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Publications:

Advanced Wound Care Adhesives with New Functional Properties
Broadly, for wounds to heal, a moist, clean and warm environment is required. A moist wound bed easily promotes growth factors and many cell types including epithelial cells to migrate, facilitating wound edge contraction. Thus, appropriate dressings play a significant role to create and maintain such environment. [1] Silicone adhesives are silicone elastomers which are not fully crosslinked but remain close to the gelation threshold (i.e. with a low crosslinking degree). [2, 3] Within the field of advanced wound care, silicone adhesives are currently the preferred, state-of-the-art adhesive system due to its gentle skin adhesion properties. However, due to their hydrophobic nature, current silicone adhesives for wound care face challenges when it comes to moisture handling. Here, we propose a novel, skin-friendly, industrially relevant glycerol-silicone hybrid adhesive with improved moisture handling due to the incorporation of emulsified glycerol (Figure 1). Various parameters will be taken into account in order to develop a relevant adhesive, in particular glycerol content, glycerol domain size and adhesive thickness to allow for a controlled moisture absorption.

A thermo-reversible silicone elastomer with remotely controlled self-healing
Soft thermoplastic elastomers with increased durability and reliability are in high demand for a broad spectrum of applications. Silicone elastomers are soft and durable, but they are not thermoplastic in nature, and under extreme conditions such as high voltage or large deformations, reliability may also suffer. Thus, as a solution to these shortcomings, which are typical of silicone elastomers, it is natural to propose a thermo-reversible, self-healing, and recyclable silicone-based elastomer. Stimuli-responsivity is imparted to the silicone polymer by incorporating supramolecular 2-ureido-4[1H]-pyrimidone (UPy) self-assembling motifs via free radical polymerisation. Self-healing of the novel elastomer may be triggered by both direct and indirect heating, the latter by means of incorporating Fe3O4 particles into the elastomer and subsequent exposure to an alternating magnetic field. As a consequence of temperature responsiveness and high thermal stability, the elastomer is proven recyclable, by withstanding multiple reprocessing procedures with no substantial effects on the resulting properties. The synergy of these valuable characteristics makes this novel material a smart candidate for innumerable applications where soft and reliable elastomers are sought.

General information
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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Coloplast Danmark AS
Authors: Chiaula, V. (Intern), Mazurek, P. (Intern), Nielsen, A. C. (Ekstern), Tornøe, J. (Ekstern), Skov, A. L. (Intern)
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A thermo-reversible silicone elastomer with remotely controlled self-healing
Soft thermoplastic elastomers with increased durability and reliability are in high demand for a broad spectrum of applications. Silicone elastomers are soft and durable, but they are not thermoplastic in nature, and under extreme conditions such as high voltage or large deformations, reliability may also suffer. Thus, as a solution to these shortcomings, which are typical of silicone elastomers, it is natural to propose a thermo-reversible, self-healing, and recyclable silicone-based elastomer. Stimuli-responsivity is imparted to the silicone polymer by incorporating supramolecular 2-ureido-4[1H]-pyrimidone (UPy) self-assembling motifs via free radical polymerisation. Self-healing of the novel elastomer may be triggered by both direct and indirect heating, the latter by means of incorporating Fe3O4 particles into the elastomer and subsequent exposure to an alternating magnetic field. As a consequence of temperature responsiveness and high thermal stability, the elastomer is proven recyclable, by withstanding multiple reprocessing procedures with no substantial effects on the resulting properties. The synergy of these valuable characteristics makes this novel material a smart candidate for innumerable applications where soft and reliable elastomers are sought.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Ogliani, E. (Intern), Yu, L. (Intern), Javakhishvili, I. (Intern), Skov, A. L. (Intern)
Deeper Insight into the Dielectric Breakdown of Elastomers

Dielectric elastomers find more and more uses but nevertheless the fundamentals behind the electrical breakdown of these thin and elastic films are still not fully understood and elucidated. Dielectric breakdown strength measurement is one of the most common methods to evaluate stability of polymers in an electric field. This breakdown test has been extensively used over many years and is still gaining on importance, due to an increasing demand on novel polymeric materials applied in high electric fields, such as: dielectric or transport layers, modern devices or flexible electronics[1].

There are only few theoretical models that assess the physical processes occurring during a breakdown phenomenon, for example: the hole-induced breakdown model, the electron-trapping breakdown model, the resonant-tunneling-induced breakdown model and the filamentary model [2]. All these theories consider movements of electrons from electrodes to polymer film samples. Other theory is the so-called electro-mechanical model [3]. It implies, that polymer films are not always smooth, and when an electric field is applied, the force gets bigger at the thinnest spot of the film. For that reason the film sample starts to deform and when electric strength is reached at the thinnest spot, breakdown occurs [3]. This is also referred to as electro-mechanical instability (EMI) and has been extensively studied by modelling [4]–[6].

In this work a high-speed camera is used in order to capture macroscopic processes taking place during the dielectric breakdown to verify if the time-scale and behavior of the electrical breakdown can elucidate the underlying behavior.
Enhancing the electro-mechanical properties of polydimethylsiloxane elastomers through blending with poly(dimethylsiloxane-co-methylphenylsiloxane) copolymers

In this work, improved electro-mechanical properties of silicone-based dielectric elastomers are achieved by means of adding so-called “voltage-stabilisers” prepared from phenyl-functional copolymers prepared using oxyanionic ring-opening polymerisation of octamethylcyclotetrasiloxane (D4) and either tetramethyltetraphenylcyclotetrasiloxane (T4) or octaphenylcyclotetrasiloxane (O4). The concentration of the voltage stabiliser was varied both by changing the molar ratio between methyl and phenyl groups in the copolymer and also by varying the amount of copolymer mixed into a PDMS-based elastomer. The phenyl-functional copolymers were generally found to disperse homogeneously in the PDMS matrix and this resulted in networks with improved mechanical and electrical properties. The developed elastomers were inherently extensible with enhanced tensile and tear strengths, due to phenyl-rich microphases acting as reinforcing domains. Furthermore, addition of phenyl-functional copolymers resulted in elastomers with increased relative permittivity and electrical breakdown strength compared to control elastomers while retaining a low dielectric loss. This demonstrates their efficiency as voltage stabilisers.
Enhancing the electro-mechanical properties of polydimethylsiloxane elastomers through blending with poly(dimethylsiloxane-co-methylphenylsiloxane) copolymers

Dielectric elastomers (DEs) hold great promise as materials for novel, advanced electromechanical applications such as actuators, generators and sensors. [1] Choosing the right polymer for the blending approach is of utmost importance to improve the electro-mechanical properties of DEs. [2] A study in our laboratories shows that sub-percentage additions of various aromatic substances can increase electrical breakdown strength significantly via voltage stabilisation, due to an electron trapping effect. [3-5]

General information
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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Technical University of Denmark
Authors: Yu, L. (Intern), Jeppe Madsen, P. (Intern), Boucher, S. (Ekstern), Skov, A. L. (Intern)
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General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Technical University of Denmark
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Electro-mechanical properties, Silicone dielectric elastomer, Voltage stabiliser, Phenyl functionalised copolymer, Blend
Enhancing the electro-mechanical properties of polydimethylsiloxane elastomers through blending with poly(dimethylsiloxane-co-methylphenylsiloxane) copolymers

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Estimation of phase separation temperatures for polyethersulfone/solvent/non-solvent systems in RTIPS and membrane properties

Phase separation temperature estimations, based on Hansen solubility parameters for poly(ethersulfone) (PES)/solvent/non-solvent systems, were carried out to study the control of phase separation temperature in a reverse thermally induced phase separation (RTIPS) process. Four membrane-forming systems were studied, namely PES/N,N-dimethylacetamide (DMAc)/diethylene glycol (DEG), PES/DMAc/polyethylene glycol 200 (PEG200), PES/DMAc/PEG300 and PES/DMAc/PEG400. The effects of PES molecular weights, PES concentrations, PEG molecular weights and ratios of non-solvent/solvent on phase separation temperature are investigated, and the theoretical Hansen solubility parameter calculation is used to establish a prediction equation for phase separation temperature. A linear relationship between the experimental data and the difference in the solubility parameters between PES and the mixed solvent was observed. When the membrane-forming temperature was higher than the cloud point, membranes with a bi-continuous structure were acquired and showed a higher pure water permeation flux than that of membranes prepared with the non-solvent induced phase separation (NIPS) process. The pure water permeation flux and the mean pore size of membranes prepared with the RTIPS process decreased in line with an increase of PES molecular weight. When the membrane formation mechanism was the RTIPS process, the mechanical properties were better than those of the corresponding membranes prepared with the NIPS process.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, Center for BioProcess Engineering, The Danish Polymer Centre, State Key Laboratory of Chemical Engineering, East China University of Science and Technology
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Glycerol-silicone elastomers as active matrices with controllable release profiles

Drug release regimes must be controlled for optimal therapeutic effect. While it is relatively straightforward to create first order release matrices, it can be challenging to avoid an initial burst. Matrices with zero-order profiles are perceived to be beneficial in many cases, but are even more difficult to formulate. We describe the straightforward synthesis of elastomeric composites prepared from silicone in which the active is dispersed in glycerol. The release of glycerol-soluble actives from films of these materials was shown to be tunable with respect to the order of release (zero- or first-order) simply by changing glycerol content. Importantly, release from the elastomers showed no burst effect. The discrete glycerol domains...
embedded within a silicone matrix act as reservoirs for active substances. Upon contact with aqueous media the active substances are released from matrices exhibiting zero-order, near zero-order or first-order release kinetics. Various parameters that could influence the release process include glycerol content, glycerol domain size or membrane thickness are thoroughly investigated, elucidating guidelines for creating matrices capable of delivering active substances at desired rates. Additionally, the composites proved to absorb significant amounts of liquid water (up to 1850 % of sample mass), a feature that can be tuned by manipulation of the composite structure.

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State: Accepted/In press
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Glycerol-silicone elastomers as active membranes for wound dressings and beyond

A green and cheap silicone-based elastomer has been developed.1,2 Through the simple mixing-in of biodiesel-originating glycerol into commercially available polydimethylsiloxane (PDMS) pre-polymer, a glycerol-in-silicone emulsion was produced. This counterintuitively stable mixture became a basis for obtaining elastomeric composites with uniformly distributed glycerol droplets. Various compositions, containing from 0 to 140 parts of glycerol per 100 parts of PDMS rubber by weight, were prepared and investigated in terms of mechanical properties as well as optical and scanning electron microscopy. The materials were proven additionally to exhibit a strong affinity to water, which was investigated by simple water absorption tests. Incorporating glycerol into PDMS decreased the Young’s modulus of the composites yet the ultimate strain of the elastomer was not compromised, even in the presence of very high loadings. The conducted experiments highlight the great potential of this new type of elastomer and reveal some possible applications especially in biomedical industry where controlled and tunable drug delivery is one of the requirements. This hybrid material was also adopted to produce glycerol-silicone elastomeric foams with adjustable densities, morphologies and mechanical properties creating a new platform for drug delivery devices.

Glycerol-silicone elastomers – current status and perspectives

Glycerol and silicone pre-polymer are two virtually immiscible liquids. However when sufficiently high shear forces are applied to a mixture of both then the glycerol phase breaks down to micro-size droplets evenly distributed within the silicone pre-polymer phase thus a glycerol-in-silicone emulsion is produced. Upon cross-linking of the silicone pre-polymer free-standing silicones with incorporated glycerol droplets are obtained as shown in Figure 1.1,2 Interestingly, mechanical properties of these composites are not compromised as the glycerol loading increases.

Glycerol-silicone elastomers became a platform for creating multiple functional smart materials, e.g. drug delivery wound care membranes, silicone foams, water absorbing silicones and magnetochromic films. Here some of the most interesting examples of glycerol-silicone elastomers applications will be presented and briefly explained elucidating the great potential of this counterintuitive composition.

General information
State: Published
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Authors: Mazurek, P. (Intern), Brook, M. A. (Ekstern), Skov, A. L. (Intern)
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Glycerol-silicone elastomers became a platform for creating multiple functional smart materials, e.g. drug delivery wound care membranes, silicone foams, water absorbing silicones and magnetochromic films. Here some of the most interesting examples of glycerol-silicone elastomers applications will be presented and briefly explained elucidating the great potential of this counterintuitive composition.

General information
State: Published
**Insight into the Dielectric Breakdown of Elastomers**

Nowadays, dielectric elastomers are used in many different fields, such as: dielectric or transport layers, modern devices or flexible electronics [1]. To test dielectric elastomer stability in electric field, dielectric breakdown measurements are used. These measurements have been used over many years and still gaining on importance, however, fundamentals behind the electrical breakdown of thin and elastic films are still not fully understood and elucidated.

There are only few theoretical models that assess the physical processes occurring during a breakdown phenomenon, for example: the hole-induced breakdown model, the electron-trapping breakdown model, the resonant-tunneling-induced breakdown model and the filamentary model [2]. In all these theories, electrons movements from electrode to polymer film samples are considered. Other theory is the, so-called, electro-mechanical model, which implies that polymer films are not always smooth, and when an electric field is applied, the force gets bigger at the thinnest spot of the film, which causes the deformation of a film. Subsequently, when electric strength is reached at the thinnest spot - breakdown occurs [3]. This is also referred to electro-mechanical instability (EMI) and has been extensively studied by modelling [4]–[7].

In this work, microscopic processes taking place during the dielectric breakdown were captured using high-speed camera, to verify if the time-scale and behavior of the electrical breakdown can elucidate the underlying behavior.

**Molecular Strategies for Improved Dielectric Elastomer Electrical Breakdown Strengths**

Dielectric elastomer transducers (actuators and generators) possess great commercial potential because they allow for novel transducer designs and applications due to—amongst others—their flexibility and low weight. On the other hand, the flexibility and inherent softness of dielectric elastomers also pose restrictions on their use, since the thin elastomers may undergo destructive deformations under large loads or in large electrical fields. In order to design better dielectric elastomers, it is crucial to understand the underlying phenomena of how thin and elastic dielectric elastomer films undergo electrical breakdown. This understanding will allow for the design of dielectric elastomers with high electrical breakdown strength and thus open up the use of films in transducers at higher electrical fields and forces. Here, the study couples intrinsic electrical breakdown strengths with well-described polymer and network characteristics, namely Kuhn parameters and crosslinking density. The universality of the developed model is illustrated by comparison over a wide range of silicone-based elastomers, such as prestretched elastomers and synthesized cross-linked bottlebrush polymers, representing both filled and unfilled elastomers. This study paves a robust way for the molecular design of elastomers into high-intrinsic electrical breakdown strength dielectric elastomers.
Optimization Techniques for Improving the Performance of Silicone-Based Dielectric Elastomers

Dielectric elastomers are possible candidates for realizing products that are in high demand by society, such as soft robotics and prosthetics, tactile displays, and smart wearables. Diverse and advanced products based on dielectric elastomers are available; however, no elastomer has proven ideal for all types of products. Silicone elastomers, though, are the most promising type of elastomer when viewed from a reliability perspective, since in normal conditions they do not undergo any chemical degradation or mechanical ageing/relaxation. Within this review, different pathways for improving the electro-mechanical performance of dielectric elastomers are highlighted. Various optimization methods for improved
energy transduction are investigated and discussed, with special emphasis placed on the promise each method holds. The compositing and blending of elastomers are shown to be simple, versatile methods that can solve a number of optimization issues. More complicated methods, involving chemical modification of the silicone backbone as well as controlling the network structure for improved mechanical properties, are shown to solve yet more issues. From the analysis, it is obvious that there is not a single optimization technique that will lead to the universal optimization of dielectric elastomer films, though each method may lead to elastomers with certain features, and thus certain potentials.

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Web of Science (2015): Indexed yes
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Scopus rating (2014): SJR 0.802 SNIP 1.055 CiteScore 1.66
Web of Science (2014): Indexed yes
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Scopus rating (2012): SJR 0.778 SNIP 0.951 CiteScore 1.46
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Web of Science (2011): Indexed yes
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Scopus rating (2010): SJR 1.098 SNIP 1.151
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 1.289 SNIP 1.092
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.266 SNIP 1.175
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 1.015 SNIP 1.154
Web of Science (2007): Indexed yes
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Screen the best ionic liquids for keratin dissolution by using COSMO-RS

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Chinese Academy of Sciences
Authors: Liu, X. (Intern), Nie, Y. (Ekstern), Zhang, S. (Ekstern), Skov, A. L. (Intern)
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Screen the best ionic liquids for keratin dissolution by using COSMO-RS

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Chinese Academy of Sciences
Authors: Liu, X. (Intern), Nie, Y. (Ekstern), Zhang, S. (Ekstern), Skov, A. L. (Intern)
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Simulation of Thermal Breakdown in a Multi-layered Stack of Dielectric Elastomers
Several aging mechanism are prone to occur during operation of dielectric elastomers. Some breakdown mechanisms are somewhat instantaneous, such as electrical, electro-mechanical and thermal breakdowns, while others are slow in order of hours, such as electrical and water trees. One of the most significant aging mechanisms is thermal breakdown, which increases its frequency significantly when stacking multiple layers of dielectric elastomers. Thermal breakdown occurs due to build-up of heat within the stacked dielectric elastomer. Heat is generated mainly through Joule heating, and if the heat generated exceeds the heat loss at the surface of the stack, the temperature will increase exponentially and a thermal breakdown is likely to occur. Thermal breakdown may happen either locally or macroscopically.

The focus of our work is to obtain a better understanding of thermal breakdown in a multi-layered stack of dielectric elastomers. This we obtain by performing numerical simulations in COMSOL Multiphysics® where joule heating and deformation, due to an externally applied voltage, is combined. From the simulation results the importance of thermal breakdown has been examined, and furthermore it has been studied how various parameters affects the point of thermal breakdown. The material of interest is PDMS which is modelled using experimentally determined material parameters and using the Yeoh model as the hyperelastic material model.

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Madsen, L. R. (Intern), Hassager, O. (Intern), Skov, A. L. (Intern)
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Structure-property relationship in silicone networks
Silicone networks find use in broad range of applications in electronic, medical, automobile, aerospace and many other industrial fields. The inherent softness and strong covalent bonds between silicium and oxygen provide the silicone network with most of its favorable properties such as thermal and oxidation stability, flexibility, hydrophobicity, and low surface energy. However, silicone elastomers suffer from poor mechanical properties due to the inherent softness of the network. The mechanical properties of the silicone networks can be improved e.g. by addition of fillers or by chemical modification of the network. Nevertheless, such modifications often comes together with deterioration of some of the desirable properties mentioned above.[1]

In this work, different silicone network structures (Fig.1) and their input on the mechanical properties will be investigated with the mail goal to maintain the favorable properties of silicone elastomers while improving the mechanical properties. The different types of silicone network structures will be formed by condensation curing reaction and evaluated with respect to the mechanical properties and long-term stability.

General information
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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, CHEC Research Centre, The Hempel Foundation Coatings Science and Technology Centre (CoaST)
Thermal degradation mechanisms of silicone elastomer

Polydimethylsiloxane (PDMS) is the most extensively used polymer among silicones. It finds application in an unlimited number of fields, including use in high temperature environments. PDMS meets these demanding requirements, due to its excellent thermal stability. The thermal degradation of linear PDMS occurs by a molecular mechanism or a radical mechanism depending on the temperature and the heating rate.[1,2] Nevertheless, the thermal degradation of cross-linked PDMS networks has not been thoroughly investigated yet. In this work, the thermal degradation mechanisms and thermal degradation products of silicone elastomers are studied. Thermogravimetric analysis (TGA) performed in inert atmosphere was carried out on PDMS networks synthesized with different stoichiometric ratios (r). Extraction of the samples was exploited to remove unreacted chains, with the aim of determining to which extent the degradation is influenced by the sol fraction and the amount of dangling chains present in the network (Figure 1). Furthermore, long-term isothermal TGA measurements were performed to recover the degradation products of the thermally treated elastomers. Soluble degradation products were analysed by size exclusion chromatography (SEC), while TGA coupled with FTIR was used to detect the released volatile degradation products.

Thermo-reversible silicone elastomer with remotely controlled self-healing

Soft thermoplastic elastomers with increased durability are in high demand for a broad spectrum of applications. Silicone elastomers are soft and durable, but they are not thermoplastic in nature and, under extreme conditions, reliability may also suffer. Thus, as a solution to these shortcomings, we report the synthesis of a thermo-reversible, self-healing and recyclable silicone elastomer, namely P(PDMSMA-co-UPyMA)[1]. Stimuli-responsivity is imparted to the silicone copolymer by incorporating supramolecular 2-ureido-4[1H]-pyrimidone (UPy)[2] self-assembling motifs (Figure 1). Self-healing of the novel elastomer may be triggered by both direct and indirect heating[3], the latter by means of incorporating Fe3O4 particles into the elastomer and subsequent exposure to an alternating magnetic field (AMF).
copolymers possess thermoplastic properties due to the reversible nature of the UPy self-associating dimers. In addition, self-healing properties were sought by alternative approaches. In particular, induction heating represents a novel method to externally trigger healing of polymers. This method consists of the incorporation of magnetic particles in the matrix and the exposure of the composite to an alternating magnetic field (AMF). As a consequence, the heat developed by the process activates network rearrangement. Controlled self-healing by use of an AMF generates heat rapidly and locally, and, above all, it is contactless.

In this work, we successfully report the development of a novel thermoplastic and self-healing silicone elastomer, namely P(PDMSMA-co-UPyMA). The remotely controlled healing of the damaged material was performed through exposure to an AMF of the composite elastomer with 20wt% Fe3O4 particle filler. Self-healing efficiency was determined by percentage of restored tensile stress and tensile strain of the healed sample compared to the native material. Scanning electron microscopy (SEM) was exploited to evaluate the morphology of samples at the healed interface. Moreover, the cross-linked copolymer was proven remouldable multiple times without showing considerable physical degradation. Hence, this material may be considered as an excellent candidate for recyclable silicone elastomers. In addition, the described self-healing approach was applied using the same conditions to another previously reported copolymer P(MEA-co-UPyMA). P(PDMSMA-co-UPyMA) and P(MEA-co-UPyMA) bear identical self-complementary motifs, although differing in the main repeating unit. Comparison of their self-healing performance under unchanged conditions aimed at assessing the versatility of the described self-healing method.

**General information**

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- **Organisations:** Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
- **Authors:** Ogliani, E. (Intern), Yu, L. (Intern), Javakhishvili, I. (Intern), Skov, A. L. (Intern)
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**Weibull Analysis of Electrical Breakdown Strength as an Effective Means of Evaluating Elastomer Thin Film Quality**

To realize the commercial potential of dielectric elastomers, reliable, large-scale film production is required. Ensuring proper mixing and subsequently avoiding demixing after, for example, pumping and coating of elastomer premix in an online process is not facile. Weibull analysis of the electrical breakdown strength of dielectric elastomer films is shown to be an effective means of evaluating the film quality. The analysis is shown to be capable of distinguishing between proper and improper mixing schemes where similar analysis of ultimate mechanical properties fails to distinguish.

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- **Organisations:** Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Technical University of Denmark
- **Authors:** Silau, H. (Intern), Stabell, N. B. (Intern), Petersen, F. R. (Ekstern), Pham, M. (Ekstern), Yu, L. (Intern), Skov, A. L. (Intern)
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Degradation patterns of silicone-based dielectric elastomers in electrical fields
Silicone elastomers have been heavily investigated as candidates for the flexible insulator material in dielectric elastomer transducers and are as such almost ideal candidates because of their inherent softness and compliance. However, silicone elastomers suffer from low dielectric permittivity. This shortcoming has been attempted optimized through different approaches during recent years. Material optimization with the sole purpose of increasing the dielectric permittivity may lead to the introduction of problematic phenomena such as premature electrical breakdown due to high leakage currents of the thin elastomer film. Within this work, electrical breakdown phenomena of various types of permittivity-enhanced silicone elastomers are investigated. Results showed that different types of polymer backbone chemistries lead to differences in electrical breakdown patterns, which were revealed through SEM imaging. This may pave the way towards a better understanding of electrical breakdown mechanisms of dielectric elastomers and potentially lead to materials with increased electrical breakdown strengths.
Silicone elastomers, Dielectric, Electrical breakdown, Voltage stabilization, Characterization

Electrical breakdown phenomena of dielectric elastomers

Silicone elastomers have been heavily investigated as candidates for dielectric elastomers and are as such almost ideal candidates with their inherent softness and compliance but they suffer from low dielectric permittivity. This shortcoming has been sought optimized by many means during recent years. However, optimization with respect to the dielectric permittivity solely may lead to other problematic phenomena such as premature electrical breakdown. In this work, we investigate the electrical breakdown phenomena of various types of permittivity-enhanced silicone elastomers. Two types of silicone elastomers are investigated and different types of breakdown are discussed. Furthermore the use of voltage stabilizers in silicone-based dielectric elastomers is investigated and discussed.
Silicone elastomers have been heavily investigated as candidates for dielectric elastomers and are as such almost ideal candidates with their inherent softness and compliance but they suffer from low dielectric permittivity.[1] This shortcoming has been sought optimized by many means during recent years. However, optimization with respect to the dielectric permittivity solely may lead to other problematic phenomena such as premature electrical breakdown. In this work, we focus on the chloro propyl functionalized silicone elastomers prepared in Madsen et al[2] and we investigate the electrical breakdown patterns of two similar chloro propyl functionalized silicone elastomers which break down electrically in a rather different way as well as we compare them to a silicone based reference. Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray Spectroscopy (EDS) are used to evaluate the elastomers after electrical breakdown.
Electrical breakdown phenomena of dielectric elastomers

Silicone elastomers have been heavily investigated as candidates for dielectric elastomers and are as such almost ideal candidates with their inherent softness and compliance but they suffer from low dielectric permittivity.[1] This shortcoming has been sought optimized by many means during recent years. However, optimization with respect to the dielectric permittivity solely may lead to other problematic phenomena such as premature electrical breakdown.

In this work, we focus on the chloro propyl functionalized silicone elastomers prepared in Madsen et al[2] and we investigate the electrical breakdown patterns of two similar chloro propyl functionalized silicone elastomers which break down electrically in a rather different way as well as we compare them to a silicone based reference. Thermogravimetric analysis (TGA) and scanning electron microscopy (SEM) are used to evaluate the elastomers before and after electrical breakdown. It was shown the chemically very similar silicone elastomers broke down electrically in very different ways. These observations emphasize that the modification of the silicone backbone may open up for completely new possibilities for stabilizing the silicone elastomer electrically. In order to tailor the elastomers, more knowledge is needed but these copolymers pave the first path towards a better understanding of the complex connection between electrical and thermal stability. Minor changes in the polymer backbone structure result in changes in electrical breakdown patterns and understanding why is crucial for enabling design for extraordinarily stable elastomers and thus ultimately reliable dielectric elastomer based products.
Helbredstjek af dansk sundhedsteknologi: Sektorudviklingsrapport
Sådan kan samarbejde mellem industrien, universiteterne og sundhedsvæsenet skabe gode løsninger til forebyggelse, diagnostik, patientbehandling og rehabilitering.

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Hvornår bryder en kunstig muskel sammen? Simulering af tærmske sammenbrud i dielektriske elastomerer
Dielektriske elastomerer kan blandt andet bruges til kunstige muskler, højtalere og til at udvinde energi fra havbølger. Ved brug af dielektriske elastomerer genereres varme grundet den elektriske modstand i materialet, hvilket kan lede til, at materialet bryder sammen. Tre essentielle parametre påvirker sammenbruddet af dielektriske elastomerer.

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Modelling of Thermal Breakdown in Dielectric Elastomers
Dielectric elastomers are a promising category of smart materials, which may find application within many fields such as soft robotics, wave-energy harvesting and loud speakers. A dielectric elastomer consists of a thin, stretchable polymer film sandwiched between two compliant electrodes. When an external voltage is applied to the electrodes, an electrostatic
pressure across the elastomer is generated, which will cause the electrodes to attract one another. Thereby the thickness of the elastomer is decreased and the cross sectional area of the elastomer is increased. When the voltage is switched off, the elastomer regenerates its original shape.

Several electrical aging mechanisms are known to occur during operation; some cause fast breakdown while others cause slow degradation of the dielectric elastomer. One of the most significant fast aging mechanisms is thermal breakdown. Thermal breakdown initiates when the heat produced within the elastomer, mainly joule heating, exceeds the heat loss to the surroundings. This may be either locally or macroscopically.

We strive to enhance the understanding of thermal breakdown in dielectric elastomer by performing numerical simulation of the actuation of dielectric elastomer transducers in stacked configuration, an example of such a simulation is shown in Figure 1. Multiple simulations using experimental data for PDMS have been performed using COMSOL Multiphysics, from which the key parameters affecting thermal breakdown have been identified. In this presentation we will present the findings and identify the optimal operating conditions for a PDMS dielectric elastomer in order to minimize thermal breakdown.

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**Modelling of Thermal Breakdown in Dielectric Elastomers**

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**Modelling of Thermal Breakdown in Dielectric Elastomers**

Dielectric elastomers are a promising category of smart materials, which may find application within many fields such as soft robotics, wave-energy harvesting and loud speakers [1]. A dielectric elastomer consists of a thin, stretchable polymer film sandwiched between two compliant electrodes. When an external voltage is applied to the electrodes, an electrostatic pressure across the elastomer is generated, which will cause the electrodes to attract one another. Thereby the thickness of the elastomer is decreased and the cross sectional area of the elastomer is increased. When the voltage is switched off, the elastomer regenerates its original shape.

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Optimisation of Silicone-based Dielectric Elastomer Transducers by Means of Block Copolymers - Synthesis and Compounding

Emerging artificial muscle technology has developed from metal-based robotics to soft-type robotics made from soft matter. Research into artificial muscle technology based on soft matter has been conducted mainly in order to mimic soft and robust human muscle. In this regard, dielectric elastomers have been studied. Their actuation occurs when Maxwell stress exceeds elastic stress in the presence of an electrical field, resulting in contraction in thickness and planar expansion in the area. As well as an actuator, dielectric elastomers can be used as generators and sensors. As a dielectric elastomer, silicones have been used extensively in many applications, due to favourable properties such as thermal stability, non-conductivity, high gas permeability and low toxicity. However, silicones have a low dielectric constant and thereby low energy density. In order to enhance actuation performance, it is the aim of this research to develop silicone elastomers with a high dielectric constant and high electrical breakdown strength, as well as a low Young's modulus.

In this Ph.D. thesis, two methods were developed to enhance silicone properties such as the dielectric constant and electrical breakdown strength. The first method was devised to enhance the dielectric constant of silicone elastomers through the use of a polydimethylsiloxane-polyethylene glycol (PDMS-PEG) copolymer, in order to obtain an elastomer
with high electrical energy. PDMS-PEG copolymers were synthesised and blended in commercial silicone and subsequently cross-linked. The relative permittivity of cross-linked silicone with 5 wt% of PDMS-PEG copolymers increased by nearly 50%, without compromising dielectric loss and mechanical properties, compared to the commercial silicone elastomer.

The second investigated method involved enhancing the electrical breakdown strength of silicone by using an aromatic voltage stabiliser. Here, polyphenylmethylsiloxane (PPMS), which contained aromatic voltage stabilisers, was bonded covalently to PDMS through a hydrosilylation reaction obtaining PDMS-PPMS copolymers. The synthesised copolymers were subsequently cross-linked with a vinyl cross-linker. The obtained cross-linked PDMS-PPMS copolymers were inherently soft and robust with increased electrical breakdown strength (21%) compared to the reference elastomer without an aromatic voltage stabiliser.

The conducting polymer was developed through the use of a multi-walled carbon nanotube (MWCNT) in a PDMS-PEG matrix as a compliant electrode of dielectric elastomers. The conductive PDMS-PEG copolymer was incorporated with surface-treated MWCNT, in order to obtain highly conductive elastomer. The prepared sample with 4 parts per hundred rubber (phr) MWCNT was soft and the resulting conductivity of the cross-linked PDMS-PEG copolymer with the addition of MWCNT was high, at 10-2 S cm-1, nearly equivalent to a commonly used commercial conducting polymer.

In this thesis, the elastomer and electrode system is referred to as a ‘dielectric elastomer transducer.’

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Post Curing as an Effective Means of Ensuring the Long-term Reliability of PDMS Thin Films for Dielectric Elastomer Applications
Post curing can be used to facilitate volatile removal and thus produce polydimethylsiloxane (PDMS) films with stable elastic and electrical properties over time. In this study, the effect of post curing was investigated for commercial silicone elastomer thin films as a means of improving long-term elastomer film reliability. The Young’s moduli and electrical breakdown strengths of commercial (silica-reinforced) PDMS elastomer films, with and without additional 35 parts per hundred rubber titanium dioxide (TiO2), were investigated after high-temperature (200°C) post curing for various time spans. The elastomers were found to contain less than 2% of volatiles (significantly higher for TiO2-filled samples), but nevertheless a strong effect from post curing was observed. The young’s moduli as well as the strain-dependent behavior were found to change significantly upon post curing treatment, where Young’s moduli at 5% strain increase with post curing. Furthermore, the determined dielectric breakdown parameters from Weibull analyses showed that greater electrical stability and reliability could be achieved by post curing the PDMS films before usage, and this method therefore paves a way toward more reliable dielectric elastomers.

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Silicone-based Dielectric Elastomers

Efficient conversion of energy from one form to another (transduction) is an important topic in our daily life, and it is a necessity in moving away from the fossil based society. Dielectric elastomers hold great promise as soft transducers, since they are compliant and light-weight amongst many favorable properties. Their transduction principle lies in direct conversion of electrical energy into mechanical energy or vice versa with no need for gearing and with no stand-by energy consumption. This should in principle support a more efficient use of energy, a hot topic in our current society where energy efficient solutions are highly sought. These properties allow for interesting products ranging very broadly, e.g. from eye implants over artificial skins over soft robotics to huge wave energy harvesting plants. All these products utilize the inherent softness and compliance of the dielectric elastomer transducers.
The subject of this thesis is improvement of properties of silicone-based dielectric elastomers with special focus on design guides towards electrically, mechanically, and electromechanically reliable elastomers. Strategies for improving dielectric elastomer performance are widely investigated but rarely discussed in the context of mechanical integrity and thus product reliability. Focus here is on long-term reliability of the dielectric elastomers and how to achieve this by means of careful elastomer design. This thesis presents methods and results of analyses acquired in the cross-disciplinary, collaborative effort on dielectric elastomers funded by Innovationsfonden Denmark (formerly Advanced Technology Foundation) with the materials workgroup headed by the author. Main contributors to the work have been research scientists at Danfoss PolyPower, colleagues from the Danish Polymer Centre, as well as 7 PhD students and 5 postdocs being involved in the project. International collaborators were also part of the project at various stages. The studies behind this thesis have been conducted over a period of about 5 years, and 10 selected papers describing the main results are included as appendices. They were chosen to represent the prime results obtained within the project. Most of the technical aspects discussed in this thesis are contained within these references. Several other important works have been omitted in the appendices in order to keep the thesis relatively short and concise.

Throughout the thesis the articles within the appendices are referred to as A1-A10. For all of these articles, the author was the principal investigator. Other dielectric-elastomer related papers by the author - either as a principal investigator or co-investigator- are referred to as A11-A51. Chapters 1 to 5 of the thesis present a coherent summary of the included papers in a common context, emphasizing the overall purpose and flow of the analysis thematically. Unpublished work is included as well to facilitate coupling between approaches as well as to provide the full, comprehensive story. Chapter 6 gives a conclusion, and in Chapter 7 a personal perspective on the future of dielectric elastomer research is given.

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### Silicone elastomers with covalently incorporated aromatic voltage stabilisers
When optimising dielectric elastomers (DEs) a conflict exists, namely that for large achievable actuation strains softness is required, but with increased softness electrical breakdown strength decreases. Herein, soft dielectric silicone elastomers with increased electrical breakdown strength, due to the incorporation of an aromatic voltage stabiliser, were prepared by cross-linking synthesised polydimethylsiloxane–polyphenylmethylsiloxane (PDMS–PPMS) copolymers. PPMS possesses voltage stabilisation capabilities but is immiscible in PDMS, and thus the copolymerisation of the two components was necessary for homogeneity. Concentrations of the voltage stabiliser were varied by changing the molecular weights of the PPMS in the copolymer. The developed elastomers were inherently soft with enhanced electrical breakdown strengths, due to delocalised electrons of the aromatic constituent. An optimum concentration was found for the voltage stabilisation effect. The relative permittivities of the PDMS–PPMS elastomers varied from 3.4 to 3.9 and therefore were also improved from pure PDMS elastomers. The elastomers were furthermore non-conductive and possessed low dielectric losses. These properties are evaluated as favourable for soft actuation.

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Soft and flexible conductive PDMS/MWCNT composites

Conductive elastomers based on MWCNT in polydimethylsiloxane (PDMS) have been prepared by a range of dispersion methods such as ultrasonication, speedmixing and roll milling in combination with physical or covalent modification. The ionic liquid (IL), 1-ethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide, was used to pre-disperse MWCNT in a MWCNT/IL-gel that was used for preparation of MWCVNT/PDMS composites. The method was seen to be effective at low levels of MWCNT, but required combination with a roll mill to obtain a stable dispersion at 4 wt % MWCNT. With higher amounts of MWCNT a reduction in conductivity was observed, which was attributed to a change in morphology occurring between 4 and 5 wt % MWCNT. As an alternative to IL dispersing aids a novel functionalized MWCNT was prepared by free radical polymerization using α-methacryloxypropyl-polydimethylsiloxane, which could be used directly for preparation of MWCNT/PDMS composites. Composites prepared by use of the IL dispersion method, use of a roll mill or by use of the f-MWCNT all had conductivities around 0.005–0.01 s/cm and retained conductivity upon extension.

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The mechanism of fracture for entangled polymer liquids in extensional flow

In uniaxial extensional flow of entangled polymer liquids, different rupture modes may happen, including necking and fracture. Malkin and Petrie [1] proposed a “master curve” dividing the flow behavior into four zones based on the stretch rate: (I) Flow zone; (II) Transition zone; (III) Rubbery zone; and (IV) Glass-like zone. The master curve shows that steady extensional flow can only be reached in Zone I where the stretch rate is very slow, while rupture happens in Zones II-IV with faster stretch rate. Furthermore, Wang et al. [2-4] reported experimental data that matches the master curve and suggested the mechanism of rupture in Zone III and IV is disentanglement and chain scission, respectively. In this work we measure two groups of entangled polystyrene solutions. In one group the samples have the same entanglement molecular weight (Me) but different number of entanglements (Z), and in the other group the samples have the same Z but different Me. We show that uncontrolled filament stretching, steady extensional flow can be reached in Zones I-III, while fracture happens in Zone IV. The critical strain at fracture decreases with increasing stretch rate, which is in agreement with the master curve in Zone IV. However, with faster rate, a constant critical strain is observed, which is not shown in the original master curve. The value of the constant critical strain seems to be related to the maximum stretch ratio of the polymer chain (determined by Me), but not influenced by Z. The results are also compared with the critical strain of chemically crosslinked polymer networks.

Voltage-stabilised elastomers with increased relative permittivity and high electrical breakdown strength by means of phase separating binary copolymer blends of silicone elastomers

Increased electrical breakdown strength and increased dielectric permittivity of silicone-based dielectric elastomers are achieved by means of the addition of so-called voltage-stabilisers prepared from PDMS–PPMS copolymers as well as PDMS–PEG copolymers in order to compensate for the negative effect of softness on electrical stability of silicone elastomers. The voltage-stabilised elastomer, incorporating a high-permittivity PDMS–PEG copolymer, possesses increased relative permittivity, high electrical breakdown strength, excellent network integrity and low dielectric loss and paves the way towards specialised silicone elastomers for dielectric elastomer transducer products with inherent softness and electrical stability, and thus increased actuation at a given voltage.
ZnO as a cheap and effective filler for high breakdown strength elastomers

Cheap, high-performance dielectric elastomers are in high demand from industry concerning new products based on dielectric elastomer transducers. However, formulating an elastomer that fulfils all the requirements for dielectric elastomers is difficult and, first and foremost, not cheap. In this article, we explore the use of a cheap and abundant metal oxide filler, namely ZnO, as a filler in silicone-based dielectric elastomers. The electro-mechanical properties of the elastomer composites are investigated, and their performance is evaluated by means of figures of merit. Various commercial silicone elastomers and a self-formulated silicone elastomer are utilised as elastomer matrices, the effects of which on the final properties of the elastomer composite are investigated.
A multiple length scale description of the mechanism of elastomer stretching

Conventionally, the stretching of rubber is modeled exclusively by rotations of segments of the embedded polymer chains; i.e. changes in entropy. However models have not been tested on all relevant length scales due to a lack of appropriate
probes. Here we present a universal X-ray based method for providing data on the structure of rubbers in the 2-50 angstrom range. First results relate to the elongation of a silicone rubber. We identify several non-entropic contributions to the free energy and describe the associated structural changes. By far the largest contribution comes from structural changes within the individual monomers, but among the contributions is also an elastic strain, acting between chains, which is 3-4 orders of magnitude smaller than the macroscopic strain, and of the opposite sign, i.e. extension of polymer chains in the direction perpendicular to the stretch. This may be due to trapped entanglements relaxing to positions close to the covalent crosslinks.

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Authors: Neuefeind, J. (Ekstern), Skov, A. L. (Intern), Daniels, J. E. (Ekstern), Honkimaki, V. (Ekstern), Jakobsen, B. (Ekstern), Oddershede, J. (Intern), Poulsen, H. F. (Intern)
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A simple method for reducing inevitable dielectric loss in high-permittivity dielectric elastomers
Commercial viability of dielectric elastomers (DEs) is currently limited by a few obstacles, including high driving voltages (in the kV range). Driving voltage can be lowered by either decreasing the Young's modulus or increasing the dielectric
permittivity of silicone elastomers, or a combination thereof. A decrease in the Young's modulus, however, is often accompanied by a loss in mechanical stability, whereas increases in dielectric permittivity are usually followed by a large increase in dielectric loss followed by a decrease in breakdown strength and thereby the lifetime of the DE. A new soft elastomer matrix, with high dielectric permittivity and a low Young's modulus, aligned with no loss of mechanical stability, was prepared through the use of commercially available chloropropyl-functional silicone oil mixed into a tough commercial liquid silicone rubber silicone elastomer. The addition of chloropropyl-functional silicone oil in concentrations up to 30 phr was found to improve the properties of the silicone elastomer significantly, as dielectric permittivity increased to 4.4, dielectric breakdown increased up to 25% and dielectric losses were reduced. The chloropropyl-functional silicone oil also decreased the dielectric losses of an elastomer containing dielectric permittivity-enhancing TiO2 fillers. Commercially available chloropropyl-functional silicone oil thus constitutes a facile method for improved silicone DEs, with very low dielectric losses.

**General information**

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Bimodal condensation silicone elastomers as dielectric elastomers

Lately, dielectric elastomers (DEs) which consist of an elastomer sandwiched between electrodes on both sides, have gained interest as materials for actuators, generators, and sensors. An ideal elastomer for DE uses is characterized by high extensibility, flexibility and good mechanical fatigue as well as high electrical and mechanical breakdown strengths.

[1] Most model elastomers are prepared by an end-linking process using a crosslinker with a certain functionality \( f \) and a linear polymer with functional groups in both ends, and the resulting networks are so-called unimodal networks where unimodal refers to that there is one polymer only in the system. As an alternative to unimodal networks there are the bimodal networks where two polymers with significantly different molecular weights are mixed with one crosslinker.

[2] Silicone rubber can be divided into condensation type and addition type according to the curing reaction. The advantages of condensation silicones compared to addition are the relatively low cost, the curing rate largely being independent of temperature, the excellent adhesion, and the catalyst being nontoxic. [3] In this work, a series of bimodal condensation silicone elastomers were prepared by mixing different mass ratios (9:1, 8:2, 7:3, 6:4, 5:5, 4:6) between long polydimethylsiloxane (PDMS) chains and short PDMS chains. The resulting elastomers were investigated with respect to their rheology, dielectric properties, tensile strength, electrical breakdown, as well as thermal stability. The bimodal elastomers reinforce themselves at large strain and the high electrical breakdown strength is obtained due both to the low extensibility of the short chains that attach strongly the long chains and to the extensibility of the last ones that retards the rupture process. Moreover, a series of elastomers with the same mass ratio (7:3) between long and short PDMS chains were made at different humidity (90%, 70%, 50%, 30%, 10%) at 23°C. The dielectric and mechanical properties of the resulting elastomers were shown to depend strongly on the atmospheric humidity level. In addition, the top and bottom surfaces of the elastomer (7:3) prepared at 23°C and 50% humidity were tested by water contact angle and optical microscope. The results show the bimodal condensation elastomer possesses structural heterogeneity, which may lead to favourable properties for DE applications.
Bimodal condensation silicone elastomers as dielectric elastomers

Lately, dielectric elastomers (DEs) which consist of an elastomer sandwiched between electrodes on both sides, have gained interest as materials for actuators, generators, and sensors. An ideal elastomer for DE uses is characterized by high extensibility, flexibility and good mechanical fatigue as well as high electrical and mechanical breakdown strengths. [1] Most model elastomers are prepared by an end-linking process using a crosslinker with a certain functionality f and a linear polymer with functional groups in both ends, and the resulting networks are so-called unimodal networks where unimodal refers to that there is one polymer only in the system. As an alternative to unimodal networks there are the bimodal networks where two polymers with significantly different molecular weights are mixed with one crosslinker. [2]Silicone rubber can be divided into condensation type and addition type according to the curing reaction. The advantages of condensation silicones compared to addition are the relatively low cost, the curing rate largely being independent of temperature, the excellent adhesion, and the catalyst being nontoxic. [3]In this work, a series of bimodal condensation silicone elastomers were prepared by mixing different mass ratios between long polydimethylsiloxane (PDMS) chains and short PDMS chains. The resulting elastomers were investigated with respect to their rheology, dielectric properties, tensile strength, electrical breakdown, thermal stability, as well as hydrophobicity and morphology.

Dielectric Elastomers (DEs) as EAPs: Materials

Dielectric elastomers (DEs) which consist of an elastomer sandwiched between electrodes on both sides, have gained interest as materials for actuators, generators, and sensors. An ideal elastomer for DE uses is characterized by high extensibility, flexibility and good mechanical fatigue as well as high electrical and mechanical breakdown strengths. [1] Most model elastomers are prepared by an end-linking process using a crosslinker with a certain functionality f and a linear polymer with functional groups in both ends, and the resulting networks are so-called unimodal networks where unimodal refers to that there is one polymer only in the system. As an alternative to unimodal networks there are the bimodal networks where two polymers with significantly different molecular weights are mixed with one crosslinker. [2]Silicone rubber can be divided into condensation type and addition type according to the curing reaction. The advantages of condensation silicones compared to addition are the relatively low cost, the curing rate largely being independent of temperature, the excellent adhesion, and the catalyst being nontoxic. [3]In this work, a series of bimodal condensation silicone elastomers were prepared by mixing different mass ratios between long polydimethylsiloxane (PDMS) chains and short PDMS chains. The resulting elastomers were investigated with respect to their rheology, dielectric properties, tensile strength, electrical breakdown, thermal stability, as well as hydrophobicity and morphology.
Electrical Breakdown and Mechanical Ageing in Dielectric Elastomers

Dielectric elastomers (DE) are used in various applications, such as artificial eye lids, pressure sensors and human motion energy generators. For many applications, one of the major factors that limits the DE performance is premature electrical breakdown. There are many approaches that have been reported to increase the breakdown strength of DEs such as compositing and pre-stretching. Some of the techniques, however, affect other parameters related to DEs negatively. For instance, the elastomers with hard filler particles (e.g. metal oxides) used as DEs experience difficulties to maintain their long-term mechanical reliability as they are susceptible to Mullins effects as the results of pre-stretching. Therefore, two strategies are developed in this thesis in order to produce DEs with high electrical performance and long-term electromechanical reliability. The first strategy is to study the mechanisms behind the electrical breakdown of DEs and the second strategy is to investigate the long-term electromechanical reliability of DEs. In the first strategy, the electrothermal breakdown in polydimethylsiloxane (PDMS) elastomers was modelled in order to evaluate the thermal mechanisms behind the electrical failures. From the modelling based on the fitting of experimental data, it showed that the electrothermal breakdown of the PDMS elastomers was strongly influenced by the increase in both relative permittivity and conductivity. In addition to that, a methodology in determining the parameters that affect the breakdown strength of the pre-stretched DEs was developed. Breakdown strength was determined for samples with and without volume conservation and was found to depend strongly on the strain and the thickness of the samples.

In order for DEs to be fully implementable in commercial products, the lifetime of elastomer materials needs further investigation. Therefore, in the second strategy, several DE parameters such as Young’s moduli, breakdown strengths and dielectric permittivities of PDMS elastomers filled with hard filler particles were investigated after being subjected to pre-stretching for various timespans. The study showed that electromechanical reliability when pre-stretching was difficult to achieve with PDMS elastomers filled with hard filler particles. Subsequently, the long-term mechanical and electrical reliability was further investigated to the PDMS elastomers filled with the soft fillers (e.g. oils). Interestingly, the results also showed that soft fillers significantly influence the long-term electromechanical reliability of PDMS elastomers. However, despite the pre-stretched PDMS elastomers filled with hard and soft filler experience difficulties to maintain their long-term electromechanical reliability, the study paves the way for electromechanically reliable DEs by indicating that simply post-curing PDMS elastomers before use.

Therefore in the last part of this thesis, the effect of post-curing was investigated for PDMS elastomer thin-films as a means of improving the long-term elastomer film electromechanical reliability. The PDMS elastomers were found to contain less than 2% of volatiles but nevertheless a strong effect from post-curing was observed. Furthermore, the determined electrical breakdown parameters from Weibull analyses showed that greater electrical reliability could be achieved by post-curing the PDMS elastomers before usage, and this method therefore paves a way towards more electromechanically reliable DEs.

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Entangled Polymer Melts in Extensional Flow
Many commercial materials derived from synthetic polymers exhibit a complex response under different processing operations such as fiber formation, injection moulding, film blowing, film casting or coatings. They can be processed both in the solid or in the melted state. Often they may contain two or more different polymers in addition to additives, fillers or solvents in order to modify the properties of the final product. Usually, it is also desired to improve the processability. For
example the supplement of a high molecular weight component improves the stability in elongational flows. On the other hand, addition of low-volatility solvents to polymers is also a common industrial practice that others a means for lowering the $T_g$ of the polymers. Moreover industrial polymers present a wide distribution of chain lengths and/or branched architectures that strongly influence their response.

Understanding the behaviour of polymer melts and solutions in complex non-linear flows is crucial for the design of polymeric materials and polymer processes. Through rheological characterization, in shear and extensional flow, of model polymer systems, i.e. narrow molar mass distribution polymer melts and solutions or well defined polymer molecules architecture, researchers develop constitutive equations that can relate the stress induced into a material with its flow deformation history. Indeed experiments on samples with well-defined structure supply data that can be compared with models.

Current models have been shown to be quite successful in describing the dynamics of polymers although they are still continuously challenged by new experimental data on model polymer systems. At the same time, new methods for generating extensional flows [McKinley and Sridhar (2002), Sentmanat (2004), Bach et al. (2003b)] are being constantly refined to improve the quality of the data and to explore a wider range of rates and deformations. Moreover, recently rheometry methods have been supplemented by other techniques such as dielectric spectroscopy that can probe chain dynamics and neutron scattering which can monitor macromolecular chain orientation associated with induced flow fields. This work concerns linear and non-linear rheology of polystyrene melts and solutions coupled with neutron scattering experiments. The aim of this thesis is to investigate the extensive properties of well characterized polymer samples and provide new experimental data on extensional rheology that can validate constitutive models. Moreover we show how the extensional technique may be used in combination with small-angle neutron scattering (SANS) to perform single chain structural studies after uniaxial elongation both after steady extensional flow and at several times during true stress relaxation. Extensional experiments have been performed on a Filament Stretching Rheometer (FSR), placed at the Technical University of Denmark (DTU), equipped with an online controlled scheme that allows to operate in controlled strain rate or controlled stress mode. High temperatures measurements can be performed due to an oven that surrounds the sample environment. Also a new implemented version of the device, named VADER 1000, has been employed to prepare the neutron scattering samples. The reduced dimension, compared to the FSR, and the particular design of the oven meets the requirement of fast cooling of the sample, so that it can freeze the particular macromolecular orientation of the chains at different stages of the stretching or relaxing of the sample.

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Department of Micro- and Nanotechnology, Amphiphilic Polymers in Biological Sensing
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**Glycerol as high-permittivity liquid filler in dielectric silicone elastomers**

A recently reported novel class of elastomers was tested with respect to its dielectric properties. The new elastomer materials based on a commercially available poly(dimethylsiloxane) composition, which has been modified by embedding glycerol droplets into its matrix. The approach has two major advantages that make the material useful in a dielectric actuator. First, the glycerol droplets efficiently enhance the dielectric constant, which can reach astonishingly high values in the composite. Second, the liquid filler also acts as a softener that effectively decreases the elastic modulus of the composite. In combination with very low cost and easy preparation, the two property enhancements lead to an extremely attractive dielectric elastomer material. Experimental permittivity data are compared to various theoretical models that predict relative permittivity changes as a function of filler loading, and the applicability of the models is discussed. VC 2016 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 2016, 133, 44153.

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Glycerol as high-permittivity liquid filler in dielectric silicone elastomers

A recently reported novel class of elastomers was tested with respect to its dielectric properties. The new elastomer material is based on a commercially available polydimethylsiloxane (PDMS) composition, which has been modified by embedding glycerol droplets into its matrix. The approach has two major advantages that make the material useful in a dielectric actuator. First, the glycerol droplets efficiently enhance the dielectric constant which can reach very high values in the composite. Second, the liquid filler also acts as a softener that effectively decreases the elastic modulus of the composite. In combination with very low cost and easy preparation, the two property enhancements lead to a very attractive dielectric elastomer material. Experimental permittivity data are compared to various theoretical models that predict relative-permittivity changes as a function of filler loading, and the applicability of the models is discussed.

General information
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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, University of Potsdam
Authors: Mazurek, P. S. (Intern), Yu, L. (Intern), Skov, A. L. (Intern), Gerhard, R. (Ekstern), Wirges, W. (Ekstern)
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Publication: Research - peer-review › Poster – Annual report year: 2017

Green silicone elastomer obtained from a counterintuitively stable mixture of glycerol and PDMS
A green and cheap silicone-based elastomer has been developed. Through the simple mixing-in of biodiesel-originating glycerol into commercially available polydimethylsiloxane (PDMS) pre-polymer, a glycerol-in-PDMS emulsion was produced. This counterintuitively stable mixture became a basis for obtaining elastomeric composites with uniformly distributed glycerol droplets. Various compositions, containing from 0 to 140 parts of glycerol per 100 parts of PDMS by weight, were prepared and investigated in terms of ATR-FTIR, broadband dielectric spectroscopy, mechanical properties as well as optical and scanning electron microscopy. The materials were proven additionally to exhibit a strong affinity to water, which was investigated by simple water absorption tests. Incorporating glycerol into PDMS decreased the Young's modulus of the composites yet the ultimate strain of the elastomer was not compromised, even in the presence of very high loadings. The conducted experiments highlight the great potential of this new type of elastomer and reveal some possible applications.

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- Web of Science (2016): Indexed yes  
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  - Scopus rating (2015): SJR 1.144 SNIP 1.277 CiteScore 3.72  
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- BFI (2012): BFI-level 1  
  - Scopus rating (2012): SJR 1.589 SNIP 1.777 CiteScore 3.74  
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- BFI (2011): BFI-level 1  
  - Scopus rating (2011): SJR 1.623 SNIP 1.797 CiteScore 4.04  
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- Web of Science (2011): Indexed yes  
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- Web of Science (2008): Indexed yes  
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- Web of Science (2007): Indexed yes  
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- Web of Science (2005): Indexed yes  
- Scopus rating (2004): SJR 1.747 SNIP 1.724  
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- Web of Science (2003): Indexed yes  
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- Web of Science (2001): Indexed yes  
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Interpenetrated polymer networks based on commercial silicone elastomers and ionic networks with high dielectric permittivity and self-healing properties

The dielectric elastomers (DEs) technology can be used in many advanced applications, such as actuators, generators and sensors, showing advantageous and promising properties[1]. However, the main disadvantage is the high driving voltage required for the actuation process which limits the applicability. One method used to avoid this limitation is to increase the dielectric permittivity of the material in order to improve the actuation response at a given field. Recently, interpenetrating polymer networks (IPNs) based on covalently cross-linked commercial silicone elastomers and ionic networks from amino- and carboxylic acid- functional silicones have been designed[2] (Figure 1). This novel system provides both the mechanical stability and the high breakdown strength given by the silicone part of the IPNs and the high permittivity and the softening effect of the ionic network. Thus these improved properties are achieved without consequently increased Young’s moduli and decreased breakdown strength compared, for example, with other silicone elastomers containing fillers. In particular, the interpenetrating systems show dielectric permittivity ε’ from 6,7 to 2 x 103 at low frequencies (0,1 Hz), and the commercial elastomers RT625 and LR3043/30 provide the best viscoelastic properties to the systems, since they maintain low viscous losses upon addition of ionic network. The values of the breakdown strength in all cases remain higher than that of the reference pure PDMS network (ranging from 45 V/μm to 90V/μm)[3]. In addition, the ionic part of the interpenetrating systems, based on non-covalent interactions, provides promising selfhealing properties both upon mechanical rupture and upon electrical breakdown. The systems are capable of recovering and to support more than 100% elongation of the reassembled samples (Figure 2). This additional advantage achieved by the IPNs represents an encouraging step forward in the challenge of increasing the life-time of the DEs.

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Interpenetrated polymer networks based on commercial silicone elastomers and ionic networks with high dielectric permittivity and self-healing properties

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Interpenetrating polymer networks based on commercial silicone elastomers and ionic networks with high dielectric permittivity and self-healing properties

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Linear Viscoelastic and Dielectric Relaxation Response of Unentangled UPy-Based Supramolecular Networks
Supramolecular polymers possess versatile mechanical properties and a unique ability to respond to external stimuli. Understanding the rich dynamics of such associative polymers is essential for tailoring user-defined properties in many products. Linear copolymers of 2-methoxyethyl acrylate (MEA) and varying amounts of 2-ureido-4[1H]-pyrimidone (UPy) quadruple hydrogen-bonding side units were synthesized via free radical polymerization. Their linear viscoelastic response was studied via small amplitude oscillatory shear (SAOS). The measured linear viscoelastic envelope (LVE) resembles that of a well-entangled polymer melt with a distinct plateau modulus. Dielectric relaxation spectroscopy (DRS) was employed to independently examine the lifetime of hydrogen bond units. DRS reveals a high frequency α-relaxation associated with the dynamic glass transition, followed by a slower α*-relaxation attributed to the reversible UPy hydrogen bonds. This time scale is referred to as the bare lifetime of hydrogen bonding units. Using the sticky Rouse model and a renormalized lifetime, we predict satisfactorily the LVE response for varying amounts of UPy side groups. The deviation from the sticky Rouse prediction is attributed to polydispersity in the distribution of UPy groups along the chain backbone. We conclude that the response of associating polymers in linear viscoelasticity is general and does not depend on the chemistry of association, but rather on the polymer molecular weight (MW) and MW distribution, the number of stickers per chain, \( n_s \), and the distribution of stickers along the backbone.

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Centro de Física de Materiales, Drexel University
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Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 2.357 SNIP 1.599 CiteScore 5.82
Web of Science (2015): Indexed yes
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Scopus rating (2014): SJR 2.524 SNIP 1.695 CiteScore 5.83
Matematisk beskrivelse af dielektrisk styrke

Dielektriske elastomerer (DE) er en relativ ny type materiale, som har stort potentiale blandt andet inden for medico- og energiteknologi

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Mechanical and Electrical Ageing Effects on the Long-Term Stretching of Silicone Dielectric Elastomers with Soft Fillers:

**Long-Term Stretching of Silicone Dielectric Elastomers**

Dielectric elastomer materials for actuators need to be soft and stretchable while possessing high dielectric permittivity. Soft silicone elastomers can be obtained through the use of silicone oils, while enhanced permittivity can be obtained through the use of dipolar groups on the polymer backbone. Such elastomers are prepared by adding soft fillers to a strong and relatively stiff elastomer, Elastosil LR3043/50. The long-term stability of the materials is tested by straining the elastomers 60% statically for up to 3 months. The results show that soft fillers significantly influence the long-term stability of silicone elastomers, with electrical breakdown strength being the most influenced. Especially high concentrations of silicone oils should be avoided for long-term mechanical stability.
Mechanically compliant electrodes and dielectric elastomers from PEG-PDMS copolymers

Soft conducting elastomers have been prepared from polydimethylsiloxane-polyethylene glycol (PDMS-PEG) copolymer and surfactant-stabilized multi-walled carbon nanotubes (MWCNTs). The copolymer was chain-extended with PDMS of molecular weight 17.2 kg mol⁻¹ in order to obtain a crosslinkable PDMS with molecular weight around 20 – 30 kg mol⁻¹. MWCNTs were treated with surfactant and sonicated for better dispersion in the polymer matrix. The conductivity and mechanical properties of conducting elastomers were thoroughly investigated including stress and strain at break. The developed conducting elastomers showed high conductivity combined with inherent softness. The high conductivity and softness, PDMS-PEG copolymers with incorporated MWCNTs hold great promises as compliant and highly stretchable electrodes for stretchable devices such as electro-mechanical transducers.

General information

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: A Razak, A. H. (Intern), Madsen, F. B. (Intern), Skov, A. L. (Intern)
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Monolithic PDMS Laminates for Dielectric Elastomer Transducers through Open-Air Plasma Treatment

The present study investigates the use of an open-air plasma-treatment system for the surface modification of polydimethylsiloxane (PDMS), in order to improve layer-to-layer adhesion. The procedure presented herein is more cost efficient compared to conventional vacuum-based plasma-treatment, and it is performed at different speeds and distances away from the nozzle, to investigate how these two parameters influence the resulting interfacial layer of two fully cured PDMS films. The plasma-treatment is determined not to alter mechanical properties compared to the single film, while peel forces are sufficient to avoid delamination during operation.
Novel high dielectric constant hybrid elastomers based on glycerol-insilicone emulsions

Novel hybrid elastomers were prepared by speedmixing of two virtually immiscible liquids – glycerol and polydimethylsiloxane (PDMS) prepolymer. Upon crosslinking of the PDMS phase of the resulting glycerol-in-silicone emulsion freestanding films were obtained. In this way glycerol became uniformly distributed within PDMS in shape of discrete droplets thus acting as a high dielectric constant filler efficiently enhancing the dielectric constant of the composites. Low- and high-voltage dielectric spectroscopy measurements were conducted in order to verify applicability of the composites as dielectric elastomer actuators. Conductivities of samples based on various PDMS compositions with different loadings of embedded glycerol were thoroughly investigated providing useful information about the dielectric behavior.

Remotely Activated Microcapsules for Oil Recovery Treatments

Water-flooding is a commonly used oil recovery method in mature reservoirs, because injected water enhances oil sweep efficiency and leads to increased oil production. Due to the occurrence of fractures in water-flooded reservoirs, excessive water production is observed. Hence, water shut-off treatments are extensively investigated, though currently applied materials still suffer from some disadvantages. The main drawback is lack of control over the setting of plugs in the fracture, and this may cause blocking of the injection well and the formation of the plug before placing the material in the fracture. Secondly, only a few developed materials prevent from blocking oil-rich pores, and yet they are not able to create a permanent plug, as they do not form a covalently bonded structure.

The aim of the project is to design a novel plugging material which can block fractures without affecting the pores and
ensure permanent water shut-off treatment. To eliminate the danger of premature plug formation, stimulus-responsive materials were investigated. Considering that fractures are extremely hard-to-access places, the designed material should respond to a remotely applied stimulus, in order to achieve better control over plug formation. The developed material consists of vinyl-functional polydimethylsiloxane (PDMS) microparticles and microcapsules with an encapsulated PDMS cross-linker. Due to reactions between the released cross-linker and vinyl groups on the PDMS microparticles’ surface, a covalently bonded elastomer plug is formed. The size of both components is adjusted to allow for creating the plug without affecting the pores. Control over cross-linking is achieved through an encapsulation process. Microcapsules prepared via a phase separation method are impermeable at 50°C, thereby eliminating premature plug formation. It is shown that after heating microcapsules to 120°C, the cross-linker is released from a poly(methyl methacrylate) (PMMA) shell. Moreover, gamma irradiation of the microcapsules decreases the Tg of the polymeric shell, changing the permeability of the microcapsules. Due to the irradiation process, microcapsules become permeable at lower temperatures, and a remotely applied activation has been achieved. Applying the alternating magnetic field (AMF) was another activation method investigated as part of this research. Magnetic nanoparticles, which generate heat on exposure to the AMF, were encapsulated successfully within the polymeric shell, and as a result, heating microcapsules when they are subjected to the AMF is possible. As an alternative to remote activation methods, solvent-flushing was examined, allowing for an investigation into the plug being cross-linked under high-pressure. Compressed material created a uniform, void-less structure capable of withstanding harsh conditions in reservoirs.

General information
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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
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Rheology of Supramolecular Polymers
Supramolecular polymers are a broad class of materials that include all polymers capable of associating via secondary interactions. These materials represent an emerging class of systems with superior versatility compared to classical polymers with applications in food stuff, coatings, cost efficient processes or biomedical areas. Design and development of supramolecular polymers using ionic, hydrogen bonding or transition metal complexes with tailored properties requires deep understanding of dynamics both in linear and non-linear deformations. While linear rheology is important to understand the dynamics under equilibrium conditions, extensional rheology is relevant during the processing or in the usage of polymers utilizing supramolecular associations for example, acrylic based pressure sensitive adhesives are subjected to extensional deformations during the peeling where strain hardening is often desirable. Such data is also needed to develop sophisticated multiscale models that can later be used for predicting the flow behavior and molecular dynamics of supramolecular networks. This thesis focuses on the experimental rheological study of two class of supramolecularpolymers namely: (a) hydrogen bonding polymers, and (b) ionic bonding polymers (hereafter termed as ionomers). We study linear and non-linear rheology fora model system of entangled pure poly(n-butyl acrylate), PnBA, homopolymer and four poly(acrylic acid), PnBA-PAA, copolymers with varying AA side groups synthesized via hydrolysis of pure PnBA homopolymer. Second, we investigate the linear viscoelastic response of entangled 2-ureido-4[1H]-pyrimidones (UPy) based supramolecular networks. Third, we investigate the rheological response of ionomers synthesized via condensation of sulfonated pthalates with poly(tetramethylene glycol)PTMO, both in non-linear shear and extensional flows. Finally, we study brittle fracture of the above ionomers in extensional flow.

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Shabbir, A. (Intern), Hassager, O. (Intern), Skov, A. L. (Intern)
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Self-Healing, High-Permittivity Silicone Dielectric Elastomer

Currently used dielectric elastomers do not have the ability to self-heal after detrimental events such as tearing or electrical breakdown, which are critical issues in relation to product reliability and lifetime. In this paper, we present a self-healing dielectric elastomer that additionally possesses high dielectric permittivity and consists of an interpenetrating polymer network of silicone elastomer and ionic silicone species that are cross-linked through proton exchange between amines and acids. The ionically cross-linked silicone provides self-healing properties after electrical breakdown or cuts made directly to the material due to the reassembly of the ionic bonds that are broken during damage. The dielectric elastomers presented in this paper pave the way to increased lifetimes and the ability of dielectric elastomers to survive millions of cycles in high-voltage conditions.

Silicone elastomers with aromatic voltage stabilizers

A method to synthesise a polydimethylsiloxane-polyphenylmethylsiloxane (PDMS-PPMS) copolymer with electron-trapping effects has been developed utilising a quick hydrosilylation reaction at ambient temperature and crosslinking via
Silicone elastomers with aromatic voltage stabilizers

Electrical breakdown causes short-circuiting of dielectric elastomers (DEs) resulting in irreversible destruction of the DE. The electrical field at which the un-actuated elastomer breaks down is known as the electrical breakdown strength. Numerous studies have been performed in order to obtain elastomers with high relative permittivity and low Young’s modulus in order to increase the actuation performance at a given voltage, but the optimised elastomers often possess relatively low electrical breakdown strength. On the other hand, increasing the electrical breakdown strength of DEs allows for larger actuation due to the possibility of utilizing larger electrical fields. Many studies on electrical breakdown of pre-stained DEs have been conducted, however less efforts have been focused on improving the electrical breakdown strength such as by blending in additives or by polymer structure modifications. In order to increase the electrical breakdown strength of polymers for e.g. the cable industry, additives like aromatic voltage stabilizers are used. Earlier works on using voltage stabilizers in polymers have mainly focused on polyethylene with the purpose of reducing power loss for high voltage insulation cables. As an alternative to utilise additives as voltage stabilizers, grafting aromatic compounds to silicone backbones may overcome the common problem of insolubility of the aromatic voltage stabilizer in the silicone elastomers due to phase separation. Preventing phase separation during preparation and during operation is a key requirement for long life-times of DEs.

A method to synthesise a polydimethylsiloxane-polyphenylmethylsiloxane (PDMS-PPMS) copolymer with electron-trapping effects has been developed utilising a quick hydrosilylation reaction at ambient temperature and crosslinking via hydrosilylation by a vinyl-functional crosslinker. The mechanism of electron-trapping by aromatic compounds grafted to silicone backbones in a crosslinked PDMS is illustrated in Fig. 1. The electrical breakdown strength, the storage modulus and the loss modulus of the elastomer were investigated, as well as the excitation energy from the collision between electron carriers and benzene rings in PDMS-PPMS copolymer was measured by UV-vis spectroscopy. The developed elastomers were inherently soft with enhanced electrical breakdown strength due to delocalized pi-electrons of aromatic rings attached to the silicone backbone. The dielectric relative permittivity of PDMS-PPMS copolymers remained between 2 to 3 with low conductivity and low dielectric loss as well as high storage moduli with low viscous loss, thereby maintaining the electro-mechanical integrity of the elastomer.

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Silicone backbones in a crosslinked PDMS is illustrated in Fig. 1. The electrical breakdown strength, the storage modulus and the loss modulus of the elastomer were investigated, as well as the excitation energy from the collision between electron carriers and benzene rings in PDMS-PPMS copolymer was measured by UV-vis spectroscopy. The developed elastomers were inherently soft with enhanced electrical breakdown strength due to delocalized pi-electrons of aromatic rings attached to the silicone backbone. The dielectric relative permittivity of PDMS-PPMS copolymers remained between 2 to 3 with low conductivity and low dielectric loss as well as high storage moduli with low viscous loss, thereby maintaining the electro-mechanical integrity of the elastomer.

Silicone elastomers with superior softness and dielectric properties
Dielectric elastomers (DEs) change their shape and size under a high voltage or reversibly generate a high voltage when deformed. The obstacle of high driving voltages, however, limits the commercial viability of the technology at present. Driving voltage can be lowered by decreasing the Young's modulus and increasing the dielectric permittivity of silicone elastomers. One such prominent method of modifying the properties is by adding suitable additives. [1] The major drawbacks for adding solid fillers are agglomeration and increasing stiffness which is often accompanied by the decrease of electrical breakdown and achievable strain.[2] In this work, three liquid additives - inert silicone oil, chloropropyl-functional silicone oil, and synthesized chloropropyl-functional copolymer - were blended into commercial silicone elastomers, and their properties were investigated. The functional groups were determined by NMR and morphology structures were investigated by optical microscopy. The resulting elastomers were evaluated with respect to their dielectric permittivity, tear and tensile strengths, as well as electrical breakdown strength, as well as low amounts of additives whereas it decreased at larger amounts. The elastomers became increasingly soft with increasing addition of soft filler. Both inert and chloropropyl-functional silicone oils enhanced the molecular motions of the network substructures via dynamic dilution effects but the viscous losses also increased with increasing amount of silicone oils. Cross-linkable chloropropyl-functional copolymer offered a high level of mechanical integrity of the blended elastomers thus consequent low viscous losses. The dielectric permittivity of chloropropyl-functional blended elastomers increased greatly compared to the pristine commercial elastomer, while the dielectric losses remained at a low level. The increase in dielectric permittivity stemmed from the high dipole moment of the chloride groups. Furthermore, the alkyl chloride units yielded a larger free volume resulted in a less dense material with a lower Young's modulus.[3]
structures were investigated by optical microscopy. The resulting elastomers were evaluated with respect to their dielectric permittivity, viscoelasticity and tensile strengths, as well as electrical breakdown.

Silicone elastomers with superior softness and dielectric properties

Silicone-modified graphene oxide fillers via the Piers-Rubinsztajn reaction

While graphene or graphene oxide can make significant improvements in the properties of a wide variety of polymeric materials, their incorporation can be challenged by incompatibility with the polymeric matrix. The modification of graphene oxide with silicones or silanes using the Piers-Rubinsztajn reaction improves dispersibility in nonpolar materials, including organic solvents and silicone pre-elastomers. A high loading (up to 10 wt %) of modified graphene oxide in silicone elastomers could be achieved, which resulted in enhanced mechanical performance and reduced gas permeability.
Silicone rubbers for dielectric elastomers with improved dielectric and mechanical properties as a result of substituting silica with titanium dioxide

One prominent method of modifying the properties of dielectric elastomers (DEs) is by adding suitable metal oxide fillers. However, almost all commercially available silicone elastomers are already heavily filled with silica to reinforce the otherwise rather weak silicone network and the resulting metal oxide filled elastomer may contain too much filler. We therefore explore the replacement of silica with titanium dioxide to ensure a relatively low concentration of filler. Liquid
Silicone rubber (LSR) has relatively low viscosity, which is favorable for loading inorganic fillers. In the present study, four commercial LSRs with varying loadings of silica and one benchmark room-temperature vulcanizable rubber (RTV) were investigated. The resulting elastomers were evaluated with respect to their dielectric permittivity, tear and tensile strengths, electrical breakdown, thermal stability and dynamic viscosity. Filled silicone elastomers with high loadings of nano-sized titanium dioxide (TiO2) particles were also studied. The best overall performing formulation had 35 wt.% TiO2 nanoparticles in the POWERSIL® XLR LSR, where the excellent ensemble of relative dielectric permittivity of 4.9 at 0.1 Hz, breakdown strength of 160 V μm−1, tear strength of 5.3 MPa, elongation at break of 190%, a Young’s modulus of 0.85 MPa and a 10% strain response (simple tension) in a 50 V μm−1 electric field was obtained.

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Synthesis and characterization of UV photocrosslinkable hydrogels with poly(N-vinyl-2-pyrrolidone): Determination of the network mesh size distribution
Hydrogels of poly(n-vinyl-2-pyrrolidone) were produced by UV irradiation of aqueous solutions of the polymer in presence of hydrogen peroxide, used as initiator. The mechanical and the nanostructural properties of the gels were characterized by a combination of experimental techniques including rheology, low field nuclear magnetic resonance spectroscopy (LF-NMR), and small angle X-ray scattering. Different irradiation doses as well as polymer and initiator concentrations were tested in the characterization. The study elucidates the relationship between different methods to estimate the mesh size of the gel polymeric network. Moreover, a novel correlation model was developed based on Chui and Scherer theories for the interpretation of LF-NMR dataset of polymer solutions and networks.

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Organisations: Department of Micro- and Nanotechnology, Nanoprobes, Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Intelligent Drug Delivery and Sensing Using Microcontainers and Nanomechanics, University of Trieste, National Research Council of Italy

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Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
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BFI (2014): BFI-level 1
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BFI (2013): BFI-level 1
Scopus rating (2013): SJR 0.827 SNIP 1.167 CiteScore 2.41
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BFI (2012): BFI-level 1
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BFI (2011): BFI-level 1
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Scopus rating (2005): SJR 0.265 SNIP 0.465
Scopus rating (2004): SJR 0.228 SNIP 0.42
Scopus rating (2003): SJR 0.24 SNIP 0.33
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The Current State of Silicone-Based Dielectric Elastomer Transducers

Silicone elastomers are promising materials for dielectric elastomer transducers (DETs) due to their superior properties such as high efficiency, reliability and fast response times. DETs consist of thin elastomer films sandwiched between compliant electrodes, and they constitute an interesting class of transducer due to their inherent lightweight and potentially large strains. For the field to progress towards industrial implementation, a leap in material development is required, specifically targeting longer lifetime and higher energy densities to provide more efficient transduction at lower driving voltages. In this review, the current state of silicone elastomers for DETs is summarised and critically discussed, including commercial elastomers, composites, polymer blends, grafted elastomers and complex network structures. For future developments in the field it is essential that all aspects of the elastomer are taken into account, namely dielectric losses, life-time and the very often ignored polymer network integrity and stability.

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Authors: Madsen, F. B. (Intern), Daugaard, A. E. (Intern), Hvilsted, S. (Intern), Skov, A. L. (Intern)
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Scopus rating (2016): CiteScore 4.19 SJR 1.711 SNIP 0.937
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BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.78 SNIP 1.06 CiteScore 4.66
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.795 SNIP 1.133 CiteScore 4.72
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 1.769 SNIP 1.123 CiteScore 4.78
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BFI (2012): BFI-level 1
Scopus rating (2012): SJR 2.096 SNIP 1.244 CiteScore 4.7
ISI indexed (2012): ISI indexed yes
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BFI (2011): BFI-level 1
Scopus rating (2011): SJR 1.999 SNIP 1.119 CiteScore 4.47
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 2.042 SNIP 1.144
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 2.226 SNIP 1.155
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The role of transport processes on dielectric elastomer performance

A dielectric elastomer (DE) is a thin elastomer film sandwiched between two compliant electrodes, and upon application of an electrical field the DE increases in area and decreases in width. DEs are soft transducers that can be used as actuators, generators and sensors. They are sought optimized by various means, but most of them include introducing high-permittivity particles into the elastomer film. These particles should theoretically increase the actuation performance, but unfortunately they also cause premature electrical breakdown to occur.

The scope of this project is to understand the parameters that lead to breakdown by modelling the performance of DEs during operation. A breakdown may arise from the effect of several different electrical aging mechanisms, which can be categorized as either degradation or intrinsic breakdown mechanisms based on the required time to cause a breakdown of the materials, illustrated in Figure 1. The mechanisms of most importance are electrical and thermal breakdown, which are also the ones of most interest in this project. A preliminary model of thermal breakdown will be used as one of the starting points for further modelling of breakdown of DEs during operation.
A new soft dielectric silicone elastomer matrix with high mechanical integrity and low losses

Though dielectric elastomers (DEs) have many favourable properties, the issue of high driving voltages limits the commercial viability of the technology. Driving voltage can be lowered by decreasing the Young's modulus and increasing the dielectric permittivity of silicone elastomers. A decrease in Young's modulus, however, is often accompanied by the loss of mechanical stability and thereby the lifetime of the DE. A new soft elastomer matrix, with no loss of mechanical stability and high dielectric permittivity, was prepared through the use of alkyl chloride-functional siloxane copolymers. Furthermore, the increase in dielectric permittivity (43%) was obtained without compromising other important properties of DEs such as viscous and dielectric losses as well as electrical breakdown strength.

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Scopus rating (2016): CiteScore 3.06 SJR 0.889 SNIP 0.757
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 0.947 SNIP 0.834 CiteScore 3.42
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BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.113 SNIP 0.962 CiteScore 3.87
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Scopus rating (2013): SJR 1.119 SNIP 0.904 CiteScore 3.74
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Scopus rating (2012): SJR 0.872 SNIP 0.619 CiteScore 2.4
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Bibliographical note
A soft and conductive PDMS-PEG block copolymer as a compliant electrode for dielectric elastomers

Conductive PDMS-PEG block copolymers (Mn = 3 – 5 kg/mol) were chain-extended (Mn = 30 – 45 kg/mol) using hydrosilylation reaction as presented in figure 1. Subsequently, the extended copolymers were added to a conductive nano-filler (multi-walled carbon nanotubes – MWCNTs) in order to enhance conductivity. The combination of soft chainextended PDMS-PEG block copolymers and conductive MWCNTs results in a soft and conductive block copolymer composite which potentially can be used as a compliant and highly stretchable electrode for dielectric elastomers. The addition of MWCNTs into the PDMS-PEG matrix not only increases the conductivity, but also increases mechanical strength by reinforcing the network. However, incorporating MWCNTs into the PDMS-PEG matrix is challenging due to strong van der Waals forces between the MWCNTs. In the present study, MWCNTs were dispersed in organic solvent (N-methyl pyrrolidinone) with 1 wt% of surfactant (Triton X-100). The dispersion of MWCNTs in PDMS-PEG systems shown in figure 2 where MWCNTs (dark areas) are well-distributed in the system indicating an acceptable dispersional though some big clusters appear in the optical microscope image. The conductivity of 4 phr MWCNTs is 10-3 S/cm compared to 10-1 S/cm of a non-stretchable reference conducting silicone elastomer (LR3162 from Wacker). Furthermore, PDMS-PEG block copolymer with 4 phr MWCNTs (Young’s modulus, Y = 0.26 MPa) is softer and more stretchable than LR3162 (Y = 1.17 MPa).

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A solvent evaporation route towards fabrication of hierarchically porous ZSM-11 with highly accessible mesopores

A route to generate hierarchically porous zeolite ZSM-11 has been paved via solvent evaporation induced self-assembly assisted by hexadecyltrimethoxysilane to produce a preformed dry gel, followed by its subsequent transformation into zeolite via steam-assisted-crystallization. The crystallization in dry gel has been found to undergo an orientated attachment growth mechanism whereby hexadecyltrimethoxysilane directs the formation of auxiliary mesopores and inhibits the fusion of primary nucleates. Measurements such as XRD, SEM, TEM, N2-physisorption, and TEM for an inverse replica of Pt derived from hierarchical ZSM-11 have been conducted to characterize the textural properties of the material. Ammonia temperature-programmed-desorption (NH3-TPD) measurements and infrared spectra using probe molecules such as pyridine (Py-IR) and 2,4,6-collidine (Coll-IR) have been collected to investigate the acidic properties as well as the accessibility of the acid sites. The hierarchical ZSM-11 possesses more acid sites on the mesopore surfaces that are accessible towards large probe molecules such as 2,4,6-collidine. This improvement together with the enhanced pore-connectivity brings about an increase in 1,3,5-triisopropylbenzene cracking activity and benzene selectivity with respect to a conventional counterpart.

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Bilaterally Microstructured Thin Polydimethylsiloxane Film Production

Thin PDMS films with complex microstructures are used in the manufacturing of dielectric electro active polymer (DEAP) actuators, sensors and generators, to protect the metal electrode from large strains and to assure controlled actuation.

The current manufacturing process at Danfoss PolyPower A/S produces films with a one-sided microstructured surface only. It would be advantageous to produce a film with both surfaces microstructured, as this increases the film’s performance efficiency. The new technique introduced herein produces bilaterally microstructured film by combining an embossing method with the existing manufacturing process. In employing the new technique, films with microstructures on both surfaces are successfully made with two different liquid silicone rubber (LSR) formulations: 1) pure XLR630 and 2) XLR630 with titanium dioxide (TiO2). The LSR films (∼70 μm) are cast on a carrier web substrate using a coating blade. The carrier web, which has a sinusoidal corrugation with wave height of 7μm and a wave period of 7μm on its surface, imparts corrugations to the bottom surface of the film. The elastomer film on the carrier web is preheated to the gel point, where the elastomer film can retain an imprint made on it. The preheated film at gel point is embossed between the rolls of a gravure lab coater, which corrugates the top surface of the film. The films are then heated, in order to cure completely. For the LSR systems used in this process, the optimum conditions for preheating are 110°C for 4–7 s, while for embossing the temperature is 110°C with 25 psi pressure between the rolls at a speed of 1.4 rpm. Scanning electron microscope (SEM) images confirm the formation of microstructures on both the surfaces of the film.

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Controlled release in hard to access places by poly(methyl methacrylate) microcapsules triggered by gamma irradiation

Gamma irradiation was investigated as a triggering stimulus for the activation of poly(methyl methacrylate) (PMMA) microcapsules. PMMA was exposed to varying doses of irradiation and analyzed by differential scanning calorimetry, size-exclusion chromatography, and nuclear magnetic resonance. It was found that the glass transition temperature (Tg) of the polymer decreases at low irradiation doses. Additionally, Tg can be physically adjusted by adding a plasticizer, and both kinds of microcapsules were successfully prepared with non-plasticized and plasticized PMMA shell. Finally, impermeable microcapsules were shown to become permeable after irradiation and release an encapsulated cross-linker, which
enables the remotely controlled formation of polydimethylsiloxanes in traditionally unavailable places. Therefore, the activation method has significant implications for industrial application.
Design of Elastomer Structure to Facilitate Incorporation of Expanded Graphite in Silicones Without Compromising Electromechanical Integrity

The development of elastomer materials with a high dielectric permittivity has attracted increased interest over the past years due to their use in, for example, dielectric elastomers. For this particular use, both the electrically insulating properties - as well as the mechanical properties of the elastomer - have to be tightly controlled in order not to destroy favorable elastic properties by the addition of particles. This study focuses on improving the electromechanical properties of an enhanced PDMS matrix with expanded graphite (EG) as filler. The PDMS matrix is crosslinked by means of an 8-functional crosslinker, which allows for development of a suitable network matrix. The dielectric permittivity was increased by almost a factor of 4 compared to a benchmark silicone elastomer.
Dielectric elastomers, with very high dielectric permittivity, based on silicone and ionic interpenetrating networks

Dielectric elastomers (DEs), which represent an emerging actuator and generator technology, admittedly have many favourable properties, but their high driving voltages are one of the main obstacles to commercialisation. One way to reduce driving voltage is by increasing the ratio between dielectric permittivity and the Young's modulus of the elastomer. One system that potentially achieves this involves interpenetrating polymer networks (IPNs), based on commercial silicone elastomers and ionic networks from amino- and carboxylic acid-functional silicones. The applicability of these materials as DEs is demonstrated herein, and a number of many and important parameters, such as dielectric permittivity/loss, viscoelastic properties and dielectric breakdown strength, are investigated. Ionic and silicone elastomer IPNs are promising prospects for dielectric elastomer actuators, since very high permittivities are obtained while dielectric breakdown strength and Young's modulus are not compromised. These good overall properties stem from the softening effect and very high permittivity of ionic networks – as high as $\varepsilon' = 7500$ at 0.1 Hz – while the silicone elastomer part of the IPN provides mechanical integrity as well as relatively high breakdown strength. All IPNs have higher dielectric losses than pure silicone elastomers, but when accounting for this factor, IPNs still exhibit satisfactory performance improvements.
In this study a new method of multi-walled carbon nanotube (MWCNT) incorporation was employed in the preparation of ultraviolet (UV) curable MWCNT-filled poly(dimethyl siloxane) (PDMS) composites. The composites were designed to contain amounts of MWCNT above the percolation threshold, without becoming conductive. Ultrasonicated and dispersed MWCNTs were co-precipitated together with an excess of short chain alpha, omega-vinyl terminated PDMS with a deficient amount of thiol-crosslinker and a photoinitiator (2,2-dimethoxy-2-phenylacetophenone, DMPA) into MeOH. The entire mixture was UV irradiated, resulting in a layer of hyperbranched PDMS forming around the MWCNTs. This MWCNT mixture was added to a hyperbranched long chain PDMS to provide concentrations of MWCNT of 0.33%, 0.66% and 1%, and a fully crosslinked system was obtained in a final photochemical curing. Rheology of the composites showed a moderate decrease in storage modulus (G') across the entire frequency range in line with an increasing amount of MWCNT, thus demonstrating that the rheological percolation threshold was not reached throughout the concentration range. Dielectric spectroscopy measurements showed an increase in permittivity in line with an increasing MWCNT content as well as the desired frequency-dependent conductivity for all samples. The composites showed moderate dielectric breakdown strength of 48 V μm(-1) at 0.33 wt% MWCNT, which decreased throughout the samples to 20 V μm(-1) at 1 wt%. Temperature-dependent AC conductivity studies revealed that an increase in the sample temperature could explain the premature breakdown observed for those composites with higher MWCNT loading.
Enhancement of dielectric permittivity by incorporating PDMS-PEG multiblock copolymers in silicone elastomers

A silicone elastomer from PDMS-PEG multiblock copolymer has been prepared by use of silylation reactions for both copolymer preparation and crosslinking. The dielectric and mechanical properties of the silicone elastomers were carefully investigated, as well as the morphology of the elastomers was investigated by SEM. The developed silicone elastomers were too conductive to be utilized as dielectric elastomers but it was shown that when the above silicone elastomers were mixed with a commercial silicone elastomer, the resulting elastomer had very favourable properties for dielectric elastomers due to a significantly increased dielectric permittivity. The conductivity also remained low due to the resulting discontinuity in PEG within the silicone matrix.
Enhancing relative permittivity by incorporating PDMS-PEG multi block copolymers in binary polymer blends

Polydimethylsiloxanes (PDMS) are well-known to actuate with relatively large strains due to low modulus, but they possess low permittivity. Contrary, polyethylene glycols (PEG) are not stretchable but possess high permittivity. Combination of the two polymers in a block copolymer depicts a possibility for substantial improvement of properties such as high permittivity, stretchability and non-conductivity if carefully designed. The objective is to synthesize PDMS-PEG multiblock copolymers assembling into discontinuous morphologies in PEG based on variation of volume fractions of PDMS. The utilized synthesis of PDMS-PEG multiblock copolymer is based on hydrosilylation reaction, which is amended from Klasner et al.1 and Jukarainen et al.2 Variation in the ratio between the two constituents introduces distinctive properties in terms of dielectric permittivity and rheological behaviour. PDMS-PEG multiblock copolymers are, however, shown to be conductive (figure 1.a) and thus not capable of actuating. By incorporating conductive PDMS-PEG multiblock copolymers into commercial PDMS elastomer, the discontinuity in PEG can be obtained and the relative permittivity ($\varepsilon'$) is significantly enhanced 60% with 5wt% of PDMS-PEG block copolymer incorporated in the PDMS network (figure 1.b).

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Enhancing relative permittivity by incorporating PDMS-PEG multiblock copolymers in binary polymer blends

Polydimethylsiloxane (PDMS) elastomers are well-known to be soft and highly stretchable, yet they never achieve maximum elongation when utilised as dielectric elastomers, simply because their dielectric permittivity remains rather low. Conversely, polyethyleneglycols (PEG) are not stretchable, but they do possess high permittivity. Combining two such polymers in a block copolymer allows for further crosslinking and presents the possibility of substantial improvements in the actuation response of the resulting dielectric elastomer – if carefully designed. The objective is to synthesise a PDMS-PEG multiblock copolymer, assembling it into discontinuous morphologies in PEG in a continuous PDMS network. PDMS-PEG multiblock copolymers, however, have been shown to be conductive and thus not capable of actuating. By incorporating conductive PDMS-PEG multiblock copolymers into a commercial PDMS elastomer, the discontinuity in PEG can be acquired and the relative permittivity ($\varepsilon'$) is significantly enhanced (60%) with 5wt% of PDMS-PEG block copolymer incorporated into the silicone elastomer.

Functional silicone copolymers and elastomers with high dielectric permittivity

Dielectric elastomers (DEs) are a new and promising transducer technology and are often referred to as ‘artificial muscles’, due to their ability to undergo large deformations when stimulated by electric fields. DEs consist of a soft and thin elastomeric film sandwiched between compliant electrodes, thereby forming a capacitor [1]. Silicone elastomers are one of the most used materials for DEs due to their high efficiency, fast response times and low viscous losses. The major disadvantage of silicone elastomers is that they possess relatively low dielectric permittivity, which means that a high electrical field is necessary to operate the DE. The necessary electrical field can be lowered by creating silicone elastomers with higher dielectric permittivity, i.e. with a higher energy density. The aim of this work is to create new and improved silicone elastomers with high dielectric permittivity. This was done through the synthesis of new functionalizable siloxane copolymers [2] that allow for the attachment of high dielectric permittivity molecules through copper-catalyzed azide-alkyne 1,3-dipolar cycloaddition (CuAAC) reactions. The synthesised siloxane copolymers were prepared via the tris(pentafluorophenyl)borane catalysed Piers-Rubinsztajn reaction [3] and have a high degree of chemical freedom, as several parameters can be varied during the preparation phase. Thus, the space between the functional groups can be varied, by using different dimethylsiloxane spacer units between the high dielectric permittivity molecules. Furthermore, the degree of functionalization of the copolymers can be varied accurately by changing the feed of the high dielectric permittivity molecules. As a result, a completely tuneable elastomer system, with respect to functionalization, is achieved.
azide-alkyne 1,3-dipolar cycloaddition (CuAAC)[3]. The synthesised copolymers allow for a high degree of chemical freedom, as several parameters can be varied during the preparation phase. As an example, the space between the functional groups can be varied, by using different dimethylsiloxane spacer units between the functional molecules. Furthermore, the degree of functionalisation of the copolymers can be varied accurately by changing the feed of functional molecules. As a result, a completely tuneable elastomer system, with respect to functionalisation, is achieved. The second method of functionalising silicone elastomers involves the synthesis of a so-called ‘chain extender’ that allows for chemical modifications such as CuAAC. This route is promising as an easy-to-use additive to commercial RTV silicone elastomer systems. We have investigated how the different functionalisation variables affect elastomer properties including dielectric and viscoelastic properties.

High energy density interpenetrating networks from ionic networks and silicone

The energy density of dielectric elastomers (DEs) is sought increased for better exploitation of the DE technology since an increased energy density means that the driving voltage for a certain strain can be lowered in actuation mode or alternatively that more energy can be harvested in generator mode. One way to increase the energy density is to increase dielectric permittivity of the elastomer. A novel silicone elastomer system with high dielectric permittivity was prepared through the development of interpenetrating networks from ionically assembled silicone polymers and covalently crosslinked silicones. The system has many degrees of freedom since the ionic network is formed from two polymers (amine and carboxylic acid functional, respectively) of which the chain lengths can be varied, as well as the covalent silicone elastomer with many degrees of freedom arising from amongst many the varying content of silica particles. A parameter study is performed to elucidate which compositions are most favorable for the use as dielectric elastomers. The elastomers were furthermore shown to be self-repairing upon electrical breakdown.
Improving dielectric permittivity by incorporating PDMS-PEG block copolymer into PDMS network

Polydimethylsiloxane (PDMS) based elastomers are well-known to actuate with large strain mainly due to their low modulus and their non-conducting nature. On the other hand, polyethyleneglycols (PEG) are not stretchable but they have high permittivity and are conductive. Combination of the two polymers as a block copolymer depicts a possibility for substantial improvement of properties such as high permittivity and non-conductivity – if carefully designed. The objective is to synthesize PDMS-PEG multiblock copolymer assembling into different morphologies such as lamellar, cylinder, gyroid and spheres based on variation of volume fractions of PDMS and PEG. The synthesis is amended from Klasner et al. and Jukarainen et al. Variation in the ratio between the two constituents introduces distinctive properties in terms of dielectric permittivity and rheological behaviour. PDMS-PEG multiblock copolymer-based elastomers of different volume fractions exhibit high storage permittivity but they are conductive. By incorporating conductive PDMS7-PEG multiblock copolymers into a commercial non-conductive PDMS elastomer (MJK) creates promising morphology which enhances storage permittivity by 60% with 5wt% of PDMS7-PEG block copolymer incorporated in the PDMS network.

Investigation into Accessible Surface Vinyl Concentrations of Nonstoichiometric PDMS Microspheres from Hydrosilylation Reactions and Their Further Crosslinking Reactions

The introduction of surface vinyl groups to PDMS microspheres broadens the latter’s applicability range since the microspheres can be further functionalized or crosslinked into elastomers. Quantification of the surface vinyl concentration of PDMS microspheres is therefore essential. Here, a novel titration method, which is based on efficiently and covalently bonding mono-functional hydride to vinyl, is employed to determine surface vinyl concentration. The titration method exhibits good reproducibility in detection, thereby underlining its potential applicability as a general titration method for cured silicones. Vinyl functional PDMS microspheres are mixed with a hydride crosslinker and non-reactive silicone oil to create a system which allows for extensive crosslinking. Both visual observations and rheological studies show that a robust macroscopic PDMS elastomer is obtained upon crosslinking. Furthermore, the influence of stoichiometric imbalance, and the weight fraction of silicone oil on the terminal storage modulus of the macroscopic PDMS elastomer, is investigated. The wide range of stoichiometries facilitating crosslinking between the PDMS microspheres and the hydride crosslinker found in this study is beneficial for delivering PDMS elastomer to hard-to-reach places, since the sensitivity of the system on concentration fluctuations is small.
Materials science: Like cartilage, but simpler

The properties of articular cartilage, which lines bones in joints, depend partly on repulsion between components of the material. A new synthetic gel that mimics this feature has rare, direction-dependent properties.
Mechanically invisible encapsulations

Research into dielectric elastomers has intensified within the last two decades, due to the realisation that these materials undergo significant deformations when subjected to high electric fields. High efficiency, lightweight, low-cost and simple working principles are the main advantages of this technology. A major part of the research on dielectric transducers is dedicated to the development of elastomeric membranes that create a basis for each potential application. It has been recognised that higher energy density and more durable materials need to be created in order to enable the commercialisation of such devices. Therefore, this project was dedicated to exploring the possibility of using polar liquids as high dielectric constant fillers for dielectric PDMS-based elastomers. Incorporating polar liquids in the form of discrete droplets into nonpolar membrane swas expected to produce a two-fold improvement with respect to a reference material. Firstly, dielectric constant enhancement and, secondly, a Young’s modulus decrease were anticipated. In the first approach a flow-focusing microfluidic technique was employed, in order to encapsulate polar liquids within a soft elastomeric shell. The produced core-shell microspheres served as a carrier for liquids, enabling the uniform dispersion of the filler droplets within PDMS prepolymer. The dielectric constant of the prepared water-PDMS composite was proven to be enhanced by 30% following the incorporation of 4.5 wt.% of water. Due to the favourable structure of the capsules, mechanical properties remained unaffected. Importantly, the approach substantiated the high potential of liquid-PDMS composites for dielectric transducers. In the second part of the study a new method for producing liquid-PDMS composites was developed and thoroughly investigated. Applying very high shear forces to mixtures of PDMS preelastomers and polar liquids facilitated the preparation of stable and uniform emulsions. Upon crosslinking the PDMS (which formed the continuous phase of the emulsions), stable hybridelastomers were obtained. The method allowed for incorporating up to 50% by volume of various liquids, which resulted in significant improvements to the dielectric constant of the composites. An incorporation of 120 wt.% of glycerol increased the dielectric constant of a commercial PDMS composition by 380%. Additionally a three-fold decrease in the elastic modulus was observed. Although the dielectric properties of the composites were very promising, the materials exhibited leakage current at high electric fields. Therefore, further study on improving high-voltage performance was conducted, exhibiting the high potential of the material. The second approach brought a breakthrough in the research on liquid-PDMS composites. The developed technique proved to be very versatile, thereby allowing for the preparation of multiplehybrid materials with very distinct properties – an attractive proposition from the point of view of multiple scientific fields.
The thesis includes a comprehensive overview of the fundamentals behind a CFD software, as well as a more detailed review of the fluid dynamic phenomena investigated in this project. The momentum and continuity equations are presented as well as the theory behind the SST and the k-ε turbulence models. Modelling of additional variables, porous materials and two-phase flows are also introduced. The two-phase flows are modelled using the Euler-Euler method, and both dispersed and free-surface flows are simulated.

The importance of mass transfer with a focus on mixing, gas-liquid transfer of oxygen, and heterogeneous reactor systems is reviewed and mathematical models for these applications are presented. A review of how these mass transfer phenomena have been modelled in the scientific literature is also included.

The models are subsequently evaluated based on their applicability in the four case studies. The evaluations especially focus on the impact of the choice of turbulence model and other modelling decisions made by the user. The conclusion is that CFD is a highly valuable tool for modelling several important parameters in chemical and biochemical reactors but that the user must be well aware of the shortcomings with the applied models.

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Optimization of large-scale fabrication of dielectric elastomer transducers
Dielectric elastomers (DEs) have gained substantial ground in many different applications, such as wave energy harvesting, valves and loudspeakers. For DE technology to be commercially viable, it is necessary that any large-scale production operation is nondestructive, efficient and cheap. Danfoss Polypower A/S employs a large-scale process for manufacturing DE films with one-sided corrugated surfaces.

The DEs are manufactured by coating an elastomer mixture to a corrugated carrier web, thereby imprinting the corrugations onto the elastomer. The corrugated elastomer is then sputtered with metal electrodes on the corrugated surface, and due to these corrugated surfaces the metal electrodes maintain conductivities up to more than 100% strain of the elastomer film. The films are then laminated in multiple layers to fabricate DE transducers. However, the current manufacturing process is not trouble-free, and two issues in particular have great influence on the performance of DE transducers. The first issue is the release of the corrugated elastomer film from the carrier web, due to the large surface area and flexible nature of the elastomer film, while the second issue relates to the lamination of DE films. Currently, the films are contacted without adhesion, in order to yield double-sided corrugations, which in turn causes friction between the layers and thus reduces the lifespan of the transducer. Furthermore, air can be trapped in the interface, thereby causing a decrease in electrical breakdown strength. Other issues may also arise, depending on how the elements are assembled.

This thesis is based on optimising the large-scale manufacture of DE transducers. The hot embossing technology is used to impart corrugations onto elastomer film surfaces. Embossing, which was performed for samples with different r-values, showed that the process was applicable for different hardnesses of the material, while time intervals for the different materials were also determined. An attempt to solve the lamination issue was made by applying stronger adhesion methods when preparing monolithic elements. Due to the corrugations, the films were able to adhere in different configurations (back-to-back, front-to-back and front-to-front). The first route involved adhering PDMS to PDMS (back-to-back), for which two routes were followed. The first route involved using an aminosilane as an adhesion agent after modifying the PDMS surface through plasma treatment, which demonstrated that the laminates were slightly stiffer and more fragile in respect to tearing. The other route involved modifying the surfaces through plasma treatment and by adhering the layers, which showed to be a suitable method and allowed high-strength laminates to perform as monolithic elements. For the front-to-back and front-to-front configurations, conductive elastomers were utilised. One approach involved adding the cheap and conductive filler, exfoliated graphite (EG) to a PDMS matrix to increase dielectric permittivity. The results showed that even at low concentrations, EG influenced the overall performance of the reinforced elastomer matrix, indicating that increasing the concentration further to make conductive elastomers would compromise
the elastic nature of the elastomer due to large EG dimensions; consequently, EG-based elastomers as conductive adhesives were rejected. Dielectric properties below the percolation threshold were subsequently investigated, in order to conclude the study. In order to avoid destroying the network structure, carbon nanotubes (CNTs) were used as fillers during the preparation of the conductive elastomers and as received CNTs and modified CNTs were investigated. The unmodified CNTs were mixed with an ionic liquid, and two dispersion methods were investigated. The first method involved the ultrasonication of CNT/IL, which showed that conductivity increased in line with increasing CNT at concentrations lower than 5 wt% when CNTs were mixed with IL. The second method involved roll milling the composite, which decreased conductivity when concentrations exceeded 4 wt% for the CNT/IL composites. However, the obtained conductivities were increased for similar concentrations by roll milling. The modified CNTs were grafted covalently to the CNT surface with poly(methacryloyl polydimethylsiloxane), resulting in the obtained conductivities being comparable to commercially available Elastosil LR3162, even at low functionalisation. The optimized methods allow new processes for the production of DE film with corrugations and efficient DE transducers.

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PDMS and MWCNT – How to Obtain an Efficient and Controlled Distribution of Conductive Fillers in PDMS
Polydimethylsiloxane (PDMS) elastomers employing conductive fillers are used in many applications for e.g. flexible electrode materials or with lower amounts of fillers for high capacitance elastomers. Traditionally, the most used filler in these applications has been carbon black, which through high loading results in sufficiently high conductivities. During recent years, the range of conductive fillers has been extended to include i.e. expanded graphite, single walled carbon nanotubes and multi walled carbon nanotubes (MWCNT), whereof in particular MWCNT are interesting due to their outstanding electrical and mechanical properties. However, the use of MWCNT for many new applications requires efficient processing strategies in order to result in elastomers with an efficient dispersion of the nanomaterial. If it is possible to obtain an efficient dispersion of the nanomaterial in the PDMS precursors the mixture should additionally be able to crosslink without interference from the nanofiller. There are several possible pathways to obtain such dispersions, where these could be divided into two main strategies, direct mixing using processing equipment or modification of the MWCNT followed by traditional preparation of the crosslinked elastomer. Both pathways have been investigated and the presentation will outline results from both approaches.

Direct processing of the MWCNT together with PDMS prepolymers by mechanical mixing, sonication, speedmixing or roll milling have been investigated. It is very clear that in order to obtain sufficiently effective dispersion, it is necessary to use the more efficient methods such as roll milling or speed mixing to distribute the fillers. Processing aids such as ionic liquids have been tested and found effective, though it requires higher amounts of additive or combinations with the most effective mixing methods and a thorough mixing in order to provide good dispersions.

As an alternative to direct mixing, modification of MWCNT is a well-known approach to ease dispersion of nanomaterials. This can be done by surface initiated polymerizations by e.g. atom transfer radical polymerization (ATRP) using compatibilizing monomers. Through the surface initiated polymerization a thin coating of polymer is introduced on the MWCNT to prevent agglomeration and permit much easier dispersion into the targeted polymer such as a PDMS prepolymer. Through simple methods of either entrapment or free radical grafting methods functionalized MWCNT (f-MWCNT) are prepared and applied in preparation of elastomers resulting in easy and efficient dispersion of the nanofillers in the elastomer. In addition to this, the choice of method permits preparation of composites with either well distributed fillers or entrapped fillers providing access to high capacitance composites with an artificially high percolation threshold or to obtain conductive elastomers with a MWCNT loading of 5 wt% only.

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Polydimethylsiloxane microspheres with poly(methyl methacrylate) coating: Modelling, preparation, and characterization

Polydimethylsiloxane (PDMS) microspheres are prepared by mixing homogeneous dispersions of vinyl-functional PDMS and a curing agent using mechanical stirring in a series of aqueous solutions, and curing at 80°C for 2 h. In order to verify the experimental diameter and size distributions of the PDMS microspheres, the Hinze-Kolmogorov theory is applied to predict the mean diameter, and a population balance model as well as the maximum entropy formalism are used to describe the size distribution. Close agreement is found between experimental and theoretical results. Furthermore, vinyl functional PDMS microspheres were coated with poly(methyl methacrylate) (PMMA) by spin coating with different concentrations of PMMA solutions. The quality of the resulting PMMA shell is investigated using rheological measurements at 50°C with a timesweep procedure. The results strongly suggest that PMMA-coated PDMS microspheres react around 20 times slower than the uncoated ones, and that the PMMA shell significantly hinders the reaction between the PDMS microsphere and cross-linker. Thus the thin PMMA shells are very efficient in protecting the reactive PDMS microspheres, since the PMMA shell forms an impermeable barrier up to 50°C.
Preparing mono-dispersed liquid core PDMS microcapsules from thiol–ene–epoxy-tailored flow-focusing microfluidic devices

An applied dual-cure system based on thiol–ene and thiol–epoxy “click chemistry” reactions was proved to be an extremely effective and easy to use tool for preparing microfluidic chips, thereby allowing for precise control over material properties and providing the possibility of covalently bonding chip wafers. Different thiol–ene–epoxy-based polymer compositions were tested with the help of DSC and ATR FTIR, in order to investigate their physical and chemical properties. Water contact angles were determined, thus verifying the high efficiency and selectivity of the chemical surface modification of compositions in relation to high hydrophilicity and hydrophobicity. An obtained microfluidic device was subsequently used in order to produce PDMS microcapsules of very narrow size distribution and which contained various common liquids, such as water and ethanol, as well as an ionic liquid 2-hydroxyethylammonium formate.

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Silicone elastomers with high dielectric permittivity and high dielectric breakdown strength based on tunable functionalized copolymers

High driving voltages currently limit the commercial potential of dielectric elastomers (DEs). One method used to lower driving voltage is to increase dielectric permittivity of the elastomer. A novel silicone elastomer system with high dielectric permittivity was prepared through the synthesis of siloxane copolymers, thereby allowing for the attachment of high dielectric permittivity molecules through copper-catalyzed azide-alkyne 1,3-dipolar cycloaddition (CuAAC). The synthesized copolymers allow for a high degree of chemical freedom, as several parameters can be varied during the preparation phase. Thus, the space between the functional groups can be varied, by using different dimethylsiloxane spacer units between the dipolar molecules. Furthermore, the degree of functionalization can be varied accurately by changing the feed of dipolar molecules. As a result, a completely tunable elastomer system, with respect to functionalization, is achieved. It is investigated how the different functionalization variables affect essential DE properties, including dielectric permittivity, dielectric loss, elastic modulus and dielectric breakdown strength, and the optimal degree of chemical functionalization, where these important properties are not significantly compromised, is also determined. Thus, the best overall properties were obtained for a silicone elastomer prepared with 5.6 wt% of the dipolar molecule 1-ethyl-4-nitrobenzene. Here, a high increase in dielectric permittivity (~70%) was obtained without compromising other vital DE properties such as elastic modulus, gel fraction, dielectric and viscous loss and electrical breakdown strength.
Dielectric elastomer, Dielectric constant, Permittivity, Breakdown, Silicone, CuAAC, Click reaction

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Soft Functional Silicone Elastomers with High Dielectric Permittivity: Simple Additives vs. Cross-Linked Synthesized Copolymers

Though dielectric elastomers (DEs) have many favorable properties, the issue of high driving voltages limits the commercial viability of the technology. Improved actuation at lower voltages can be obtained by decreasing the Young’s modulus and/or decreasing the dielectric permittivity of the elastomer. A decrease in Young’s modulus, however, is often accompanied by the loss of mechanical stability and thereby the lifetime of the DE whereas addition of high permittivity fillers such as metal oxides often increases Young’s modulus such that improved actuation is not accomplished. New soft silicone elastomers with high dielectric permittivity were prepared through the use of chloropropyl-functional silicones. One method was through the synthesis of modular cross-linkable chloropropyl-functional copolymers that allow for a high degree of chemical freedom such that a tuneable silicone elastomer system is obtained. This system provides high dielectric permittivity without compromising other important properties of DEs such as viscous and dielectric losses as well as electrical breakdown strength. Another method was through a one-pot system where functional silicone copolymers are added as an additive to commercial silicone elastomer systems. Here the functional copolymer acts both as a permittivity enhancer and plasticizer. We show how the DE properties and the dielectric permittivity to Young’s modulus ratio are improved for these systems and we compare the use of cross-linkable polymers against the use of plasticizers with similar chemical structures on the final mechanical stability and lifetime of DEs.

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Super soft silicone elastomers with high dielectric permittivity

Dielectric elastomers (DEs) have many favourable properties. The obstacle of high driving voltages, however, limits the commercial viability of the technology at present. Driving voltage can be lowered by decreasing the Young’s modulus and increasing the dielectric permittivity of silicone elastomers. A decrease in Young’s modulus, however, is often accompanied by the loss of mechanical stability and thereby the lifetime of the DE. New soft elastomer matrices with high dielectric permittivity and low Young’s modulus, with no loss of mechanical stability, were prepared by two different approaches using chloropropyl-functional silicone polymers. The first approach was based on synthesised chloropropyl-functional copolymers that were cross-linkable and thereby formed the basis of new silicone networks with high dielectric permittivity (e.g. a 43% increase). These networks were soft without compromising other important properties of DEs such as viscous and dielectric losses as well as electrical breakdown strength. The second approach was based on the addition of commercially available chloropropyl-functional silicone oil to commercial LSR silicone elastomer. Two-fold increase in permittivity was obtained by this method and the silicone oil decreased the Young’s modulus significantly. The viscous losses, however, also increased with increasing content of silicone oil. Cross-linkable chloropropyl-functional copolymers offer a new silicone elastomer matrix that could form the basis of dielectric elastomers of the future, whereas the chloropropyl silicone oil approach is an easy tool for improvement of the properties of existing commercial silicone elastomers.

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Techniques for hot embossing microstructures on liquid silicone rubbers with fillers

Embossing is an established process for the thermoplastic elastomers but not yet for the thermosetting elastomers. It has already been shown that hot embossing is a viable technology for imprinting microstructures in addition to curing thin silicone films at their gel point. It is one of the simplest, most cost-effective, and time-saving methods for replicating microstructures. In the present study, films made from liquid silicone rubber (LSR) formulations containing fillers are hot embossed under modified operating conditions. The use of such relatively hard silicone elastomers shows the versatility of this method that has been established for softer silicone elastomers. Also, as a proof of concept, a microstructured metal (nickel (Ni)) plate is used as an embosser for the films successfully. The ideal condition for hot embossing the LSR formulation (XLR 630 with titanium dioxide fillers) with a Ni embosser is 110°C preheating for 15–35 s, embossed with 2 bar pressure, and postheating for complete curing at 110°C for 3 min showing that the process is extremely fast.

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The electrical breakdown strength of pre-stretched elastomers, with and without sample volume conservation

In practice, the electrical breakdown strength of dielectric electroactive polymers (DEAPs) determines the upper limit for transduction. During DEAP actuation, the thickness of the elastomer decreases, and thus the electrical field increases and the breakdown process is determined by a coupled electro-mechanical failure mechanism. A thorough understanding of the mechanisms behind the electro-mechanical breakdown process is required for developing reliable transducers. In this study, two experimental configurations were used to determine the stretch dependence of the electrical breakdown strength of polydimethylsiloxane (PDMS) elastomers. Breakdown strength was determined for samples with and without volume conservation and was found to depend strongly on the stretch ratio and the thickness of the samples. PDMS elastomers are shown to increase breakdown strength by a factor of ∼3 when sample thickness decreases from 120 to 30 μm, while the biaxial pre-stretching (λ = 2) of samples leads similarly to an increase in breakdown strength by a factor of ∼2.5.

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Delft University of Technology, Danfoss PolyPower A/S
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The influence of static pre-stretching on the mechanical ageing of filled silicone rubbers for dielectric elastomer applications

Dielectric elastomer (DE) pre-stretching is a key aspect of attaining better actuation performance, as it helps prevent electromechanical instability (EMI) and usually lowers the Young’s modulus, thus leading to easier deformation. The pre-stretched DE is not only susceptible to a high risk of tearing and the formation of mechanical defects, but films with sustained and substantial strain may also experience mechanical degradation. In this study a long-term mechanical reliability study of DE is performed. Young’s moduli, dielectric breakdown strengths and dielectric permittivities of commercial silica-reinforced silicone elastomers, with and without an additional 35% (35 phr) of titanium dioxide (TiO2), were investigated after being subjected to pre-stretching for various timespans at pre-stretches to strains of 60 and 120%, respectively. The study shows that mechanical stability when pre-stretching is difficult to achieve with highly filled elastomers. However, despite the negative outlook for metal oxide-filled silicone elastomers, the study paves the way for reliable dielectric elastomers by indicating that simply post-curing silicone elastomers before use may increase reliability.

Mechanically Invisible Polymer Coatings

The present invention relates to a composition comprising encapsulated particles in a polymeric material. The composition comprises a continuous phase and a discontinuous phase incorporated therein, wherein the continuous phase comprises a first polymeric material and wherein the discontinuous phase comprises particles, said particles comprising a filler material and an encapsulating coating of a second polymeric material, wherein the backbones of the first and second polymeric materials are the same. The composition may be used in electroactive polymers (EAPs) in order to obtain mechanically invisible polymer coatings.
A method of providing a barrier in a fracture-containing system

The present invention relates to a method of providing a barrier in a fracture-containing system, comprising: i) Providing a treatment fluid comprising: a) a base fluid; b) an elastomeric material, wherein said elastomeric material comprises at least one polymer capable of crosslinking into an elastomer, and c) at least one crosslinking agent; ii) Placing the treatment fluid in a fracture-containing system; iii) Allowing the elastomeric material to crosslink with itself to form a barrier in said fracture-containing system; wherein the elastomeric material and/or the crosslinking agent are of neutral buoyancy with regard to the base fluid. The invention is contemplated to having utility not only in the oil-drilling industry but also in the plugging of fractures in sewer drains, pipelines etc.

Control of PDMS crosslinking by encapsulating a hydride crosslinker in a PMMA microcapsule

In the preparation of PDMS elastomers, a combination of mixing and reactive processes constrains the applicability of the PDMS elastomer in research and applications. Separation of the mixing and reactive processes, which control PDMS crosslinking, has been achieved by encapsulating a hydride crosslinker in a PMMA shell. Microcapsules are mixed with vinyl-terminated PDMS to create a gelation system, which allows for storage at 50 °C, without premature gelation, and in addition allows for extensive crosslinking reaction at 120 °C. Both visual observations and rheological studies show that a robust PDMS elastomer is obtained upon heating the gelation system. Furthermore, the influence of stoichiometric imbalance on the equilibrium storage modulus of the PDMS network is investigated, by employing different amounts of microcapsules in vinyl-terminated PDMS. It has been found that adding microcapsules increases the equilibrium storage modulus of the PDMS elastomer until the diffusion of the hydride crosslinker is constricted. An optimum amount of crosslinker used in the control crosslinking reaction has also been found. However, compared to the pure PDMS elastomer, the modulus of the PDMS elastomer from the encapsulated system is less sensitive in relation to the stoichiometry of the system than the corresponding polymer network. This broadens the applicability range of silicone elastomers.
Dielectric electroactive polymers comprising an ionic supramolecular structure

The present invention relates to an ionic interpenetrating polymer network comprising at least one elastomer and an ionic supramolecular structure comprising the reaction product of at least two chemical compounds wherein each of said compounds has at least two functional groups and wherein said compounds are able to undergo Lewis acid-base reactions. The interpenetrating polymer network may be used as dielectric electroactive polymers (DEAPs) having a high dielectric permittivity.

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Dielectric properties of supramolecular ionic structures obtained from multifunctional carboxylic acids and amines

The dielectric properties of several supramolecular ionic polymers and networks, linked by the ammonium salts of hexamethylene diamine (HMDA), tris(2-aminoethyl)amine (TAEA), poly(propylene imine) (PPI) dendrimers and two short bis carboxymethyl ether-terminated poly(ethylene glycol)s (DiCOOH-PEG), are reported in this paper. All supramolecular ionic polymers and networks exhibit very high relative dielectric permittivities (3 × 10^2 – 10^6) at low frequencies, and significantly lower values (from 1 up to 26) at high frequencies. Additionally, the dielectric properties of supramolecular ionic networks, formed by mixing multifunctional carboxylic acids such as citric acid (CA), tricarballylic acid (TCAA), trimesic acid (TMA), ethylenediaminetetraacetic acid (EDTA) and diethylenetriaminepentaacetic acid (DETPA) with two different Jeffamine polyetheramines (designated as D400 and D2000), are investigated. Here the relative dielectric permittivities of the supramolecular ionic structures formed with the multifunctional carboxylic acids were lower than those from the supramolecular ionic structures formed with the two carboxymethyl ether-terminated poly(ethylene glycol)s.
Effects of fillers on the properties of liquid silicone rubbers (LSRs)

Dielectric electro active polymers (DEAPs) change their shape and size under a high voltage or reversibly generate a high voltage when deformed [1,2]. One prominent method to make DEAPs performances more efficient is by adding suitable fillers [3,4]. Liquid silicone rubbers (LSRs) have relatively low viscosities, which is favorable for loading of inorganic fillers [5]. In this study, commercially available fillers, such as fumed silica (SiO2), titanium dioxide (TiO2), barium titanate (BaTiO3), copper calcium titanate (CaCu3Ti4O12, CCTO), multi-walled carbon nanotubes (MWCNTs) were added into the LSRs. The filled elastomers have both favorable properties and shortcomings. The results indicate that the hydrophobic rutile TiO2 nanofiller is a good candidate for achieving higher permittivity and breakdown, as well as favorable elastic modulus of the elastomers.

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DEAPs, LSRs, Dielectric permittivity, Electrical breakdown, Young’s modulus, TiO2

Effects of fillers on the properties of liquid silicone rubbers (LSRs)

Dielectric electro active polymers (DEAPs) change their shape and size under a high voltage or reversibly generate a high voltage when deformed. To make their performances more efficient, certain properties of the polymers like their dielectric permittivity, electrical breakdown and Young’s modulus have to be modified according to the specifications of their respective applications. One such prominent method of modifying the properties is by adding suitable fillers. Liquid silicone rubbers (LSRs) have relatively low viscosities when compared with thermoplastics, which is favorable for loading of inorganic fillers. The property improvement of the filled LSRs depends on filler concentration, filler morphology, such as particle size and structure, the degree of dispersion and orientation in the matrix, and also the degree of adhesion with the polymer chains, as well as the properties of the inorganic fillers.

In this study commercially available fillers, such as fumed silica (SiO2), titanium dioxide (TiO2), barium titanate (BaTiO3), copper calcium titanate (CaCu3Ti4O12, CCTO), multi-walled carbon nanotubes (MWCNTs) were added into the LSRs and we examined how the properties of the networks were modified. The filled elastomers have both favorable properties and shortcomings. The shortcomings are of various types. Fumed silica reinforces the networks with no increase in permittivity (εr,SiO2 ~ 3.9). Barium titanate possesses high dielectric constant (εr,BaTiO3 ~ 150) but its heavy density (6.08 g/mL) deteriorates the lightweight advantage of the DEAPs. Micron-sized giant dielectric constant CCTO (εr,CCTO ~ 10000) decreases the mechanical performances of the composites. The inhomogeneous compatibility of the unmodified MWCNTs in the silicone system causes the risk of electric conductivity. Among these additives, the use of multiple titanium oxides as filler potentially suits to special applications. In the present study, a series of TiO2 fillers were blended into LSRs, such as hydrophilic/ hydrophobic, micro/ nano scale, anatase/ rutile crystal, sphere/ core-shell structure. The results indicate that the hydrophobic rutile TiO2 nanofiller is a good candidate for achieving higher permittivity and breakdown strength, as well as favorable elastic modulus of the prepared elastomers.

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Encapsulated PDMS microspheres with reactive handles

Cured poly(dimethyl siloxane) microspheres are prepared by an emulsion polymerization reaction of silicone droplets in a continuous aqueous phase. The commonly used PDMS elastomer, Sylgard 184 from Dow Corning, is used as the dispersed phase. PDMS is polymerized and cross-linked by reacting vinyl end-terminated poly(dimethyl siloxane) oligomers with dimethylmethylhydrogen siloxane cross-linkers via the hydrosilylation reaction using platinum catalyst and heat. Weight ratios of 10:1, 20:1, and 25:1 of the PDMS mixtures are used and emulsified in water using two water-soluble surfactants as stabilizers (sodium dodecyl sulphate and polyvinyl alcohol). The temperature is subsequently increased to accelerate the rate of cross-linking and prevent the prepolymer droplets from coalescing. The particle size distribution of cured PDMS microspheres is determined by Mastersizer (laser diffraction). Finally, cured PDMS microspheres are coated with poly(methyl methacrylate) using a chemical process (solvent evaporation technique). Three solvents are used in three different experiments: dichloromethane, tetrahydrofuran, and acetone. The composition and morphology of the cured PDMS microspheres and PMMA coated cured PDMS microspheres are characterized by differential scanning calorimetry, Fourier transform infrared spectroscopy in attenuated-total-reflection mode, optical microscopy, and thermogravimetric analysis. Curing profiles of PDMS elastomer with different ratios between the silicone elastomer base and the silicone elastomer curing agent are obtained. The reactivity of cured PDMS microspheres and PMMA coated cured PDMS microspheres are measured by rheology to evaluate the efficiency of the PMMA coating. © 2014 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

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Favorable electrical breakdown strengths of prestretched elastomers with and without sample volume conservation

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**Filled liquid silicone rubbers: Possibilities and challenges**

Liquid silicone rubbers (LSRs) have been shown to possess very favorable properties as dielectric electroactive polymers due to their very high breakdown strengths (up to 170 V/μm) combined with their fast response, relatively high tear strength, acceptable Young's modulus as well as they can be filled with permittivity enhancing fillers. However, LSRs possess large viscosity, especially when additional fillers are added. Therefore both mixing and coating of the required thin films become difficult. The solution so far has been to use solvent to dilute the reaction mixture in order both to ensure better particle dispersion as well as following for film formation properties. We show that the mechanical properties of the films as well as the electrical breakdown strength can be affected, and that the control of the amount of solvent throughout the coating process is essential for solvent borne processes. Another problem encountered when adding solvent to the highly filled reaction mixture is the loss of tension in the material upon large deformations. These losses are shown to be irreversible and happen within the first large-strain cycle.

**High breakdown-strength composites from liquid silicone rubbers**

In this paper we investigate the performance of liquid silicone rubbers (LSRs) as dielectric elastomer transducers. Commonly used silicones in this application include room-temperature vulcanisable (RTV) silicone elastomers and composites thereof. Pure LSRs and their composites with commercially available fillers (an anatase TiO2, a core–shell TiO2-SiO2 and a CaCu3Ti4O12 filler) are evaluated with respect to dielectric permittivity, elasticity (Young's modulus) and electrical breakdown strength. Film formation properties are also evaluated. The best-performing formulations are those with anatase TiO2 nanoparticles, where the highest relative dielectric permittivity of 5.6 is obtained, and with STX801, a core–shell morphology TiO2-SiO2 filler from Evonik, where the highest breakdown strength of 173 V μm−1 is obtained.
Improving dielectric permittivity by incorporating PDMS-PEG block copolymer into PDMS network

Polydimethylsiloxane (PDMS) is well-known to cause hydrophobic surfaces due to their low surface energy as well as they possess low conductivity and extremely low glass transition temperature. On the other hand, polyethyleneglycol (PEG) behaves hydrophilic and is a highly conductive polymer. Combination of both polymers as a block copolymer depicts a possibility for substantial improvement of properties such as high permittivity and non-conductivity – if carefully designed.
The objective of my research is to synthesize PDMS-PEG block copolymer assembling into different morphologies such as lamellae, cylinder, gyroid and spheres based on different volume fraction ratio of PDMS and PEG. The synthesis of PDMS-PEG block copolymer is based on hydrosilylation reaction occurring at 60 °C and dry condition with presence of platinum catalyst as shown in Fig. 13.4. Different volume fraction of PDMS and PEG introduces different properties in terms of contact angles, dielectric permittivity and rheological behaviour. All morphologies of PDMS-PEG block copolymer in this study exhibit high storage permittivity; at the same time the loss permittivity is even higher which implies that the synthesized PDMS-PEG block copolymers are conductive. By incorporating conductive PDMS-PEG block copolymer into commercial PDMS elastomer from Wacker Chemie, the storage permittivity is significantly enhanced by 38% with 20% of PDMS-PEG block copolymer incorporated in pure PDMS network as depicted in Fig. 2.

**General information**

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Authors: A Razak, A. H. (Intern), Szabo, P. (Intern), Skov, A. L. (Intern)
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**Large scale processing of dielectric electroactive polymers**

Efficient processing techniques are vital to the success of any manufacturing industry. The processing techniques determine the quality of the products and thus to a large extent the performance and reliability of the products that are manufactured. The dielectric electroactive polymer (DEAP) technology is relatively new and is in the initial stages of development with no established large scale manufacturing techniques. Danfoss Polypower A/S has set up a large scale manufacture process to make thin film DEAP transducers. The DEAP transducers developed by Danfoss Polypower consist of microstructured elastomer surfaces on which the compliant metallic electrodes are sputtered thus enabling large strains of non-stretchable metal electrode. Thin microstructured polydimethylsiloxane (PDMS) films are quintessential in DEAP technology due to scaling of their actuation strain with the reciprocal of square of films' thickness. Production of thin elastomer film with microstructures on one or both surfaces is therefore the crucial step in the manufacturing. The manufacture process is still not perfect and further optimization is required. Smart processing techniques are required at Danfoss Polypower to solve production issues in DEAP film manufacture. The primary issue in the processing is the release of thin PDMS films from a corrugated substrate on which the films are cured. The films are weak owing to their small thickness and low Young’s modulus. The process of peeling the films from the substrate either induces pre strain or tears the films leading to a total shut down of the manufacturing. Further problem arises in the lamination step of the manufacturing process. The thin films manufactured in the industry with the current process have only one microstructured surface. The complaint electrode is sputtered on the microstructured surface of the film. Two such films are laminated to make a single DEAP laminate with two microstructured surfaces. The lamination process introduces two problems: 1) it may entrap air bubbles and dust at the interface which will cause the films to breakdown at the operating voltages and 2) after lamination the thickness of the elastomer is double that of the single film and as thickness of the elastomer between the electrodes increases, higher voltages are required to produce a desired strain.

The objective of this research work is to find suitable solutions to the above discussed technical hitches. To facilitate easy release of the films from substrates, following three approaches are investigated: 1) chemical modification of the elastomer film by adding surface active block copolymers to reduce its surface energy, 2) use of surfactants as release agents and 3) compounding a silicone resin with the elastomer matrix. An alternate process of making microstructured films by hot embossing has been established in this research. From our experiments it has been learnt that addition curing PDMS elastomer has the unique property to retain an imprint made on it at gel point. With hot embossing technique the microstructures can be imprinted directly on the films at gel point. Thus, films can be made on a flat substrate and embossed instead of making the films on a corrugated substrate. Releasing the films from a flat substrate is easier compared to the corrugated substrate. To address the lamination problem, bilaterally microstructured films can be made by combining the hot embossing method with the existing manufacturing process eliminating the lamination step. The bilaterally corrugated monolithic films have lower thickness than the laminated films made with conventional manufacture process. This new technique developed on a lab scale is quick, economical, easy and can be implemented on a large scale. The results of all our experiments and the hot embossing technique have been discussed.

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Authors: Vudayagiri, S. (Intern), Skov, A. L. (Intern)
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Publisher: Technical University of Denmark, Department of Chemical and Biochemical Engineering
Methods to ease the release of thin polydimethylsiloxane films from difficult substrates

Silicone elastomers are used as dielectric electroactive polymers for making actuators, generators, sensors, and as artificial muscles in medical applications. Current requirements in the actuator manufacturing put a strict limitation on the thickness of the elastomers, such that a maximum permissible thickness is around 25–50 µm. The relatively small Young's modulus for these elastomers is a requirement for actuation capabilities. However, peeling and release of such films during manufacture processes are very difficult. To ease the release of the films, techniques such as the use of release agents like surfactants and detergents, incorporating resins in the silicone matrix and grafting/adding low surface energy functionalities to the silicone elastomer have been tested. The methods used are required not to interfere with the Young's modulus and the dielectric permittivity in a negative way. Polysorbate-20, a non-ionic surfactant, fulfills all requirements and gives the lowest peel forces for the films.

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Monolithic growth of partly cured polydimethylsiloxane thin film layers

The demand for monolithic structures in many applications has increased to enable more reliable and optimized performances such as for dielectric electroactive polymers (DEAPs). For the layers of the elements to grow efficiently together, it is first of all required that the layers adhere together to enable interlayer crosslinking reactions either by application of an adhesion promoter or by ensuring that there are reactive, complementary sites available on the two surfaces. Polydimethylsiloxane (PDMS) is a widely used polymer for DEAPs. In this work, two-layered PDMS films are adhered together at different curing times. The monolithic films are investigated by rheology, scanning electron microscope, mechanical testing, dielectric relaxation spectroscopy, thermal gravimetric analysis (TGA) and differential scanning calorimetry (DSC). The morphology, mechanical and dielectric properties, as well as thermal stabilities of the bilayer elastomer films are observed to change with the curing time of the monolayers before lamination. The objective of this work is to create adhesion of two layers without destroying the original viscoelastic properties of the PDMS films, and hence enable, for example, adhesion of two microstructured films which is currently a crucial step in the large-scale production of DEAPs. © 2014 The Society of Polymer Science, Japan (SPSJ) All rights reserved.
Novel encapsulation technique for incorporation of high permittivity fillers into silicone elastomers

The research on soft elastomers with high dielectric permittivity for the use as dielectric electroactive polymers (DEAP) has grown substantially within the last decade. The approaches to enhance the dielectric permittivity can be categorized into three main classes: 1) Mixing or blending in high permittivity fillers, 2) Grafting of high permittivity molecules onto the polymer backbone in the elastomer, and 3) Encapsulation of high permittivity fillers. The approach investigated here is a new type of encapsulation which does not interfere with the mechanical properties to the same content as for the traditionally applied thermoplastic encapsulation. The properties of the elastomers are investigated as function of the filler content and type. The dielectric permittivity, dielectric loss, conductivity, storage modulus as well as viscous loss are compared to elastomers with the same amounts of high permittivity fillers blended into the elastomer, and it is found that the encapsulation provides a technique to enhance some of these properties.
Novel method to prepare multiwalled carbon nanotube/poly(dimethyl siloxane) (MWCNT/PDMS) non-conducting composites

In this study a new method of carbon nanotube (CNT) incorporation was employed for the preparation of ultraviolet (UV) curable CNT filled poly (dimethyl siloxane) (PDMS) composites. The composites were designed to contain loadings of CNT above the percolation threshold without becoming conductive due to a localized distribution of CNT. Ultrasonicated and dispersed multiwalled CNTs were mixed with short chain -vinyl terminated PDMS. When the whole mixture containing dispersed CNT and short chain PDMS was irradiated with UV radiation in presence of deficient amount of hexa functional thiol PDMS crosslinker and a photoinitiator, hyperbranched PDMS layer was formed over the CNTs. The prepared hyperbranched CNTs were mixed in different weight ratios (0.33%, 0.66%, 1%) with long chain -vinyl terminated PDMS and crosslinked subsequently with the same hexa functional thiol PDMS via UV photoinitiated thiol-ene chemistry to obtain the networks. Rheology of the prepared networks showed a gradual decrease in storage modulus (G’) in the entire frequency range as the amount of CNT was increased due to a reduction in crosslinking density imposed by the CNT was increased due to a reduction in crosslinking density imposed by the CNTs. Dielectric spectroscopy measurements showed an increasing trend in permittivity in all the composites with increasing CNT loadings and AC conductivity measurements confirmed non-percolating behavior of the prepared composites.

General information
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Novel water shutoff treatment using PDMS elastomer

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PEG-bis phosphonic acid based ionic supramolecular structures
A number of supramolecular structures are prepared by mixing stoichiometric amounts of a phosphonic acid terminated poly(ethylene glycol) with propargyl amine, hexamethylene diamine, tris(2-aminoethyl)amine, and the first generation of a poly(propylene imine) dendrimer in turn at room temperature. The resulting ionic assemblies are very comprehensively characterized by ATR-FTIR, proton, and carbon-13 NMR spectroscopy that unequivocally demonstrate the ionic network formation through ammonium phosphonates. The resulting salt and ionic networks are additionally analyzed by differential scanning calorimetry and thermogravimetric analysis. The conclusion is that mixing the virgin components at room temperature spontaneously form either a salt or ionic supramolecular networks. © 2014 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

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Scopus rating (2010): SJR 0.438 SNIP 0.469
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 0.55 SNIP 0.533
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 0.565 SNIP 0.516
Scopus rating (2007): SJR 0.536 SNIP 0.51
Scopus rating (2006): SJR 0.567 SNIP 0.526
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 0.559 SNIP 0.505
Scopus rating (2004): SJR 0.493 SNIP 0.401
Scopus rating (2003): SJR 0.544 SNIP 0.503
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 0.539 SNIP 0.419
Scopus rating (2001): SJR 0.492 SNIP 0.407
Scopus rating (2000): SJR 0.726 SNIP 0.521
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 0.67 SNIP 0.694
Original language: English
Organic Chemistry, Materials Chemistry, Polymers and Plastics, Condensed Matter Physics, differential scanning calorimetry, ionic networks, IR spectroscopy, NMR spectroscopy, supramolecular chemistry, thermogravimetric analysis, Fourier transform infrared spectroscopy, Infrared spectroscopy, Ionic liquids, Mixing, Nuclear magnetic resonance spectroscopy, Organic acids, Polyethylene glycols, Propylene, Supramolecular chemistry, Thermogravimetric analysis,
Physical characterization of photocrosslinked poly(vinyl pyrrolidone) (PVP) hydrogels for drug delivery

General information
State: Published
Organisations: Department of Micro- and Nanotechnology, Nanoprobes, Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, University of Trieste, University of Copenhagen
Number of pages: 2
Publication date: 2014

Host publication information
Title of host publication: Proceedings of the 8th Symposium of The Pharmaceutical Solid State Research Cluster
Main Research Area: Technical/natural sciences
Electronic versions:
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Bibliographical note
8th Symposium of The Pharmaceutical Solid State Research Cluster (PSSRC), Ljubljana, Slovenia, September 16-18 2014,
Source: PublicationPreSubmission
Source-ID: 103224915
Publication: Research - peer-review › Conference abstract in proceedings – Annual report year: 2014

Preparation and Characterization of Silicone Liquid Core/Polymer Shell Microcapsules via Internal Phase Separation
Microcapsules with a silicone liquid core surrounded by a polymeric shell were synthesised through the controlled phase separation. The dispersed silicone phase consisted of the shell polymer PMMA, a good solvent for the PMMA (dichloromethane, DCM) and a poor solvent (methylhydrosiloxane dimethylsiloxane) for the PMMA. The morphology of the PMMA micro-capsules was investigated by ATR-FTIR and by optical microscopy. Microcapsules were prepared with different emulsifiers and different concentrations of acetone and PMMA in the oil phase. The thermal stability of the PMMA microcapsule and the content of the silicone liquid core were assessed by TGA. 1H-NMR spectroscopy and an extraction method were also used to determine the content of the silicone liquid core in the microcapsules.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Maersk Oil Research and Technology Centre
Authors: Gonzalez, L. (Intern), Kostrzewska, M. (Intern), Ma, B. (Intern), Li, L. (Intern), Hansen, J. H. (Ekstern), Hvilsted, S. (Intern), Skov, A. L. (Intern)
Pages: 1259-1267
Publication date: 2014
Main Research Area: Technical/natural sciences

Publication information
Journal: Macromolecular Materials & Engineering
Volume: 299
ISSN (Print): 1438-7492
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 2.71 SJR 0.755 SNIP 0.945
Web of Science (2017): Indexed yes
Sealing device and method for sealing fractures or leaks in wall or formation surrounding tube-shaped channel

The sealing device (1) includes an elongated body (5) adapted to be introduced into a tube-shaped channel (2) and including a sealing fluid placement section (6) arranged between a first and a second annular flow barrier (7, 8). The elongated body further includes a sealing fluid activation section (11) arranged between the second annular flow barrier (8) and a third annular flow barrier (12) and including a sealing fluid activation device (13) adapted to at least initiate or accelerate curing of the sealing fluid (17). In operation, the elongated body may be displaced along the tube-shaped channel until the sealing fluid activation section is placed at a position where sealing fluid has been ejected by the sealing fluid placement section, and the sealing fluid activation device may be activated. Thereby, sealing fluid may be cured at selected locations along the tube-shaped channel after ejection of sealing fluid.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Hansen, J. H. (Ekstern), Skov, A. L. (Intern)
Publication date: 2014
Silicone elastomers with high dielectric permittivity and high dielectric breakdown strength based on dipolar copolymers

Dielectric elastomers (DES) are a promising new transducer technology, but high driving voltages limit their current commercial potential. One method used to lower driving voltage is to increase dielectric permittivity of the elastomer. A novel silicone elastomer system with high dielectric permittivity was prepared through the synthesis of siloxane copolymers, thereby allowing for the attachment of high dielectric permittivity molecules through copper-catalysed azide-alkyne 1,3-dipolar cycloaddition (CuAAC). The copolymers have a high degree of chemical freedom, as the dimethylsiloxane spacer units between the functional groups, as well as the degree of functionalisation, can be varied. Thus, the best overall properties were obtained for an elastomer prepared with a copolymer with a 1200 g mol(-1) dimethylsiloxane spacer unit and 5.6 wt% of the high dielectric permittivity molecule 1-ethynyl-4-nitrobenzene. Here, a high increase in dielectric permittivity (similar to 70%) was obtained without compromising other favourable DE properties such as elastic modulus, gel fraction, dielectric loss and electrical breakdown strength. © 2014 Elsevier Ltd. All rights reserved.
Soft silicone based interpenetrating networks as materials for actuators

A new approach based on silicone interpenetrating networks with orthogonal chemistries has been investigated with focus on developing soft and flexible elastomers with high energy densities and small viscous losses. The interpenetrating networks are made as simple two pot mixtures as for the commercial available silylation based elastomers such as Elastosil RT625. The resulting interpenetrating networks are formulated to be softer than RT625 to increase the actuation caused when applying a voltage due to their softness combined with the significantly higher permittivity than the pure silicone elastomers.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Energy Resources Engineering
Authors: Yu, L. (Intern), Gonzalez, L. (Intern), Hvilsted, S. (Intern), Skov, A. L. (Intern)
Publication date: 2014

Host publication information
Title of host publication: Proceedings of SPIE
Volume: 9056
Publisher: SPIE - International Society for Optical Engineering
Synthesis and characterizations of high permittivity ultraviolet cured soft elastomeric networks and composites applicable as dielectric electroactive polymer

The objective of this thesis was preparation and characterizations of high permittivity ultraviolet (UV) cured elastomeric networks and composites applicable as dielectric electroactive polymers (DEAPs). At present, none of the commercially available elastomers such as acrylics, poly (dimethyl siloxane) (PDMS) and polyurethanes are designed with the requirements specific for DEAPs. Thus there is a need to develop elastomers with low elastic modulus, low viscous and dielectric losses and high relative permittivity. Interpenetrating networks and fumed silica reinforced composites of poly(propylene oxide) (PPO) were prepared which showed marked improvements in properties compared to the acrylic elastomers. But difficulties in curing by industrial processes and handling of these elastomers posed as limitations. So the focus was on optimizing UV induced thiol-ene reactions for curing commercially available PDMS. UV curing of PDMS was successfully established which eliminated the major drawbacks of widely used platinum catalyzed addition curing of PDMS. An advanced sequential curing used to form the PDMS networks showed low elastic modulus and low viscous losses than the former-developed processes due to better control over the heterogeneity of the networks. The sequential curing approach was successfully used to incorporate conductive multiwalled carbon nanotubes (MWCNTs) in higher concentrations than usual without making the elastomers conductive. The PDMS-MWCNT composites also showed high relative permittivity, low elastic modulus and low viscous and dielectric losses. Thus the elastomers developed in this project show promising properties to be considered as potential DEAPs.

Synthesis of telechelic vinyl/allyl functional siloxane copolymers with structural control

Multifunctional siloxane copolymers with terminal vinyl or allyl functional groups are synthesised through the borane-catalysed polycondensation of hydrosilanes and alkoxysilanes. Copolymers of varying molecular weights (M_w =13200 – 70 300 g mol^-1), spatially well-distributed functional groups and high end-group fidelity are obtained in a facile and robust synthetic scheme involving polycondensation, end-group transformation and different functionalisation reactions such as Cu(I)-mediated azide – alkyne cycloaddition. Pendant alkyl chloride, alkyl azide, bromoisobutyryl, 4-nitrobenzene and 1-ethyl-imidazolium chloride fragments with programmable spatial distributions are incorporated in the copolymer backbones. NMR and FTIR spectroscopy as well as size exclusion chromatography corroborate the efficacy and versatility of this modular approach.
Synthetic Strategies for High Dielectric Constant Silicone Elastomers

Dielectric electroactive polymers (DEAPs) are a new and promising transducer technology and are often referred to as 'artificial muscles', due to their ability to undergo large deformations when stimulated by electric fields. DEAPs consist of a soft and thin elastomeric film (an elastomer) sandwiched between compliant electrodes and have many interesting properties such as a simple working principle, large achievable strains, high-energy densities and the fact that they are lightweight and low in cost. Some issues, however, limit the current commercial viability of the technology, as high driving voltages (several kV's), for instance, are necessary to activate the material. Driving voltage can be lowered by increasing the energy density of the DEAP elastomer film, achieved by creating elastomers with high dielectric constants, which is a material's ability to store electrical energy. Two synthetic strategies were developed in this Ph.D. thesis, in order to create silicone elastomers with high dielectric constants and thereby higher energy densities. The work focused on maintaining important properties such as dielectric loss, electrical breakdown strength and elastic modulus. The methodology therefore involved chemically grafting high dielectric constant chemical groups onto the elastomer network, as this would potentially provide a stable elastomer system upon continued activation of the material. The first synthetic strategy involved the synthesis of a new type of cross-linker for silicone polymer networks. The silicone compatible cross-linker allowed for copper-catalysed azide-alkyne cycloadditions (CuAAC) and thereby the attachment of functional groups to the network crosslinking point. The functional groups were very well-distributed in the silicone elastomer matrix, and various functional
groups provided a number of elastomers with diverse properties. High dielectric constant molecules, such as the dipolar 4-nitrobenzene and nitroazobenzene, resulted in elastomers with an approximately 20% increase in dielectric constant at low concentrations of dipolar species (~0.5 wt%). The second synthetic strategy was to create elastomers with high concentrations of functional groups and thereby even higher dielectric constants through the synthesis of novel copolymers. Two different routes were followed to accomplish this aim. One route involved the synthesis of a so-called ‘chain extender’ that allowed for chemical modifications such as Cu- AAC. This route was promising for one-pot elastomer preparation and as a high dielectric constant additive to commercial silicone systems. The second approach used the borane-catalysed Piers-Rubinsztajn reaction to form spatially well-distributed copolymers where functional groups could be attached along the polymer backbone. The functional copolymers contained vinyl or allyl end groups that allowed for elastomer synthesis. The dielectric properties of the formed elastomers were found to increase significantly and an optimum concentration of functional groups was identified. At a concentration of 5.6 wt% of a nitrobenzene functional group the dielectric permittivity increased 70% while at this loading important properties such as electrical breakdown strength, elastic modulus and dielectric loss were not significantly compromised. The developed synthetic strategies facilitate new ways of functionalising elastomers in general and dielectric elastomers in particular.

**General information**

State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Madsen, F. B. (Intern), Skov, A. L. (Intern), Daugaard, A. E. (Intern), Hvilsted, S. (Intern)
Number of pages: 187
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**Publication information**

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Main Research Area: Technical/natural sciences
Electronic versions: Frederikke_Bahrt_Madsen_978_87_93054_56_1.pdf
Source: PublicationPreSubmission
Source-ID: 104901786
Publication: Research › Ph.D. thesis – Annual report year: 2015

**The Electrical Breakdown of Thin Dielectric Elastomers: Thermal Effects**

Dielectric elastomers are being developed for use in actuators, sensors and generators to be used in various applications, such as artificial eye lids, pressure sensors and human motion energy generators. In order to obtain maximum efficiency, the devices are operated at high electrical fields. This increases the likelihood for electrical breakdown significantly. Hence, for many applications the performance of the dielectric elastomers is limited by this risk of failure, which is triggered by several factors. Amongst others thermal effects may strongly influence the electrical breakdown strength. In this study, we model the electrothermal breakdown in thin PDMS based dielectric elastomers in order to evaluate the thermal mechanisms behind the electrical failures. The objective is to predict the operation range of PDMS based dielectric elastomers with respect to the temperature at given electric field. We performed numerical analysis with a quasi-steady state approximation to predict thermal runaway of dielectric elastomer films. We also studied experimentally the effect of temperature on dielectric properties of different PDMS dielectric elastomers. Different films with different percentages of silica and permittivity enhancing filler were selected for the measurements. From the modeling based on the fitting of experimental data, it is found that the electrothermal breakdown of the materials is strongly influenced by the increase in both dielectric permittivity and conductivity.

**General information**

State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Process Engineering and Technology, Center for Energy Resources Engineering, Delft University of Technology, Danfoss PolyPower A/S
Authors: Zakaria, S. B. (Intern), Morshuis, P. H. F. (Ekstern), Yahia, B. M. (Ekstern), Gernaey, K. (Intern), Skov, A. L. (Intern)
Number of pages: 11
Publication date: 2014

**Host publication information**

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Publisher: SPIE - International Society for Optical Engineering
ISBN (Print): 9780819499820
Main Research Area: Technical/natural sciences
Conference: SPIE Smart Structures/NDE, San Diego, CA, United States, 09/03/2014 - 09/03/2014
DEAP, PDMS, Electrothermal breakdown, Numerical method
Electronic versions:
UV-Cured, Platinum-Free, Soft Poly(dimethylsiloxane) Networks

To overcome the drawbacks exhibited by platinum-catalyzed curing of silicones, photoinitiated thiol–ene cross-linking of high-molecular-weight poly(dimethylsiloxane) (PDMS) prepolymers has been investigated as a pathway to novel soft PDMS networks, based on commercially available starting materials was developed. Through a fast and efficient two-step cross-linking reaction highly flexible PDMS elastomers were prepared.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Energy Resources Engineering
Authors: Goswami, K. (Intern), Skov, A. L. (Intern), Daugaard, A. E. (Intern)
Pages: 9230 – 9233
Publication date: 2014
Main Research Area: Technical/natural sciences

Publication information
Journal: Chemistry: A European Journal
Volume: 20
ISSN (Print): 0947-6539
Ratings:
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 4.9 SJR 2.265 SNIP 1.02
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 5.03 SJR 2.352 SNIP 1.068
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 2.461 SNIP 1.195 CiteScore 4.99
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 2.526 SNIP 1.222 CiteScore 5.51
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 2.643 SNIP 1.239 CiteScore 5.68
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 2.935 SNIP 1.291 CiteScore 5.55
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): SJR 2.902 SNIP 1.319 CiteScore 5.46
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 2.791 SNIP 1.295
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Scopus rating (2009): SJR 2.753 SNIP 1.425
Web of Science (2009): Indexed yes
Visualisation and characterisation of heterogeneous bimodal PDMS networks

The existence of short-chain domains in heterogeneous bimodal PDMS networks has been confirmed visually, for the first time, through confocal fluorescence microscopy. The networks were prepared using a controlled reaction scheme where short PDMS chains were reacted below the gelation point into hyperbranched structures using a fluorescent silicone compatible cross-linker. The formation of the hyperbranched structures was confirmed by FTIR, 1H-NMR and size exclusion chromatography (SEC). The short-chain hyperbranched structures were thereafter mixed with long-chain hyperbranched structures to form bimodal networks with short-chain domains within a long-chain network. The average sizes of the short-chain domains were found to vary from 2.1 to 5.7 mm depending on the short-chain content. The visualised network structure could be correlated thereafter to the elastic properties, which were determined by rheology. All heterogeneous bimodal networks displayed significantly lower moduli than mono-modal PDMS elastomers prepared from the long polymer chains. Low-loss moduli as well as low-sol fractions indicate that low-elastic moduli can be obtained without compromising the network's structure.
A control scheme for filament stretching rheometers with application to polymer melts

We propose a new control scheme to maintain a constant strain rate of the mid-filament diameter in a filament stretching rheometer for polymer melts. The scheme is cast as a velocity algorithm and consists of a feedback and a feed-forward contribution. The performance of the controller is demonstrated on a commercial low density polyethylene. Several strain rates and experimental conditions are tested to demonstrate the necessity of the control parameters and the limits of both the control scheme and the experimental apparatus. When the control parameters are properly tuned, the algorithm ensures that the applied strain stays within 2% of the set point and measurements can be made up to Hencky strains of 6.5.
Adhesion between Polydimethylsiloxane Layers by Crosslinking

Adhesion between two surfaces may be strongly improved by chemical crosslinking of the interfaces. Polydimethylsiloxane (PDMS) is a widely used polymer that has received considerable attention due to its unique properties, such as relatively low price, biocompatibility, flexibility, high thermal stability, and outstanding dielectric properties. The excellent performances
of PDMS elastomers enable the realization of pneumatic, electromagnetic, and thermal actuators. In this work, two-layered PDMS films were adhered together by different mixtures of crosslinkers. The double-layered films were investigated by rheology and microscopy. The objective of this work was to create adhesion of two layers without destroying the original viscoelastic properties of the PDMS films.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Yu, L. (Intern), Daugaard, A. E. (Intern), Skov, A. L. (Intern)
Pages: 47-52
Publication date: 2013
Main Research Area: Technical/natural sciences

Publication information
Journal: Advances in Science and Technology
Volume: 79
ISSN (Print): 1662-0356
Ratings:
ISI indexed (2013): ISI indexed no
ISI indexed (2012): ISI indexed no
ISI indexed (2011): ISI indexed no
Original language: English
Adhesion, Polydimethylsiloxane , Crosslinking, Doublelayered
DOIs:
10.4028/www.scientific.net/AST.79.47
Source: dtu
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Publication: Research - peer-review › Journal article – Annual report year: 2013

Concentrated Polymer Solutions are Different from Melts: Role of Entanglement Molecular Weight
We compare viscoelastic properties of several polystyrene solutions and melts with the same number of entanglements. It is demonstrated that the modulus and time can be shifted such that the linear viscoelastic properties are identical provided the number of entanglements are identical. However the nonlinear properties in strong extensional flow are different with polymer solutions showing markedly stronger extensional hardening than the corresponding melts. While increased chain extensibility for solutions may provide part of the explanation, it is demonstrated that other mechanisms are needed for a full explanation for the differences between solutions and melts.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Department of Micro- and Nanotechnology, Amphiphilic Polymers in Biological Sensing, Department of Mechanical Engineering, Manufacturing Engineering
Authors: Huang, Q. (Intern), Mednova, O. (Intern), Rasmussen, H. K. (Intern), Javier Alvarez, N. (Intern), Skov, A. L. (Intern), Almdal, K. (Intern), Hassager, O. (Intern)
Number of pages: 10
Pages: 5026–5035
Publication date: 2013
Main Research Area: Technical/natural sciences

Publication information
Journal: Macromolecules
Volume: 46
Issue number: 12
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Ratings:
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Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 5.86 SJR 2.419 SNIP 1.513
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 5.76 SJR 2.564 SNIP 1.483
Web of Science (2016): Indexed yes
Development of novel PDMS elastomers for dielectric electroactive polymers (DEAPs)

General information
Dielektrisk spektroskopi i aktion som nyttigt polymeranalyseværktøj

Dielektrisk spektroskopi er blevet langt mere tilgængeligt som karakteriseringsværktøj både offline og direkte tilsluttet forskelligt produktionsudstyr. Her gives en række eksempler på anvendelser, hvor ellers svært tilgængelig information er blevet langt nemmere at opnå vha. dielektrisk spektroskopi.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Energy Resources Engineering
Authors: Bahrt, F. (Intern), Daugaard, A. E. (Intern), Hvilsted, S. (Intern), Skov, A. L. (Intern)
Number of pages: 1
Publication date: 2013
Event: Abstract from EPF 6th summer school, Gargnano, Italy.
Main Research Area: Technical/natural sciences
Electronic versions:
  Abstract-Frederikke Bahrt.pdf

Bibliographical note
Poster presentation
Source: dtu
Source-ID: u:7509
Publication: Research - peer-review › Conference abstract for conference – Annual report year: 2013

Dielektrisk spektroskopi til karakterisering af plastmaterialer

Dielektrisk spektroskopi er blevet et vigtigt redskab i arbejde med polymere materialer. Her gives en introduktion til metoden dielektrisk spektroskopi og hvilke fænomener teknikken er baseret på med fokus på analyse af polymere systemer.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Danish Technological Institute
Authors: Daugaard, A. E. (Intern), Skov, A. L. (Intern), Bøgelund, J. P. (Ekstern)
Pages: 20-21
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Main Research Area: Technical/natural sciences
Publication information
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Volume: 94
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  ISI indexed (2012): ISI indexed no
  ISI indexed (2011): ISI indexed no
  Web of Science (2007): Indexed yes
  Web of Science (2004): Indexed yes
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  Source-ID: 127086579
Publication: Research › Journal article – Annual report year: 2013

Dielektrisk spektroskopi til karakterisering af plastmaterialer
Dielektrisk spektroskopi er blevet et vigtigt redskab i arbejde med polymere materialer. Her gives en introduktion til metoden dielektrisk spektroskopi og hvilke fænomener teknikken er baseret på med fokus på analyse af polymere systemer.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Danish Technological Institute
Authors: Bøgelund, J. P. (Ekstern), Skov, A. L. (Intern), Daugaard, A. E. (Intern)
Pages: 27-28
Publication date: 2013
Main Research Area: Technical/natural sciences
Dipolar cross-linkers for PDMS networks with enhanced dielectric permittivity and low dielectric loss

Dipole grafted cross-linkers were utilized to prepare polydimethylsiloxane (PDMS) elastomers with various chain lengths and with various concentrations of functional cross-linker. The grafted cross-linkers were prepared by reaction of two alkyne-functional dipoles, 1-ethyl-4-nitrobenzene and 3-(4-((4-nitrophenyl)diazeneyl)phenoxy)-prop-1-yn-1-ylium, with a synthesized silicone compatible azide-functional cross-linker by click chemistry. The thermal, mechanical and electromechanical properties were investigated for PDMS films with 0 to 3.6 wt% of dipole-cross-linker. The relative dielectric permittivity was found to increase by ~20% at only 0.46 wt% of incorporated dipole without significant changes in the mechanical properties. Furthermore, the dielectric losses were proved to be remarkably low while the electrical breakdown strengths were high.
Electromechanically active polymer transducers: research in Europe

Smart materials and structures based on electromechanically active polymers (EAPs) represent a fast growing and stimulating field of research and development. EAPs are materials capable of changing dimensions and/or shape in response to suitable electrical stimuli. They are commonly classified in two major families: ionic EAPs (activated by an electrically induced transport of ions and/or solvent) and electronic EAPs (activated by electrostatic forces).

These polymers show interesting properties, such as sizable active strains and/or stresses in response to electrical driving, high mechanical flexibility, low density, structural simplicity, ease of processing and scalability, no acoustic noise and, in most cases, low costs.

Since many of these characteristics can also describe natural muscle tissues from an engineering standpoint, it is not surprising that EAP transducers are sometimes also referred to as 'muscle-like smart materials' or 'artificial muscles'. They are used not only to generate motion, but also to sense or harvest energy from it. In particular, EAP electromechanical transducers are studied for applications that can benefit from their 'biomimetic' characteristics, with possible usages from the micro- to the macro-scale, spanning several disciplines, such as mechatronics, robotics, automation, biotechnology and biomedical engineering, haptics, fluidics, optics and acoustics.

Currently, the EAP field is just undergoing its initial transition from academic research into commercialization, with companies starting to invest in this technology and the first products appearing on the market.

This focus issue is intentionally aimed at gathering contributions from the most influential European groups working in the EAP field. In fact, today Europe hosts the broadest EAP community worldwide. The rapid expansion of the EAP field in Europe, where it historically has strong roots, has stimulated the creation of the 'European Scientific Network for Artificial Muscles—ESNAM', entirely focused on EAPs and gathering the most active research institutes, as well as key industrial developers and end users. The ESNAM network has received financial support from the European COST (Cooperation in Science and Technology) programme (COST Action MP1003), leading to fruitful collaboration, of which some results are showcased in this issue.
This focus issue deals with a number of relevant topics on ionic and electronic EAPs. The contents, which span highly heterogeneous and cross diverse disciplines, such as physics, chemistry, material science and engineering, embrace size scales from nano to macro, and cover different areas, such as new materials, devices and applications.

This collection of papers helps elucidate, on the one hand, how heterogeneous and dynamic the EAP field is in general and, on the other hand, the state of the art of the EAP research in Europe.

We hope that this focus issue might help to stimulate future work in this emerging field of research and generate new applications.

General information
State: Published
Organisations: Center for Energy Resources Engineering, Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Queen Mary University of London, Johannes Kepler University of Linz, Linköping University, University of Cergy-Pontoise
Authors: Carpi, F. (Ekstern), Graz, I. (Ekstern), Jager, E. (Ekstern), Skov, A. L. (Intern), Vidal, F. (Ekstern)
Publication date: 2013
Main Research Area: Technical/natural sciences

Publication information
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Volume: 22
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ISSN (Print): 0964-1726
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BFI (2017): BFI-level 1
Scopus rating (2017): SJR 1.152 SNIP 1.474 CiteScore 3.38
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): SJR 1.038 SNIP 1.599 CiteScore 3.1
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.177 SNIP 1.737 CiteScore 3.03
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.232 SNIP 1.836 CiteScore 2.9
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 1.006 SNIP 1.92 CiteScore 2.61
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 0.991 SNIP 1.723 CiteScore 2.19
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 1.149 SNIP 1.941 CiteScore 2.53
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.227 SNIP 1.864
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 1.102 SNIP 1.782
BFI (2008): BFI-level 2
Scopus rating (2008): SJR 1.058 SNIP 1.63
Factors affecting surface and release properties of thin PDMS films

Polydimethylsiloxane (PDMS) elastomers are commonly used as dielectric electroactive polymers (DEAP). DEAP films are used in making actuators, generators and sensors. In the large scale manufacture of DEAP films, release of films from the substrate (carrier web) induces some defects and pre-strain in the films which affect the overall performance of the films. The current research is directed towards investigating factors affecting the peel force and release of thin, corrugated polydimethylsiloxane films used in DEAP films. It has been shown that doping the PDMS films with small quantities of perfluoroether allylamide (PFE) lowered the surface energy which could ease the release. This is further investigated together with an evaluation of the resulting change in actuator performance. The relationship between the adhesive energy, surface energy, Young’s modulus and peel force of the films is analyzed.

General information
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BFI (2015): BFI-level 1
Scopus rating (2015): SJR 0.627 SNIP 0.678 CiteScore 1.66
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Scopus rating (2014): SJR 0.604 SNIP 0.757 CiteScore 1.58
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
High dielectric permittivity elastomers from well-dispersed expanded graphite in low concentrations

The development of elastomer materials with a high dielectric permittivity has attracted increased interest over the last years due to their use in for example dielectric electroactive polymers. For this particular use, both the electrically insulating properties - as well as the mechanical properties of the elastomer - have to be tightly controlled in order not to destroy favorable elastic properties by the addition of particles. In the following, expanded graphite in low concentrations (up to 5 wt%) are investigated as a possible candidate as filler materials in very soft elastomers, which by the addition of traditional fillers in the necessary amounts would either lose their stability or their softness. Furthermore the influence of several mixing procedures on the electrical and mechanical properties is investigated. © 2013 SPIE.
Hot embossing of microstructures on addition curing polydimethylsiloxane films
The aim of this research work is to establish a hot embossing process for addition curing vinyl-terminated polydimethylsiloxane (PDMS), which are thermosetting elastomers, based on the existing and widely applied technology for thermoplasts. To our knowledge, no known technologies or processes are commercially available for embossing microstructures and submicron structures on elastomers like silicones in large scale production of films. The predominantly used technologies to make microscale components for microfluidic devices and microstructures on PDMS elastomer is (a) reaction injection molding, (b) ultraviolet lithography, and (c) photolithography. We focus on hot embossing as it is one of the simplest, most cost-effective, and time-saving methods for replicating structures for thermoplasts. Addition curing silicones are shown to possess the ability to capture and retain an imprint made on it, 10–15 min after the gel point at room temperature. This property is exploited in the hot embossing technology.

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Energy Resources Engineering
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Scopus rating (2016): SJR 0.263 SNIP 0.566 CiteScore 0.76
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Scopus rating (2015): SJR 0.262 SNIP 0.529 CiteScore 0.61
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 0.213 SNIP 0.553 CiteScore 0.6
BFI (2013): BFI-level 1
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BFI (2012): BFI-level 1
Scopus rating (2012): SJR 0.304 SNIP 0.766 CiteScore 0.75
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Scopus rating (2011): SJR 0.373 SNIP 0.546 CiteScore 0.72
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Hot-embossing of microstructures on addition-curing polydimethylsiloxane films

To our knowledge no known technologies or processes are commercially available for embossing microstructures and sub-micron structures on elastomers like silicones in large scale production of films. The predominantly used technologies to make micro-scale components for micro-fluidic devices and microstructures on PDMS elastomer are 1) reaction injection molding 2) UV lithography and 3) photolithography, which all are time-consuming and not suitable for large scale productions. A hot-embossing process to impart micro-scale corrugations on an addition curing vinyl terminated PDMS film, which is thermosetting elastomer, was established based on the existing and widely applied technology for thermoplasts. We focus on hot-embossing as it is one of the simplest, most cost-effective and time saving methods for replicating structures for thermoplasts. Addition curing silicones are shown to possess the ability to capture and retain an imprint made on it 10-15 minutes after the gel-point at room temperature. This property is exploited in the hot-embossing technology. © 2013 SPIE.

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State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Energy Resources Engineering
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Number of pages: 16
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Ionic networks derived from the protonation of dendritic amines with carboxylic acid end-functionalized PEGs

The synthesis and characterization of novel ionic networks linked by the ammonium salts of poly(propylene imine) (PPI) dendrimers of the first (PPI G1) and second (PPI G2) generation and two short bis carboxymethyl ether terminated poly(ethylene glycol)s (DiCOOH-PEG) with different molecular weights (Mn ∼ 250 and Mn ∼ 600) are reported. Likewise, an ionic network based on PPI G1 and a long ωω-dicarboxylic acid functionalized PEG (Mn ∼ 4800) were evaluated. Simpler ionic structures based on tris(2-aminoethyl)amine or hexamethylene diamine and the short DiCOOH-PEGs are also investigated. The ionic structures formed were confirmed by differential scanning calorimetry, Fourier Transform Infrared spectroscopy in the attenuated-total-reflection mode, and 1H-13C NMR spectroscopy. A comprehensive 1H NMR analysis revealed that only the primary amines of the PPI G1 dendrimer residing at the periphery take part in the ionic network formation. In the case of PPI G2, the picture is less clear. A thorough investigation of the thermal degradation of the utilized precursors and all the ionic materials prepared was additionally performed by thermogravimetric analysis. © 2012 Wiley Periodicals, Inc. J Polym Sci Part A: Polym Chem, 2013

Intelligente materialer som kunstige muskler og energi-hættere

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Energy Resources Engineering
Authors: Bahrt, F. (Intern), Daugaard, A. E. (Intern), Hvilsted, S. (Intern), Skov, A. L. (Intern)
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Organisations: Center for Energy Resources Engineering, Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Gonzalez, L. (Intern), Skov, A. L. (Intern), Hvilsted, S. (Intern)
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Molecular Rheology of Complex Fluids

The processing of polymer materials is highly governed by its rheology, and influences the properties of the final product. For example, a recurring problem is instability in extrusion that leads to imperfect plastic parts. The ability to predict and control the rheological behavior of polymer fluids as a function of molecular chemistry has attracted a long history of collaboration between industry and academia. In industrial polymer processes, there is usually a combination of both shear and extensional flows. In some processing operations such as blow molding and fiber spinning, extensional flow is the dominant type of deformation. The polymer molecules experience a significant amount of chain orientation and stretching during these processes. Shear rheology measured by conventional shear rheometers is good at describing chain orientation, whereas extensional rheology gives a good way of inducing chain stretching. Accurate and reliable stress–strain measurements of extensional flow play a crucial role in the understanding of non–linear rheological properties of polymers. However, the non–linear extensional rheology has not been extensively studied. It is known that
the rheology of polymer melts is highly sensitive to molecular architecture, but the precise connection between architecture and non-linear rheology is still not fully understood. For example, linear polymer melts have the simplest architecture, but the possible existence of a qualitative difference on extensional steady-state viscosity between melts and solutions is still an open question. Branched polymer melts have more complex molecular structures. A stress maximum during the start-up of uniaxial extensional flow was reported in 1979 for a low-density polyethylene (LDPE) melt. Subsequently observations of a steady stress following a stress maximum were reported for two LDPE melts. However the rheological significance of the stress maximum as well as the existence of steady flow conditions following the maximum is still a matter of some debate. This thesis focuses on the experimental study of extensional rheology of linear and branched polymer melts. We report the stress–strain measurements in extensional flows using a unique Filament Stretching Rheometer (FSR) in controlled strain rate mode and controlled stress mode. Extensional flow is difficult to measure reliably in Laboratory circumstances. In this thesis we first present an updated control scheme that allows us to control the kinematics of polymer melts in an FSR, which is the foundation of our experimental work. Next we investigate four categories of polymer melts from the simplest system to the most complicated system, including 1) the narrow molar mass distribution (NMMD) linear polystyrene melts and solutions; 2) the bidisperse and polydisperse linear polystyrene melts; 3) the NMMD branched polystyrene melts; and 4) the polydisperse branched polyethylene melts. The experimental results are also compared with some developing theoretical models. Finally, to ensure the experimental data is accurate, the measurements from the FSR are compared with the data from some other extensional rheometers as well.

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Energy Resources Engineering, Technical University of Denmark
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Novel cross-linkers for PDMS networks for controlled and well distributed grafting of functionalities by click chemistry
An azide-containing, trifunctional vinyl cross-linker for silicone networks has been synthesized. The cross-linker has through Cu(i) catalyzed 1,3-cycloaddition been reacted with six different alkyne-containing chemical groups which each possess a particular functionality. The functional cross-linkers have been utilized to prepare novel polydimethylsiloxane (PDMS) networks. All functional cross-linkers were successfully incorporated into the networks and were demonstrated to be well distributed within the PDMS films. This was substantiated by fluorescence microscopy of a film prepared with the 4-methyl-umbelliferone containing cross-linker. TGA showed that a ferrocene functionality increased the thermal degradation temperature of PDMS. It was furthermore shown that the incorporation of only 0.25 wt% of the push-pull dipole, ethynyl-4-nitrobenzene, increased the dielectric permittivity of PDMS by 35%. The contact angle of PDMS films was increased from 108° to 116° by the introduction of a small poly(pentafluorostyrene) chain. Finally, 17α-ethynyl-1,3,5(10)-estratriene-3,17β-diol and 1-ethynyl-3,5-bis(trifluoromethyl)benzene were incorporated as examples of other functional groups. © 2013 The Royal Society of Chemistry.

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Energy Resources Engineering
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Novel silicone compatible cross-linkers for controlled functionalization of PDMS networks

Polydimethylsiloxane (PDMS) elastomers are excellent materials for dielectric electroactive polymers (DEAPs) due to their high efficiency and fast response. PDMS suffers, however, from low dielectric permittivity and high voltages are therefore required when the material is used for DEAP actuators. In order to improve the dielectric properties of PDMS a novel system is developed where push-pull dipoles are grafted to a new silicone compatible cross-linker. The grafted cross-linkers are prepared by reaction of two different push-pull dipole alkynes as well as a fluorescent alkyne with the new azide-functional cross-linker by click chemistry. The dipole cross-linkers are used to prepare PDMS elastomers of various chain lengths providing different network densities. The functionalized cross-linkers are incorporated successfully into the networks and are well distributed as determined by the fluorescent functional cross-linker and fluorescence microscopy. The thermal, mechanical and electro-mechanical properties of PDMS elastomers of 0 wt% to 3.6 wt% of push-pull dipole cross-linker are investigated. An increase in the dielectric permittivity of 19 % at only 0.46 wt% of pure push-pull dipole is observed. Furthermore, the dielectric losses are found to be very low while the electrical breakdown strengths are high and adequate for DEAP applications. © 2013 SPIE.
Novel silicone elastomer formulations for DEAPs

We demonstrate that the force output and work density of polydimethylsiloxane (PDMS) based dielectric elastomer transducers can be significantly enhanced by the addition of high permittivity titanium dioxide nanoparticles which was also shown by Stoyanov et al.[1] for pre-stretched elastomers and by Carpi et al for RTV silicones[2]. Furthermore the elastomer matrix is optimized to give very high breakdown strengths. We obtain an increase in the dielectric permittivity of a factor of approximately 2 with a loading of 12% TiO2 particles compared to the pure modified silicone elastomer with breakdown strengths remaining more or less unaffected by the loading of TiO2 particles. Breakdown strengths were measured in the range from approximately 80-150 V/μm with averages of the order of 120-130 V/μm for the modified silicone elastomer with loadings ranging from 0 to 12%.

General information

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Organisations: Center for Energy Resources Engineering, Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Danfoss PolyPower A/S
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Reinforced poly(propylene oxide)- a very soft and extensible dielectric electroactive polymer

Poly(propylene oxide) (PPO), a novel soft elastomeric material, and its composites were investigated as a new dielectric electroactive polymer (EAP). The PPO networks were obtained from thiol-ene chemistry by photochemical crosslinking of 1,1-diallyl PPO with a tetra-functional thiol. The elastomer was reinforced with hexamethylenedisilazane treated fumed silica to improve the mechanical properties of PPO. The mechanical properties of PPO and composites thereof were investigated by shear rheology and stress–strain measurements. It was found that incorporation of silica particles improved the stability of the otherwise mechanically weak pure PPO network. Dielectric spectroscopy revealed high relative dielectric permittivity of PPO at 103 Hz of 5.6. The relative permittivity was decreased slightly upon addition of fillers, but remained higher than the commonly used acrylic EAP material VHB4910. The electromechanical actuation performance of both PPO and its composites showed properties as good as VHB4910 and a lower viscous loss.
Stimuli-Adaptable Materials

The work presented in this Thesis deals with the development of a stimuli-adaptable polymer material based on the UV-induced dimerisation of cinnamic acid and its derivatives. It is in the nature of an adhesive to adhere very well to its substrate and therefore problems can arise upon removal of the adhesive. This is also known from skin adhesives where it is very undesirable to cause damage to the skin. The overall idea of this project was to resolve this problem by developing a material which could switch between an adhesive and a non-adhesive state. Switchable adhesion is known in the literature but the presented work has a new approach to the field by basing itself on the idea of developing a network into which a photo-active polymer is mixed and which function as an adhesive. Upon irradiation with UV-light for a short time a non-adhering inter-penetrating network material would be formed. Two simple models for the extent of reaction for the system are presented and show that the timescale for the reaction is minutes to hours. This was further investigated with IR-spectroscopy and UV-absorbance spectroscopy. UV-spectroscopy confirmed that a change in the material occurs upon irradiation with UV and that the reaction time is in the range of minutes. A number of polymer derivatives with cinnamic acid or cinnamylidene acetic acid were prepared and the material properties of these were studied before and after irradiation with UV-light. For the cinnamylidene acetic acid derivatised polymers a macroscopic change was observed upon dissolution of the irradiated compound. The irradiated polymers formed threads or fibres when exposed to water while the unexposed polymers dissolved as a hygroscopic powder. Cinnamic acid derivatised poly(ethylene glycol) (PEG) was studied in detail and three different polymers were derivatised, namely a 4-armed star PEG (Mn = 2000 g/mol), a short linear PEG (Mn = 1000 g/mol) and a long linear PEG (Mn = 4000 g/mol). The derivatised polymers were mixed to create three different photo-active materials and these were investigated with rheology before and after irradiation with UV-light for one hour. It was observed that the largest change occurred for the system consisting solely of the cinnamic acid derivatised 4-armed star PEG. The development of the material properties of this material was studied in details by exposing the compound to UV-radiation for up to 120 h and determining the rheological properties after the exposure. It was found that approx. 24 hours was needed to form a manageable film and that approx. 70 hours were needed to obtain stable rheological properties. The exposure times were larger than expected but it was assumed that this would be resolved by preparation of the inter-penetrating network materials. A number of inter-penetrating network materials where both the permanent Network as well as the switching segment was made up of PEG-polymers were prepared. It was found that for a material with relatively long chains in both permanent Network (Mn = 10000 g/mol) and switching segment (Mn = 15000 g/mol) no changes occurred upon exposure to UV-light. It is expected that this is because Mn of polymers is above the molecular entanglement weight. Inter-penetrating network materials with Mn = 4000 g/mol in the permanent network were prepared. Initially a linear photo-active PEG was mixed into the material. This material exhibited a decrease in the values for G and G after irradiation with UV-light for 30 minutes. The reason for the decrease in the two rheological parameters is unclear.
but the change encouraged further work with this type of systems. Two inter-penetrating network materials with the star-shaped cinnamic acid derivatised PEG as switching segment were prepared and irradiated with UV-light for 72 hours. The network with $r = 0.75$ in the permanent matrix proved the expectations by clearly showing a solvent effect when the photo-active polymer was introduced into the permanent network. In addition a significant increase of $G$ and $G'$ was observed after 72 h of irradiation with UV-light proving the formation of a second network consisting of the photo-active polymers. A network with $r = 0.5$ in the permanent matrix was also investigated but gave very different results due to the lower value for $r$. The applied $r$-value is significantly closer to the critical rc-value and thus resulted in problems with the film formation. The data showed that the secondary network dominates the rheological properties of this network. A material with shorter chains in the permanent network (Mn = 1000 g/mol) was also investigated but showed no change after irradiation with UV-light for 15 minutes. This is related to the stiffness of permanent matrix. First steps to creating an inter-penetrating network with two different polymers were taken by incorporating the cinnamic acid derivatised PEG-stars into a poly(propylene oxide) network. However exposure to UV-light did not result in any changes of the material properties. It was also tested if the photo-active PEG could be incorporated into a poly(dimethyl siloxane) network, but the addition of the photo-active PEG resulted in complete hindrance of the cross-linking of the poly(dimethyl siloxane). A number of problems were identified throughout the work, primarily concerning the mismatch between the expected exposure time needed to induce changes in the materials and the exposure time observed experimentally. This can partially be explained by mobility of the polymers and concentration of the photo-active cinnamic acid. Studies presented in the literature show that the position of the cinnamic acid groups is important for the dimerisation to occur. The nature of polymers makes encounters between end groups less likely and this affects the dimerisation. Furthermore, an NMR-study showed formation of the cis-isomer of cinnamic acid. The isomerisation of cinnamic acid only occurs if dimerisation is hindered. This underlines that the circumstances are not ideal for dimerisation.

General information
State: Published
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Transient overshoot extensional rheology of long-chain branched polyethylenes: Experimental and numerical comparisons between filament stretching and cross-slot flow
This work analyses the high-strain extensional behavior of long-chain branched polyethylenes, employing two novel extensional rheometer devices, the filament stretching rheometer and the cross-slot extensional rheometer. The filament stretching rheometer uses an active feedback loop to control the imposed strain rate on a filament, allowing Hencky strains of around 7 to be reached. The cross-slot extensional rheometer uses optical birefringence patterns to determine the steady-state extensional viscosity from planar stagnation point flow. The two methods probe different strain-rate regimes and in this paper we demonstrate the agreement when the operating regimes overlap and explore the steady-state extensional viscosity in the full strain-rate regime that these two complimentary techniques offer. For long-chain branched materials, the cross-slot birefringence images show a double cusp pattern around the outflow centre line (named W-cusps). Using constitutive modeling of the observed transient overshoot in extension seen in the filament stretching rheometer and using finite element simulations we show that the overshoot explains the W-cusps seen in the cross-slot extensional rheometer, further confirming the agreement between the two experimental techniques. © 2013 The Society of Rheology.

General information
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Organisations: Department of Chemical and Biochemical Engineering, Durham University, University of Cambridge, University of Leeds
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Main Research Area: Technical/natural sciences
Adhesion Between Poly(dimethylsiloxane) Layers

Different adhesion methods of poly(dimethylsiloxane) (PDMS) layers were studied with respect to adhesional force and the resulting rheology of the two-layered PDMS films were investigated. The role of adhesion between PDMS layers on the performances of two-layer structures was studied with peel strength test and by SEM pictures. The rheology of the double-layered compared to the monolayer films changed in some cases which indicates that the adhesion process needs to be carefully introduced in order not to alter the final properties.

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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Yu, L. (Intern), Daugaard, A. E. (Intern), Skov, A. L. (Intern)
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Are Entangled Polymer Melts Different From Solutions?

The possible existence of a qualitative difference on extensional steady state viscosity between polymer melts and polymer solutions is still an open question. Recent experiments [1-4] showed the extensional viscosity of both polymer melts and solutions decayed as a function of strain rate with an exponent of -0.5. When the strain rate became higher than the order of inverse Rouse time, the polymer solutions showed an upturn [1, 4]. However, in the same regime for polymer melts, the experiments were contrary: some of the experiments showed an upturn [4, 5], while others did not [2, 3].

In order to further investigate the extensional steady state viscosity of polymer melts, we carefully synthesized two monodisperse polystyrenes with molar masses of 248 and 484 kg/mole. The start-up and steady uniaxial elongational viscosity have been measured for the two melts using a filament stretching rheometer. We then compared the measurements with the bi-disperse polystyrene melts made from the above two polymers. The influence and sensitivity of impurities were studied by adding different percentages of 484k into 248k polystyrene melt. Furthermore a polydisperse polystyrene with weight average molecular weight 230 kg/mole was also measured for comparison. Possible reasons for the differences shown in the previously mentioned experiments are discussed.

General information
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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Department of Micro- and Nanotechnology, Amphiphilic Polymers in Biological Sensing, Department of Mechanical Engineering, Manufacturing Engineering
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Bimodal Networks as Candidates for Electroactive Polymers

An alternative network formulation method was adopted in order to obtain a different type of silicone based elastomeric systems - the so-called bimodal networks - using two vinyl-terminated polydimethyl siloxanes (PDMS) of different molecular weight, a labelled crosslinker (3 or 4-functional), and a platinum-catalyzed hydrosilylation reaction between the three reactants. The crosslinking reaction was initiated by mixing different amounts of short PDMS chains and long PDMS chains with the stoichiometric amount of the crosslinker in a series of mixing schemes. We visualize how the short chains organize themselves between the long chains and show how this leads to unexpectedly good properties for DEAP purposes due both to the low extensibility of the short chains that attach strongly the long chains and to the extensibility of the last ones that retards the rupture process.

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Development of new PDMS based materials for dielectric electroactive polymers (DEAPs) as actuators and generators

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Dielectric electroactive elastomers: from eye lid control to wave power generation with one single material

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Authors: Skov, A. L. (Intern)
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Main Research Area: Technical/natural sciences

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Fremstilling af dielektriske elektroaktive polymerer i stor skala

Interessen for dielektriske elektroaktive polymerfilm er de seneste år steget markant. For overhovedet at kunne producere de ønskede mængder til en konkurrencedygtig pris er optimering og effektivisering af produktionen af disse film et krav for teknologiens overlevelse.

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ISI indexed (2011): ISI indexed no
Web of Science (2007): Indexed yes
Web of Science (2004): Indexed yes
Original language: Danish
Links:
http://techmedia.swiflet.com/tm/dak/60/1/?api=togglearchive()%3b
Source: dtu
Source-ID: u::6956
Publication: Research › Journal article – Annual report year: 2013

Hot-embossing of microstructures on addition-curing PDMS

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Vudayagiri, S. (Intern), Yu, L. (Intern), Hassouneh, S. S. (Intern), Skov, A. L. (Intern)
Publication date: 2012
Main Research Area: Technical/natural sciences
Electronic versions:
Annual Polymer day-2012-Sindhu Vudayagiri.pdf
Source: dtu
Source-ID: u::5518
Publication: Research › Conference abstract for conference – Annual report year: 2012

Influence of micro- and nanofillers on electro-mechanical performance of silicone EAPs

The effect of different fillers on the mechanical and dielectric properties of soft elastomers has been investigated. It was found that the addition of a small amount of silica fillers would increase the Young’s modulus significantly but not simultaneously increase the tear strength sufficiently for processing as thin films. Addition of nanoclay and barium titanate nanoparticles to the soft elastomers was shown to be very favorable for the enhancement of the dielectric properties without increasing the Young’s modulus significantly and could be used for DEAP material in e.g. microprocessing

where the tear strength is not crucial for processing. However, for elastomer film processing it is suggested that a combination of the nanoclay or barium titanate with either silica particles or bimodal networks would give the right tear strength together with the desired increased dielectric permittivity.

General information
Investigation of the properties of fully reacted unstoichiometric polydimethylsiloxane networks and their extracted network fractions

We investigated the linear dynamic response of a series of fully reacted unstoichiometric polydimethylsiloxane (PDMS) networks and of the two corresponding network fractions namely the sol and the washed network. The sol and the washed network were separated by a simple extraction process. This way it was possible to obtain rheological data from the washed network without interference from the sol fraction, and furthermore from the sol fraction without interference from the elastic washed network. When the stoichiometry increased towards perfectly reacted networks and beyond we observed harder networks both qualitatively and by rheology and the properties of the two fractions became more and more different. At the gel point, the sol fraction and the washed network have more or less identical properties which our data also shows. The storage and loss moduli, $G'$ and $G''$, were analysed with the gel equation as proposed by Winter and Chambon (1986; 1987). We observed that one of the investigated samples which before the swelling experiment did not show any elastic response gave an elastic washed network after swelling, this was verified by analysis with the gel equation. We also calculated the weight fraction of the sol fraction by using the theory by Villar et al. (1996) and compared these with experimentally found values.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Frankær, S. M. G. (Intern), Jensen, M. K. (Intern), Bejenariu, A. G. (Intern), Skov, A. L. (Intern)
Pages: 559-567
Publication date: 2012
Main Research Area: Technical/natural sciences

Publication information
Journal: Rheologica Acta
Volume: 51
Issue number: 6
ISSN (Print): 0035-4511
Ratings:
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 1.9 SJR 0.704 SNIP 1.063
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 1.9 SJR 0.634 SNIP 1.026
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 0.876 SNIP 1.272 CiteScore 2.09
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 0.725 SNIP 1.181 CiteScore 1.72
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 0.877 SNIP 1.38 CiteScore 2.09
ISI indexed (2013): ISI indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 0.898 SNIP 1.36 CiteScore 1.8
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): SJR 1.292 SNIP 1.397 CiteScore 2.22
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Lamination of PDMS films by different methods

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Hassouneh, S. S. (Intern), Yu, L. (Intern), Skov, A. L. (Intern)
Publication date: 2012
Event: Abstract from 49th Nordic Polymer Days 2012, Copenhagen, Denmark.
Main Research Area: Technical/natural sciences

Electronic versions:
Abstract_NPD_Suzan.pdf
Source: dtu
Source-ID: u::3780
Publication: Research › Conference abstract for conference – Annual report year: 2012

Large amplitude oscillatory measurements as mechanical characterization methods for soft elastomers

Mechanical characterization of soft elastomers is usually done either by traditional shear rheometry in the linear viscoelastic (LVE) regime (i.e. low strains) or by extensional rheology in the nonlinear regime. However, in many commercially available rheometers for nonlinear extensions the measurements rely on certain assumptions such as a predefined shape alteration and are very hard to perform on soft elastomers in most cases. The LVE data provides information on important parameters for DEAP purposes such as the Young’s modulus and the tendency to viscous dissipation (at low strains only) but provides no information on the strain hardening or softening effects at larger strains, and the mechanical breakdown strength. Therefore it is obvious that LVE can not be used as the single mechanical characterization tool in large strain applications. We show how the data set of LVE, and large amplitude oscillating elongation (LAOE) and planar elongation make the ideal set of experiments to evaluate the mechanical performance of DEAPs. We evaluate the mechanical performance of several soft elastomers applicable for DEAP purposes such as...
poly(propyleneoxide) (PPO) networks\textsuperscript{3, 4} and traditional unfilled silicone (PDMS) networks\textsuperscript{5}.

**General information**

State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Skov, A. L. (Intern)
Number of pages: 8
Pages: 83401C
Publication date: 2012
Conference: Electroactive Polymer Actuators and Devices 2012, San Diego, CA, United States, 12/03/2012 - 12/03/2012
Main Research Area: Technical/natural sciences

**Publication information**

Journal: Proceedings of SPIE, the International Society for Optical Engineering
Volume: 8340
ISSN (Print): 0277-786X
Ratings:

BFI (2018): BFI-level 1
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 0.43 SJR 0.243 SNIP 0.289
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 0.42 SNIP 0.258 SJR 0.226
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 0.212 SNIP 0.239 CiteScore 0.3
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 0.217 SNIP 0.249 CiteScore 0.3
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 0.234 SNIP 0.273 CiteScore 0.26
ISI indexed (2013): ISI indexed no
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 0.219 SNIP 0.275 CiteScore 0.27
ISI indexed (2012): ISI indexed no
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 0.217 SNIP 0.286 CiteScore 0.31
ISI indexed (2011): ISI indexed no
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 0.233 SNIP 0.277
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 0.236 SNIP 0.312
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 0.245 SNIP 0.3
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 0.247 SNIP 0.376
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 0.323 SNIP 0.676
Scopus rating (2005): SJR 0.162 SNIP 0.372
Web of Science (2004): Indexed yes
Web of Science (2002): Indexed yes

Original language: English
Elastomers, Elasticity, Loss modulus, Storage modulus, Material functions, Mechanical breakdown
Electronic versions:
lao_1_.pdf
Low moduli elastomers with low viscous dissipation
A controlled reaction schema for addition curing silicones leads to both significantly lower elastic modulus and lower viscous dissipation than for the chemically identical network prepared by the traditional reaction schema.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Bejenariu, A. G. (Intern), Yu, L. (Intern), Skov, A. L. (Intern)
Pages: 3917-3923
Publication date: 2012
Main Research Area: Technical/natural sciences

Publication information
Journal: Soft Matter
Volume: 8
Issue number: 14
ISSN (Print): 1744-683X
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): SNIP 1.227 SJR 1.493 CiteScore 3.81
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 3.7 SJR 1.657 SNIP 1.173
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.634 SNIP 1.271 CiteScore 3.97
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.742 SNIP 1.243 CiteScore 4.11
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 1.748 SNIP 1.2 CiteScore 4.2
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 1.901 SNIP 1.144 CiteScore 3.96
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 2.007 SNIP 1.307 CiteScore 4.56
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 2.181 SNIP 1.375
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 2.527 SNIP 1.53
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 2.596 SNIP 1.373
Web of Science (2008): Indexed yes
Monolithic structures of DEAP films

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Hassouneh, S. S. (Intern), Skov, A. L. (Intern)
Publication date: 2012
Main Research Area: Technical/natural sciences
Electronic versions:
Abstract_APD_Suzan_05102012.pdf

New Cross-linkers for PDMS Networks

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Energy Resources Engineering
Authors: Bahrt, F. (Intern), Daugaard, A. E. (Intern), Hvilsted, S. (Intern), Skov, A. L. (Intern)
Number of pages: 1
Publication date: 2012
Event: Abstract from 49th Nordic Polymer Days 2012, Copenhagen, Denmark.
Main Research Area: Technical/natural sciences
Electronic versions:
Abstract Frederikke Bahrt.pdf

Novel silicone compatible cross-linkers for controlled and well distributed functionalization of PDMS networks

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Center for Energy Resources Engineering
Authors: Bahrt, F. (Intern), Daugaard, A. E. (Intern), Hvilsted, S. (Intern), Skov, A. L. (Intern)
Number of pages: 1
Planar elongation of soft elastomeric networks

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Goswami, K. (Intern), Román Marín, J. M. (Intern), Skov, A. L. (Intern)
Publication date: 2012
Event: Abstract from 49th Nordic Polymer Days 2012, Copenhagen, Denmark.
Main Research Area: Technical/natural sciences
Electronic versions:
kaustav_goswami_abstract_npd_2012.pdf
Links:
http://www.npd2012.dk/
Source: dtu
Source-ID: u::3781
Publication: Research › Conference abstract for conference – Annual report year: 2012

Preparation and Characterization of Anisotropic Films by Curing Hard Silicone Microspheres with Soft Silicone

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Gonzalez, L. (Intern), Skov, A. L. (Intern)
Number of pages: 1
Publication date: 2012
Event: Abstract from 49th Nordic Polymer Days 2012, Copenhagen, Denmark.
Main Research Area: Technical/natural sciences
Electronic versions:
Abstract_ligo_NPD_2012.pdf
Links:
http://www.npd2012.dk/
Source: dtu
Source-ID: u::3739
Publication: Research › Conference abstract for conference – Annual report year: 2012

Role of stoma effluent on adhesion between skin-friendly adhesives and skin

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Coloplast Danmark A/S
Authors: Ravni, C. (Ekstern), Hansen, K. (Ekstern), Daugaard, A. E. (Intern), Skov, A. L. (Intern)
Number of pages: 1
Publication date: 2012
Event: Abstract from 49th Nordic Polymer Days 2012, Copenhagen, Denmark.
Main Research Area: Technical/natural sciences
Electronic versions:
Abstract_Clemence.pdf
Links:
http://www.npd2012.dk/
Source: dtu
Source-ID: u::3736
Publication: Research › Conference abstract for conference – Annual report year: 2012
Stress relaxation and reversed flow of low-density polyethylene melts following uniaxial extension.
The extensional dynamics of two low-density polyethylene melts Lupolen 3020D and Lupolen 1840D, both showing a stress overshoot in start-up of uniaxial extension [Rasmussen, H. K., J. K. Nielsen, A. Bach, and O.Hassager, 'Viscosity overshoot in the start-up of uniaxial elongation of low density polyethylene melts,' J. Rheol. 49(2), 369-381 (2005)], have been further investigated in stress relaxation and reversed flow. After the overshoot, the stress relaxation has a remarkably faster decrease of the transient stress than when the relaxation is initiated before the overshoot. The measurements from the reversed flow also show that the melts appear less elastic after the overshoot. Multi mode versions of the pom-pom and interchain pressure model fit the data quantitatively up to the stress maximum, but neither model captures the qualitative behavior after the maximum.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Department of Mechanical Engineering, Manufacturing Engineering
Authors: Huang, Q. (Intern), Rasmussen, H. K. (Intern), Skov, A. L. (Intern), Hassager, O. (Intern)
Pages: 1535-1554
Publication date: 2012
Main Research Area: Technical/natural sciences

Publication information
Journal: Journal of Rheology
Volume: 56
Issue number: 6
ISSN (Print): 0148-6055
Ratings:
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 3.17 SJR 1.326 SNIP 1.564
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 3.1 SJR 1.438 SNIP 1.523
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 1.43 SNIP 1.531 CiteScore 2.67
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 1.696 SNIP 1.565 CiteScore 3.29
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 1.319 SNIP 1.63 CiteScore 2.96
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 1.359 SNIP 1.617 CiteScore 2.72
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): SJR 1.96 SNIP 1.839 CiteScore 3.34
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 1.831 SNIP 1.495
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Scopus rating (2009): SJR 1.796 SNIP 1.539
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.86 SNIP 2.468
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 1.753 SNIP 1.777
Scopus rating (2006): SJR 1.472 SNIP 1.706
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 1.921 SNIP 1.64
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 1.72 SNIP 1.812
Scopus rating (2003): SJR 1.865 SNIP 1.762
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 1.882 SNIP 1.994
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 2.622 SNIP 1.952
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 1.729 SNIP 1.794
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 1.899 SNIP 2.004
Original language: English
Electronic versions:
JoR.pdf
Links:
http://journalofrheology.org/resource/1/jorhd2/v56/i6/p1535_s1

Bibliographical note
© 2012 by The Society of Rheology, Inc.
Source: du
Source-ID: n::oai:DTIC-ART:ebsco/370362273::20217
Publication: Research - peer-review › Journal article – Annual report year: 2012

Surface and release properties of PDMS films

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Vudayagiri, S. (Intern), Skov, A. L. (Intern)
Publication date: 2012
Event: Abstract from 49th Nordic Polymer Days 2012, Copenhagen, Denmark.
Main Research Area: Technical/natural sciences
Electronic versions:
sindhu_vudayagin_abstract_npd2012.pdf
Source: du
Source-ID: u::5517
Publication: Research › Conference abstract for conference – Annual report year: 2012

Transient Overshoot Extensional Rheology: Experimental and Numerical Comparisons
The true steady state value for the extensional stress growth is still an open topic in the field of polymer melts. Shear and extensional flows are often used to fit the various parameters in constitutive models. Extensional flow is much more effective than shear flow at orientating and stretching polymer chains, and not having steady state values for the extensional viscosity makes understanding and modelling molecular rheology in extensional flow challenging. Here we present a comparison between three extensional rheometers: the Sentmanat extensional rheometer (SER), the filament stretching rheometer (FSR) and the cross-slot extensional rheometer (CSER). The first two are uni-axial stretching rheometers and the third is a planar extensional rheometer. The FSR and CSER are capable of achieving steady state flows, although in different strain-rate regimes.
The SER has been a widely adopted tool as it conveniently adapts to a standard shear rheometer. However, the SER is only capable of reaching Hencky strains up to 4. We compare this to the transient measurements of the FSR which uses an active feedback system to control sample necking and is capable of Hencky strains of around 7. We then compare the steady state measurements from the FSR to the predictions of the CSER, which measures the viscosity using optical flow induced birefringence techniques. The FSR is capable of measuring a steady state for strain rates less than 0.5/s and the CSER for strain rates typically in the range of 0.1/s to 10/s. Although we are comparing uni-axial to planar flow, these flows produce the same polymer deformation and hence stress in
the strain hardening regime. The techniques show a good quantitative agreement where the two experimental windows overlap.

In comparing the FSR to the CSER we are able to explain an observed feature of the optical birefringence stress patterns, known as W-cusping. It has been shown that the Pompom model, a constitutive model for branched polymers, does not capture this feature. Here we show the cause of the observed W-cusps is due to the presence of a transient overshoot in the extensional viscosity. We present a modified Pompom model capable of capturing these overshoots and show finite element simulations of the cross-slot flow using this model. In doing this we show how the overshoot causes the W-cusp and how the shape of the cusps is dependent upon the difference between the maximum extensional viscosity and the steady state value.

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre, Department of Mechanical Engineering, Manufacturing Engineering, Durham University, University of Leeds, Universite Catholique de Louvain
Authors: Hoyle, D. (Ekstern), Huang, Q. (Intern), Auhl, D. (Ekstern), Rasmussen, H. K. (Intern), Skov, A. L. (Intern), Harlen, O. (Ekstern), Hassager, O. (Intern), McLeish, T. (Ekstern)
Publication date: 2012
Event: Abstract from 16th International Congress on Rheology, Lisbon, Portugal.
Main Research Area: Technical/natural sciences
Electronic versions:
ICR2012-p48.pdf

Bibliographical note
Oral Presentation.
Source: dtu
Source-ID: u::5236
Publication: Research › Conference abstract for conference – Annual report year: 2012

Udvikling af materialer til kunstige muskler
Ultrapræsterende højtalere, bølgeenergi-generatorer med energiproduktion i enorme mængder, aktuatorer uden standby-energiforbrug og bløde deformerbare kunstige muskler kan måske snart realiseres vha. dielektriske elektroaktive polymerer (DEAP).

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Bahrt, F. (Intern), Daugaard, A. E. (Intern), Hvilsted, S. (Intern), Skov, A. L. (Intern)
Pages: 48-51
Publication date: 2012
Main Research Area: Technical/natural sciences

Publication information
Journal: Dansk Kemi
Volume: 93
Issue number: 9
ISSN (Print): 0011-6335
Ratings:
ISI indexed (2013): ISI indexed no
ISI indexed (2012): ISI indexed no
ISI indexed (2011): ISI indexed no
Web of Science (2007): Indexed yes
Web of Science (2004): Indexed yes
Original language: Danish
Electronic versions:
Frederikke Bahrt Madsen.pdf
Links:
http://techmedia.swiflet.com/tm/dak/56/1/
Source: dtu
Source-ID: u::5707
Publication: Research › Journal article – Annual report year: 2012
Unimodal and bimodal networks performance as dielectric electroactive polymers (DEAPs)

General information
State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Bejenariu, A. G. (Intern), Daugaard, A. E. (Intern), Skov, A. L. (Intern)
Publication date: 2012
Event: Abstract from 49th Nordic Polymer Days 2012, Copenhagen, Denmark.
Main Research Area: Technical/natural sciences
Electronic versions:
Abstract_NPD_31May2012KBH_1_.pdf
Source: dtu
Source-ID: u::3655
Publication: Research › Conference abstract for conference – Annual report year: 2012

A rheological study on bimodal networks applied as electroactive polymers

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering
Authors: Bejenariu, A. G. (Intern), Skov, A. L. (Intern)
Publication date: 2011
Main Research Area: Technical/natural sciences
Source: orbit
Source-ID: 279082
Publication: Research › Conference abstract for conference – Annual report year: 2011

Cinnamic Acid Derivatised Poly(Ethylene Glycol) as a Bioinspired UV-Adaptable Material

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, CHEC Research Centre, Technical University of Denmark
Publication date: 2011
Main Research Area: Technical/natural sciences
Electronic versions:
Abstract_SarahMariaFrankaer.pdf
Source: orbit
Source-ID: 278470
Publication: Research › Conference abstract for conference – Annual report year: 2011

Experimental evaluation of the pseudotime principle for nonisothermal polymer flows
We have applied a series of start-up of uniaxial extensions to very high strain followed by stress relaxation. A potential temperature change was applied during the stress relaxation. We used two thermorheological simple polymers; a linear polystyrene and a branched low density polyethylene. Experiments performed with temperature changes during the stress relaxation were shown to be identical with isothermal ones in the “pseudotime”, within the accuracy of the experiments. This verifies that the pseudotime approach seems to be the general basis for nonisothermal microstructural modeling for flow of polymers. The pseudotime is given as $\xi(t) = \int_{t_0}^{t} \frac{1}{aT(T(t))} dT$, where $aT$ are the well established time-temperature superposition shift factors, calculated from the past temperatures (at time $t_0$) in a particle path.

General information
State: Published
Organisations: Manufacturing Engineering, Department of Mechanical Engineering, The Danish Polymer Centre, Department of Chemical and Biochemical Engineering
Authors: Yu, K. (Intern), Rasmussen, H. K. (Intern), Skov, A. L. (Intern)
Pages: 1059-1067
Publication date: 2011
Main Research Area: Technical/natural sciences

Publication information
Influence of micro- and nanofillers on DEAP performance

The formulation of dielectric electroactive polymers (DEAP) is a challenging task as the elastomer part of DEAP is a multi-component system with strict requirements to the electromechanical performance. In traditional formulated products such as paints there is a huge variety of filler materials available and the filler particles are used both to lower the price and to increase the performance of the material. In DEAP there is a strong interplay between the electrical and mechanical properties as one property may be improved and another worsened when changing the formulation so in many cases optimization of the properties ends in a trade-off of some kind. In this study we focus on the addition of well-known commercially available filler particles and evaluate their performance in elastomers applied for DEAP since the price of the fillers also have to be taken into account when formulating. The effect of particle size and particle size distribution on the electrical and viscoelastic properties is investigated. Both nano- and microscale particles of different compositions are mixed with a platinum cured silicone elastomer matrix [1] in different combinations, and the dielectric and viscoelastic properties are measured. The dielectric properties are improved mainly by decreasing the particle size as was also shown by Kofod et al [2] for acrylic networks whereas the elastic properties are influenced in a less trivial way. The filler particles increase the storage modulus and make the films easier to handle but the formulation of the ‘raw’ silicone matrix has to be modified as the filler particles in some instances interact with the polymers and hence require a changed stoichiometry to ensure complete reaction of both reactive species in the silicone matrix.

General information

State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering
Authors: Skov, A. L. (Intern), Bejenariu, A. G. (Intern), Daugaard, A. E. (Intern)
Publication date: 2011
Event: Abstract from 6th World Congress on Biomimetics, Artificial Muscles and Nano-Bio, Cergy-Pontoise, France.
Main Research Area: Technical/natural sciences
Electronic versions:
anneladegaardskov_biomimetics2011-2.pdf
Links:
http://biomimetics2011.u-cergy.fr/

Influence of micro- and nanofillers on electro-mechanical performance of silicone EAPs

A well-known technique to improve the tear strength of rubbers is to add micro- or nanofillers. The effects of different micro- and nanofillers on the electro-mechanical behavior of silicone based dielectric elastomer are studied in this work. Several types of fillers and varied loadings are used to increase the overall permittivity of the silicone networks. The effect of particle size and particle size distribution on the electrical and viscoelastic properties is investigated. The aim of the paper is to find the right filler and the appropriate concentration for a suitable EAP material, such that an optimized formulation schema can be proposed.

General information

State: Published
Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Bejenariu, A. G. (Intern), Daugaard, A. E. (Intern), Skov, A. L. (Intern)
Publication date: 2011
Event: Abstract from 1st International conference on Electromechanically Active Polymer (EAP) transducers & artificial muscles, Pisa, Italy.
Main Research Area: Technical/natural sciences
Source: dtu
Source-ID: u::3652
Publication: Research › Conference abstract for conference – Annual report year: 2012

New Cross-linkers for PDMS Networks

General information
New elastomeric silicone based networks applicable as electroactive systems

Commercial elastomer materials are available for dielectric electroactive polymer (DEAP) purposes but the applied commercial elastomers have not been developed with the specific application in mind. It is therefore obvious that optimization of the elastomer material should be possible. In this study we focus on optimization of the mechanical properties of the elastomer and show that it is possible to lower the elastic modulus and still not compromise the other required mechanical properties such as fast response, stability, low degree of viscous dissipation and high extensibility. The elastomers are prepared from a vinyl-terminated polydimethyl siloxane (PDMS) and a 4-functional crosslinker by a platinum-catalyzed hydrosilylation reaction between the two reactants. Traditionally, elastomers based on hydrosilylation are prepared via a ‘one-step two-pot’ procedure (with a mix A and a mix B mixed in a given ratio). An alternative network formulation method is adopted in this study in order to obtain an elastomeric system with controlled topology – a so-called bimodal network. Bimodal networks are synthesized using a ‘two-step four-pot’ mixing procedure which results in a nonhomogeneous network structure which is shown to lead to novel mechanical properties due to the low extensibility of the short chains and the high extensibility of the long chains. The first ensures stability and the last retards the rupture process thereby combining two desired properties for DEAP purposes without necessarily compromising the viscous dissipation. Several elastomers are prepared and tested for the linear viscoelastic behaviour, i.e. behaviour in the small-strain limit (up to approximately 10% strain). The bimodal networks are, however, capable of extensions up to several times their initial length but the focus here is the small-strain limit.

General information

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Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Technical University of Denmark
Authors: Bejenariu, A. G. (Intern), Boll, M. (Ekstern), Lotz, M. R. (Intern), Vraa, C. (Ekstern), Skov, A. L. (Intern)
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Main Research Area: Technical/natural sciences

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New Soft Polymeric Materials Applicable as Elastomeric Transducers

An elastomer is a material characterized by the capability to regain its original size and shape after being deformed (stretched or distorted). An ideal elastomer for electroactive polymer (EAP) applications is a system characterized by high extensibility, flexibility and a good mechanical fatigue. Dielectric elastomers (DEs) are part of electronic EAPs presenting a good combination of electromechanical properties such as high achievable strains and stresses, fast response speeds, long lifetime, high reliability and high efficiency1. Subjected to a voltage, a polymeric electroactive material sandwiched between two compliant electrodes will reduce its thickness and expand its area. The electrical energy transformed into mechanical energy is called actuation and it is studied in the technology of elastomeric transducers. While DEs deform under high voltage, the actuation varies for different materials (ceramics2, glassy polymers3 or soft polymeric networks4-6). The strain actuation for stiff materials such as glassy or semicrystalline polymers is limited by the electrical breakdown7, while the deformation upon actuation for soft materials is limited by the electromechanical breakdown8. This paper presents new soft polymeric materials based on silicone with improved mechanical properties. Silicone elastomers exhibit good characteristics including biocompatibility, oxidation resistance, thermal stability, fast mechanical response with good reproducibility and stable mechanical behaviour over a wide range of temperature10-13. However, silicone elastomer has weak intermolecular forces among polymeric chains, which limits its mechanical strength. Mechanical properties may be improved using different methods (adding fillers14, interpenetrating network synthesis15 or bimodal network synthesis16). In the present study hyperswollen silicone networks are synthesized and rheologically characterized. Their viscoelastic properties make them good candidates for elastomeric transducers. Silicone networks are synthesized using a hydrosilylation reaction at room temperature between vinyl-terminated polydimethyl siloxanes (PDMS), a 4-functional crosslinker and a platinum catalyst. A ‘one-step two-pot’ mixing procedure is applied and to each premix 70% solvent (heptane) is added. The use of solvent causes networks with fewer entanglements, thus giving the polymeric chains opportunity to act as undisturbed ideal springs. The viscoelastic behavior as function of the applied frequency (LVE diagram) is shown for different hyperswollen networks with varying stoichiometric imbalance (r). The results are compared with results of similar un-swollen networks. The hyperswollen networks are significantly softer and
still easy to handle. From a mechanical point of view, the materials for EAPs use have to be soft with sufficient mechanical strength so the rupture of the material can be avoided at high strain actuation. Considering the EAP requirements and the experimental data for the hyperswollen networks based on silicone, these materials may be considered as good alternatives for the EAP application.

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State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering
Authors: Bejenariu, A. G. (Intern), Skov, A. L. (Intern)
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http://biomimetics2011.u-cergy.fr/
Source: orbit
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Publication: Research - peer-review › Poster – Annual report year: 2011

Novel Ionic Networks derived from the Protonation of Poly(Propylene Imine) Dendrimers with Carboxylic Acid-Telechelic PEG's

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Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering
Authors: Gonzalez, L. (Intern), Hviilsted, S. (Intern), Skov, A. L. (Intern)
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Reversed planar elongation of soft polymeric networks
The newly developed planar elongation fixture, designed as an add-on to the filament stretch rheometer, is used to measure reversible large amplitude planar elongation on soft elastomers. The concept of this new fixture is to elongate an annulus, by keeping the perimeter constant. The deformation on the cylindrical probe is measured using digital imaging, and it is found that the diameter only changes a few percent compared with the initial diameter. Additionally, it is found that a time-strain separable K-BKZ model based on a Doi–Edwards (DE) network strain (without independent alignment) captures the experimental data well. In particular, it is observed that it reproduces the deformation on the cylindrical probe, and this observation confirms our previous statement that the deformation on the cylindrical probe is highly sensitive towards the choice of strain tensor. The stress itself is well described by both the modified Lodge and the DE network strain. The amount of work needed during the deformation is calculated, and it is illustrated how the sample itself contributes with work upon flow reversal. As the stress is time-strain separable the energy loss originates from linear viscoelastic relaxation even in the nonlinear regime, and the loss can be determined solely from linear viscoelastic measurements.

**General information**
State: Published
Organisations: Department of Chemical and Biochemical Engineering, Manufacturing Engineering, Department of Mechanical Engineering, The Danish Polymer Centre
Authors: Jensen, M. K. (Intern), Rasmussen, H. K. (Intern), Skov, A. L. (Intern), Hassager, O. (Intern)
Pages: 729-740
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Silicone based bimodal networks applicable as electroactive systems

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Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Technical University of Denmark
Authors: Bejenariu, A. G. (Intern), Lotz, M. (Ekstern), Boll, M. (Ekstern), Vraa, C. (Ekstern), Skov, A. L. (Intern)
Publication date: 2011

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Silicone_based_bimodal_networks_applicable_as_electroactive_systems_Abstract.pdf
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Stress maximum and steady extensional flow of branched polymer melts

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Manufacturing Engineering, Department of Mechanical Engineering, Durham University, University of Cambridge, University of Leeds
Authors: Huang, Q. (Intern), Skov, A. L. (Intern), Rasmussen, H. K. (Intern), Hoyle, D. M. (Ekstern), McLeish, T. C. (Ekstern), Harlen, O. (Ekstern), Hassel, D. (Ekstern), Lord, T. D. (Ekstern), Mackley, M. R. (Ekstern), Hassager, O. (Intern)
Publication date: 2011
Event: Abstract from 83rd Annual Meeting of The Society of Rheology, Cleveland, OH, United States.
Main Research Area: Technical/natural sciences
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Publication: Research - peer-review › Conference abstract for conference – Annual report year: 2011

Teaching chemical product design to engineering students: course contents and challenges
Chemical product design is not taught in the same way as traditional engineering courses like unit operations or transport phenomena. This paper gives an overview of the challenges that we, as teachers, have faced when teaching chemical product design to engineering students. Specific course contents and relevant teaching methods are discussed.

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State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, CHEC Research Centre
Authors: Skov, A. L. (Intern), Kiil, S. (Intern)
Publication date: 2011
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Main Research Area: Technical/natural sciences
Products, Innovation, Production, Design
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UV-initierede "smart materials"

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, CHEC Research Centre
Authors: Frankær, S. M. G. (Intern), Skov, A. L. (Intern), Kiil, S. (Intern)
Publication date: 2011
Main Research Area: Technical/natural sciences
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Rheology of Cross-linked Polymer Networks


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Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering
Authors: Jensen, M. K. (Intern), Hassager, O. (Intern), Skov, A. L. (Intern), Bach, A. (Ekstern)
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Large Amplitude Oscillatory Extension of Soft Polymeric Networks

sing a filament stretching rheometer (FSR) surrounded by a thermostatic chamber and equipped with a micrometric laser it is possible to measure large amplitude oscillatory elongation (LAOE) on elastomeric based networks with no base flow as in the LAOE method for polymer melts. Poly(dimethylsiloxane) (PDMS) based networks, considered as a model system for studying the development of soft elasticity, are mechanically characterized through reversed LAOE deformation, applying oscillatory measurements with several amplitudes and frequencies.

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Manufacturing Engineering, Department of Mechanical Engineering, CHEC Research Centre
Authors: Bejenariu, A. G. (Intern), Rasmussen, H. K. (Intern), Skov, A. L. (Intern), Hassager, O. (Intern), Frankær, S. M. (Intern)
Planar elongation of soft polymeric networks

A new test fixture for the filament stretch rheometer (FSR) has been developed to measure planar elongation of soft polymeric networks with application towards pressure-sensitive adhesives (PSAs). The concept of this new geometry is to elongate a tube-like sample by keeping the perimeter constant. To validate this new technique, soft polymeric networks of poly(propylene oxide) (PPO) were investigated during deformation. Particle tracking and video recording were used to detect to what extent the imposed strain rate and the sample perimeter remained constant. It was observed that, by using an appropriate choice of initial sample height, perimeter, and thickness, the planar stretch ratio will follow $\lambda(t) = \frac{h(t)}{h(0)} = \exp((\varepsilon)_{\text{overdot}} t)$, with $h(t)$ being the height at time $t$ and $(\varepsilon)_{\text{overdot}}$ the imposed constant strain rate. The perimeter would decrease by a few percent only, which is found to be negligible. The ideal planar extension in this new fixture was confirmed by finite element simulations. Analysis of the stress difference, $\sigma_{zz} - \sigma_{xx}$, showed a network response similar to that of the classical neo-Hookean model. As the Deborah number was increased, the stress difference deviated more from the classical prediction due to the dynamic structures in the material. A modified Lodge model using characteristic parameters from linear viscoelastic measurements gave very good stress predictions at all Deborah numbers used in the quasi-linear regime.

General information

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Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Manufacturing Engineering, Department of Mechanical Engineering, Administration, Coloplast Danmark A/S
Authors: Jensen, M. K. (Intern), Hassager, O. (Intern), Rasmussen, H. K. (Intern), Skov, A. L. (Intern), Bach, A. (Ekstern), Koldbech, H. V. (Intern)
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Reversed extensional flow on polyisoprene melts

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State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Manufacturing Engineering, Department of Mechanical Engineering, Universite Catholique de Louvain
Authors: Bejenariu, A. G. (Intern), Rasmussen, H. K. (Intern), Auhl, D. (Ekstern), Skov, A. L. (Intern), Hassager, O. (Intern)
Publication date: 2010

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Main Research Area: Technical/natural sciences
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Publication: Research - peer-review › Article in proceedings – Annual report year: 2010

Reverse extensional flow on polyisoprene melts

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Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Manufacturing Engineering, Department of Mechanical Engineering, Universite Catholique de Louvain
Authors: Bejenariu, A. G. (Intern), Rasmussen, H. K. (Intern), Auhl, D. (Ekstern), Skov, A. L. (Intern), Hassager, O. (Intern)
Publication date: 2010

Host publication information
Title of host publication: Nordic Rheology Conference
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Source-ID: 271959
Publication: Research - peer-review › Article in proceedings – Annual report year: 2010
Reversible large amplitude planar extension of soft elastomers

The newly developed planar elongation fixture, designed as an add-on to the filament stretch rheometer, is used to measure reversible large amplitude planar elongation on soft elastomers. The concept of the new fixture is to elongate an annulus by keeping the perimeter constant. The deformation on the cylindrical probe is measured using digital imaging, and it is found that the diameter drops a few percent only compared to the initial diameter. Additionally it is found that a new approximation to the Doi-Edwards (DE) model, without independent alignment, captures the experimental data very well. In particular it is observed that this new approximation reproduces the order of magnitude of the deformation on the cylindrical probe. In fact it is demonstrated that the deviation from an ideal planar extension of the cylindrical probe is highly sensitive towards the choice of strain tensor. When analyzing the measured stress data, it is observed that there is some elastic recovery when reversing the flow. This is analyzed calculating the amount of work needed during the deformation, and it is observed that the sample itself contributes with work upon flow reversal.

Reversible planar elongational of soft polymeric networks

Silicone based electroactive polymers: application driven development
A comparative study on the influence of the platinum catalyst in poly(dimethylsiloxane) based networks synthesis

The aim of the project is to find the best of three Pt catalysts and their appropriate quantity in order to obtain soft networks in one hour at room temperature. How the choice of catalyst influences the final elastomeric properties is also evaluated. The differences between the catalysts are the solvent and the platinum concentration.

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Risø National Laboratory for Sustainable Energy
Authors: Bejenariu, A. G. (Intern), Poulsen, J. Ø. (Ekstern), Skov, A. L. (Intern), Henriksen, L. C. (Ekstern)
Number of pages: 282
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Volume: 17
Publisher: Chalmers tekniska högskola
Main Research Area: Technical/natural sciences
Solid Oxide Fuel Cells, Materials research
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Source: orbit
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Publication: Research › peer-review › Article in proceedings – Annual report year: 2009

Characterization of Polymer Networks by Swelling
The linear dynamic response of crosslinked PDMS samples with different stoichiometries, as well as the sol and network fractions, was investigated. It was found that all the networks showed similar properties after removal of the sol indicating only small deviations in the network structure upon changing the stoichiometric imbalance.

General information
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Organisations: CHEC Research Centre, Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Frankær, S. M. (Intern), Jensen, M. K. (Intern), Bejenariu, A. G. (Intern), Skov, A. L. (Intern), Hassager, O. (Intern)
Publication date: 2009

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Main Research Area: Technical/natural sciences
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Publication: Research › Article in proceedings – Annual report year: 2009

Elongational dynamics of multiarm polystyrene
The startup of uni-axial elongational flow followed by stress relaxation and reversed bi-axial flow has been measured for a branched polystyrene melt with narrow molar mass distribution using the filament stretching rheometer. The branched polystyrene melt was a multiarm A(q)-C-C-A(q) pom-pom polystyrene with an estimated average number of arms of q=2.5. The molar mass of each arm is about 28 kg/mole with an overall molar mass of M-w=280 kg/mole. An integral molecular stress function constitutive formulation within the "interchain pressure" concept agrees reasonably well with the experiments.

General information
State: Published
Organisations: Manufacturing Engineering, Department of Mechanical Engineering, The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Technical University of Denmark
Authors: Rasmussen, H. K. (Intern), Skov, A. L. (Intern), Nielsen, J. K. (Intern), Laille, P. (Ekstern)
Linear rheology of cross-linked polypropylene oxide as a pressure sensitive adhesive

Soft polymer networks are commonly used as pressure sensitive adhesives (PSAs). This is due to their unique ability to deform and yet to resist flow. These contradictory requirements indicate that the mechanical properties are finely tuned, and that the types of deformation upon application are carefully considered. Two main mechanisms must be considered when studying adhesives, that is the debonding and bonding mechanisms. Linear rheology is used to study the debonding mechanisms to gain better understanding of the peeling process of the PSAs. A variety of PSAs are prepared by mixing a linear vinyl terminated polymer with a silane terminated f-functional cross-linker, with \( f > 2 \). The stoichiometric imbalance, \( r \) (silane to vinyl ratio), the molecular weight of the linear polymer, \( M \), and the cross-linker functionality, \( f \), are used as adjustable parameters to tune the properties of the cross-linked networks. The adhesive performance was tested with 90 degrees peel tests at three peel rates and thicknesses, and it was observed that the peel force varies with \( r \), \( M \) and \( f \) and also the peel rate. The fundamental viscoelastic parameters that govern the PSA performance of cross-linked systems were used to state an empirical relation for the peel force. The relation is combining the peel force with the loss tangent at the peel frequency and the equilibrium modulus. Based on this, basic guidelines for selecting the appropriate polymer/cross-linker system to achieve the target performance are given.
New large amplitude oscillatory elongation method applied on elastomeric PDMS networks
The reversed deformation measurements give important information about the entropic state of the sample and about the behaviour of the polymer inside it. Even though there exist important stretching methods studies through rheometry [5], to our knowledge this is the first elongational method applied on elastomers for measuring the elastic recovery through oscillations at a constant strain.

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Event: Poster session presented at 5th Annual European Rheology Conference, Cardiff, United Kingdom.
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Publication: Research - peer-review › Journal article – Annual report year: 2009

Optimization of the synthesis parameters for obtaining PDMS samples suitable for filament stretch rheometer measurements
The filament stretch rheometer (FSR) can be used for large amplitude oscillatory elongation measurements (LAOE) on polymer networks. During sample formation the shape and integrity of the samples can be affected. This research paper presents the results of an optimization study concerning the synthesis parameters for obtaining silicone samples suitable for FSR.

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Source: orbit
Source-ID: 244298
Publication: Research - peer-review › Article in proceedings – Annual report year: 2009

Planar Elongation Measurements on Soft Elastomers
A new fixture to the filament stretch rheometer (FSR) has been developed to measure planar elongation of soft polymeric networks. To validate this new technique, soft polymeric networks of poly(propyleneoxide) (PPO) were investigated during deformation.

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Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Department of Mechanical Engineering, Coloplast Danmark A/S
Authors: Jensen, M. K. (Intern), Skov, A. L. (Intern), Rasmussen, H. K. (Intern), Bach, A. (Ekstern), Hassager, O. (Intern)
Publication date: 2009

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Self-consisting modeling of entangled network strands and dangling ends
Text of Abstract We seek knowledge about the effect of dangling ends and soluble structures of stoichiometrically imbalanced networks. To interpretate our recent experimental results we seek a molecular model that can predict LVE data. The discrete slip-link model (DSM) has proven to be a robust tool for LVE and non-linear rheology predictions for linear chains, and it is thus used to analyze the experimental results. We divide the LVE predictions into three domains; 1) the low frequency region, where $G'$ is a plateau, $G_0$, the intermediate frequency region, where $G'$ and $G''$ are parallel and 3) the high frequency region, where $G'$ levels off to an entanglement plateau, $G_N0$, close to that of the linear polymer. The latter region is seldom obtained in experiments, while it is obtained in simulations since these start at zero time. Initially we consider a stoichiometrically balanced network, we call this an ideal entangled network (IEN). We simulate monodisperse poly(propylene oxide) with an average number of entanglements of ~3.8. Such lightly entangled networks show a $G_0$ that is about 24% lower than $G_N0$. This decrease is a result of monomer fluctuations between entanglements. Additionally we observe that $G'$ is dominating at all frequencies compared to $G''$. Experimental observations of stoichiometrically imbalanced networks shows that $G'$ and $G''$ are of the same order of magnitude at intermediate frequencies, hence the DSM suggests that energy dissipation is largely a result of dangling ends and soluble structures. Energy dissipation is increased by adding a fraction of dangling ends, $w_{DE}$, to the ensemble. We find that when $w_{DE}=0.6$, $G_0$ is about 75% lower than $G_N0$, this suggests that the fraction of network strands, $w_{NS}=1-w_{DE}$, largely influences the plateau value at low frequencies. Soluble strands can also be added to the theory which is expected to increase energy dissipation further.
Constitutive equation for polymer networks with phonon fluctuations

Recent research by Xing [Phys. Rev. Lett. 98, 075502 (2007)] has provided an expression for the Helmholtz free energy related to phonon fluctuations in polymer networks. We extend this result by constructing the corresponding nonlinear constitutive equation, usable for entirely general, volume conserving deformation fields. Constitutive equations for the sliplink model and the tube model are derived and the three models are examined by comparison with each other and with data from Xu and Mark [Rubber Chem. Technol. 63, 276 (1990)] and Wang and Mark [J. Polym. Sci., Part B: Polym. Phys. 30, 801 (1992)]. Elastic moduli are derived for the three models and compared with the moduli determined from the chemical stoichiometry. We conclude that the sliplink model and the phonon fluctuation model are relatively consistent with each other and with the data. The tube model seems consistent neither with the other models nor with the data.
Constitutive equation for polymer networks with phonon fluctuations

Recent research by Xing et al. [Phys. Rev. Lett. 98, 075502, 2007] has provided an expression for the Helmholtz free energy related to phonon fluctuations in polymer networks. We extend this result by constructing the corresponding nonlinear constitutive equation, usable for entirely general, volume conserving deformation fields. Constitutive equations for the sliplink and the tube model are applied. The three models are examined by comparison with each other and with data from Xu and Mark [Rubber Chem. Technol. 63, 276, 1990] and Wang and Mark [J. Polym. Sci. Part B: Polym. Phys. 30, 801, 1992] on end-linked polydimethylsiloxane networks. Elastic moduli are derived for the three models and compared with the moduli determined from the chemical stoichiometry. We conclude that the sliplink model and the phonon fluctuation model are relatively consistent with each other and also with the data. The tube model seems consistent neither with the other models nor with the data. © 2008 American Institute of Physics.

General information
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Organisations: Department of Chemical and Biochemical Engineering, The Danish Polymer Centre
Authors: Hansen, R. (Intern), Hassager, O. (Intern), Skov, A. L. (Intern)
Pages: 1369-1371
Publication date: 2008

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Title of host publication: CP1027, The XVth International Congress on Rheology, The Society of Rheology 80th Annual Meeting
Measurement of Reversed Extension Flow using the Filament Stretch Rheometer

The measurement of material functions with reversed extension flow is demonstrated using the Filament Stretching Rheometer (FSR). This includes startup of uniaxial elongational flow (potentially until steady state) followed by biaxial squeezing, and large amplitude oscillatory elongation (LAOE). The latter is applicable on highly extensible elastomers, whereas in LAOE measurements on liquids (including polymer melts) the LAOE flow needs to be imposed upon a constant strain rate uniaxial elongation. The used Filament Stretching Rheometer allows measurements on polymeric fluids (including polymeric melts) from room temperature until 200 degrees C.

General information
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Organisations: Manufacturing Engineering, Department of Mechanical Engineering, The Danish Polymer Centre, Department of Chemical and Biochemical Engineering
Authors: Rasmussen, H. K. (Intern), Skov, A. L. (Intern), Nielsen, J. K. (Intern), Laille, P. (Ekstern)
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Publication date: 2008

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Main Research Area: Technical/natural sciences
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Optimering af hudlæbere

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Manufacturing Engineering, Department of Mechanical Engineering, Coloplast Danmark A/S
Authors: Jensen, M. K. (Intern), Skov, A. L. (Intern), Bach, A. (Ekstern), Hassager, O. (Intern), Rasmussen, H. K. (Intern)
Pages: 14-17
Publication date: 2008
Main Research Area: Technical/natural sciences

Physical and chemical properties of dielectric elastomers

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Solar Energy Programme, Risø National Laboratory for Sustainable Energy
Authors: Skov, A. L. (Intern), Sommer-Larsen, P. (Intern)
Number of pages: 344
Pages: 25-32
Publication date: 2008

Rheology and Adhesive Performance of Soft Polymeric Networks

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Coloplast Danmark A/S
Authors: Jensen, M. K. (Intern), Hassager, O. (Intern), Skov, A. L. (Intern), Bach, A. (Ekstern)
Publication date: 2008
Kinetic investigation of the addition curing vulcanization

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering
Authors: Skov, A. L. (Intern)
Publication date: 2007

Host publication information
Title of host publication: ANNUAL TRANSACTIONS OF THE NORDIC RHEOLOGY SOCIETY
Volume: 15
Main Research Area: Technical/natural sciences
Conference: 16th Nordic Rheology Conferences, Stavanger, Norway, 01/01/2007
Source: orbit
Source-ID: 220419
Publication: Research › Article in proceedings – Annual report year: 2007

Dynamic Dilution Effects in Polymeric Networks
The relaxation processes occurring in slightly and well-entangled polydimethylsiloxane (PDMS) networks are investigated. Swelling experiments are performed in order to determine the sol fractions. The low-frequency linear rheology of the two types of networks reveal two significant relaxation processes, namely the reptation of linear species within the network and the arm withdrawal process of star arms in the sol fraction and of dangling single-chain ends attached to the network. The relaxation spectra are influenced by the stoichiometry to a large extent due to dynamic dilution effects caused by the change in the amount of dangling arms and solubles with stoichiometry. The star arm relaxation is suppressed by washing out the sol fraction which is seen as a clear example of the dynamic dilution effect arising from the small amount of non-reactive PDMS.

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Polymers for Energy Technology, Polymer Department, Risø National Laboratory for Sustainable Energy
Authors: Skov, A. L. (Intern), Sommer-Larsen, P. (Intern), Hassager, O. (Intern)
Number of pages: 11
Pages: 042
Publication date: 24 Jul 2006
Main Research Area: Technical/natural sciences

Publication information
Journal: e-polymers
ISSN (Print): 1618-7229
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): SNIP 0.43 SJR 0.335 CiteScore 1.02
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): SJR 0.292 SNIP 0.345 CiteScore 0.64
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 0.167 SNIP 0.13 CiteScore 0.28
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 0.135 SNIP 0.07 CiteScore 0.15
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 0.145 SNIP 0.086 CiteScore 0.14
Some experimental results for end-linked polydimethylsiloxane network system

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Risø National Laboratory
Authors: Larsen, A. L. (Intern), Larsen, P. S. (Ekstern), Hassager, O. (Intern)
Pages: 1-18
Publication date: 2004
Main Research Area: Technical/natural sciences

Publication information
Journal: e-Polymers
Volume: 050
Original language: English
Links:
http://www.e-polymers.org
Source: orbit
Source-ID: 138300
Publication: Research - peer-review › Journal article – Annual report year: 2004

Elastic properties of nonstoichiometric reacted PDMS networks
The influence of stoichiometry on the elastic modulus of eight-functional end-linked poly(dimethylsiloxane) (PDMS) networks was investigated by extensional rheometry with extensions up to more than 100%, and the stress-strain relation was found to be almost linear—a characteristic property for a network structure with an eight-functional cross-linker. The experimental data were compared to a stochastic model taking into account entanglements and to Monte Carlo simulations. The Mooney-Rivlin model was furthermore used to fit the data, and the dependency of C-1 and C-2 parameters on the stoichiometric ratio was investigated in order to clarify especially the influence of trapped entanglements acting either as chemical cross-links or as sliding links. It was found that including a locking factor dividing trapped entanglements into locked entanglements and slip-links could explain our data obtained for the Mooney-Rivlin constants. It was furthermore found that trapped entanglements dominate when there is an excess of cross-linker, ensuring that all long difunctional DMS chains are bound to the infinite network in both ends.

General information
State: Published
Tailoring Silicone Networks

General information
State: Published
Organisations: The Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Risø National Laboratory for Sustainable Energy
Authors: Skov, A. L. (Intern), Sommer-Larsen, P. (Intern), Hassager, O. (Intern)
Publication date: 2003

Host publication information
Title of host publication: Proceedings of the Nordic Polymer Days 2003
Main Research Area: Technical/natural sciences
Source: orbit
Source-ID: 41358
Publication: Research › Conference abstract in proceedings – Annual report year: 2003

Projects:

Viscoelastic Response of Double Dynamics Polymer Networks under Transient Elongation
Department of Chemical and Biochemical Engineering
Period: 01/06/2018 → 31/05/2021
Number of participants: 4
Phd Student:
Wang, Wendi (Intern)
Supervisor:
Hassager, Ole (Intern)
Huang, Qian (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Marie Curie (EU-stipendium)
Project: PhD

Coating with inherent sensing functionality based on dielectric elastomers
Department of Chemical and Biochemical Engineering
Period: 01/02/2018 → 31/01/2021
Number of participants: 3
Phd Student:
Heavy Duty Coatings
Department of Chemical and Biochemical Engineering
Period: 01/02/2018 → 31/01/2021
Number of participants: 4
PhD Student:
Ekbrant, Björn Erik Fristrup (Intern)
Supervisor:
Ambat, Rajan (Intern)
Paulsen, Andreas Lundtang (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Industrial PhD
Project: PhD

Structure property relationships in coatings
Department of Chemical and Biochemical Engineering
Period: 01/02/2018 → 31/01/2021
Number of participants: 4
PhD Student:
Juraskova, Alena (Intern)
Supervisor:
Dam-Johansen, Kim (Intern)
Olsen, Stefan Møller (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Samfinansieret - Andet
Project: PhD

Strechable conductive elastomers
Department of Chemical and Biochemical Engineering
Period: 01/01/2018 → 31/12/2020
Number of participants: 3
PhD Student:
Shao, Jiang (Intern)
Supervisor:
Skov, Anne Ladegaard (Intern)
Main Supervisor:
Daugaard, Anders Egede (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Stipendie fra udlandet
Study on self-healing and high-permittivity silicone elastomer/keratin composite materials

Department of Chemical and Biochemical Engineering
Period: 01/12/2017 → 30/11/2020
Number of participants: 4
Phd Student:
Liu, Xue (Intern)
Supervisor:
Nie, Yi (Ekstern)
Zhang, Suo-Jiang (Ekstern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Stipendie fra udlandet
Project: PhD

Functional Polysilazanes for Coating Applications

Department of Chemical and Biochemical Engineering
Period: 01/09/2017 → 31/01/2018
Number of participants: 4
Phd Student:
Kristiansen, Thomas (Intern)
Supervisor:
Dam-Johansen, Kim (Intern)
Daugaard, Anders Egede (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Samfinansieret - Andet
Project: PhD

Advanced wound care adhesives with new functional properties

Department of Chemical and Biochemical Engineering
Period: 01/08/2017 → 31/07/2020
Number of participants: 6
Phd Student:
Chiaula, Valeria (Intern)
Supervisor:
Mazurek, Piotr Stanislaw (Intern)
Nielsen, Anders Christian (Ekstern)
Tornøe, Jens (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Industrial PhD
Project: PhD

Synthesis and characterization of hydrogels to be used as dielectric elastomers

Department of Chemical and Biochemical Engineering
Antimicrobial Polymers for Catheter Coatings

Department of Chemical and Biochemical Engineering
Period: 01/01/2017 → 31/12/2019
Number of participants: 4
Phd Student:
Andersen, Christian (Intern)
Supervisor:
Madsen, Niels Jørgen (Ekstern)
Skov, Anne Ladegaard (Intern)
Main Supervisor:
Daugaard, Anders Egede (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Industrial PhD
Project: PhD

Mechanical and photochemical stabilization of flexible organic solar cells

Department of Chemical and Biochemical Engineering
Period: 01/01/2017 → 31/12/2019
Number of participants: 4
Phd Student:
Ogliani, Elisa (Intern)
Supervisor:
Hvilsted, Søren (Intern)
Yu, Liyun (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Samfinansieret - Andet
Project: PhD

Heat Transfer in dielectric elastomers

Department of Chemical and Biochemical Engineering
Period: 01/05/2016 → 30/04/2019
Number of participants: 3
Phd Student:
Christensen, Line Riis (Intern)
Supervisor:
Hassager, Ole (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)
Silicone elastomers with aromatic voltage stabilizers

Department of Chemical and Biochemical Engineering

The Danish Polymer Centre
Period: 01/11/2015 → 01/07/2016
Number of participants: 2
Aromatic voltage stabilizers, Silicone elastomers, Electron-trapping effect, PDMS-PPMS copolymers
Project ID: 52070
Project participant:
A Razak, Aliff Hisyam (Intern)
Skov, Anne Ladegaard (Intern)

On the correlation between Young’s modulus and melt flow in fiber spinning operations

Department of Chemical and Biochemical Engineering

Period: 15/09/2014 → 13/11/2017
Number of participants: 6
Phd Student:
Wingstrand, Sara Lindeblad (Intern)
Supervisor:
Szabo, Peter (Intern)
Main Supervisor:
Hassager, Ole (Intern)
Examiner:
Skov, Anne Ladegaard (Intern)
Peters, Gerrit W.M. (Ekstern)
von Ruymbeke, Evelyne (Ekstern)

Enhancement of dielectric permittivity by incorporating PDMS-PEG multiblock copolymers in silicone elastomers

A silicone elastomer from PDMS-PEG multiblock copolymer has been prepared by use of silylation reactions for both copolymer preparation and crosslinking. The dielectric and mechanical properties of the silicone elastomers were carefully investigated, as well as the morphology of the elastomers was investigated by SEM. The developed silicone elastomers were too conductive to be utilized as dielectric elastomers but it was shown that when the above silicone elastomers were mixed with a commercial silicone elastomer, the resulting elastomer had very favourable properties for dielectric elastomers due to a significantly increased dielectric permittivity. The conductivity also remained low due to the resulting discontinuity in PEG within the silicone matrix.

Department of Chemical and Biochemical Engineering

Period: 01/01/2014 → 31/12/2014
Number of participants: 3
PDMS-PEG block copolymers, high permittivity, silicone elastomers, hydrosilylation
Project ID: 52070
Project participant:
A Razak, Aliff Hisyam (Intern)
Szabo, Peter (Intern)
A chemical approach to avoiding electromechanical instabilities in dielectric electroactive polymer films

Department of Chemical and Biochemical Engineering
Period: 01/12/2013 → 15/05/2017
Number of participants: 6
Phd Student: A Razak, Aliff Hisyam (Intern)
Supervisor: Szabo, Peter (Intern)
Main Supervisor: Skov, Anne Ladegaard (Intern)
Examiner: Daugaard, Anders Egede (Intern)
Carpi, Federico (Ekstern)
Sommer-Larsen, Peter (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Stipendie fra udlandet

Relations
Publications:
Optimisation of Silicone-based Dielectric Elastomer Transducers by Means of Block Copolymers - Synthesis and Compounding
Project: PhD

Supramolecular Polymeric Rheology

Department of Chemical and Biochemical Engineering
Period: 01/10/2013 → 14/12/2016
Number of participants: 6
Phd Student: Shabbir, Aamir (Intern)
Supervisor: Skov, Anne Ladegaard (Intern)
Main Supervisor: Hassager, Ole (Intern)
Examiner: Daugaard, Anders Egede (Intern)
Creton, Costantino (Ekstern)
Ianniruberto, Giovanni (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Marie Curie (EU-stipendium)

Relations
Publications:
Rheology of Supramolecular Polymers
Project: PhD

Microcapsules as delivery vehicles

Department of Chemical and Biochemical Engineering
Period: 15/09/2013 → 15/03/2017
Number of participants: 6
Phd Student:
Mazurek, Malgorzata Natalia (Intern)
Supervisor:
Hvilsted, Søren (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)
Examiner:
Daugaard, Anders Egede (Intern)
Hietala, Sami Heikki Olavi (Ekstern)
Paulsen, Andreas Lundtang (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Institut stipendie (DTU) Samf.

Relations
Publications:
Remotely Activated Microcapsules for Oil Recovery Treatments
Project: PhD

Extensional Rheology of Model Polymers
Department of Chemical and Biochemical Engineering
Period: 01/04/2013 → 16/06/2016
Number of participants: 7
Phd Student:
Hengeller, Ludovica (Intern)
Supervisor:
Almdal, Kristoffer (Intern)
Skov, Anne Ladegaard (Intern)
Main Supervisor:
Hassager, Ole (Intern)
Examiner:
Szabo, Peter (Intern)
Ianniruberto, Giovanni (Ekstern)
Vlassopoulos, Dimitrios (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: 1/3 FUU, 1/3 inst 1/3 Andet

Relations
Publications:
Entangled Polymer Melts in Extensional Flow
Project: PhD

Synthesis and Evaluation of Structures Lipids based Polyurethanes for Sensors, Actuators and Generators trough Monte Carlo Simulation
Department of Chemical and Biochemical Engineering
Period: 15/01/2013 → 26/05/2016
Number of participants: 7
Phd Student:
Zakaria, Shamsul Bin (Intern)
Supervisor:
Daugaard, Anders Egede (Intern)
Gernaey, Krist V. (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)
Examiner:
Daugaard, Anders Egede (Intern)
Jager, Edwin Willem Harm (Ekstern)
Sarban, Rahimullah (Intern)

**Financing sources**
Source: Internal funding (public)
Name of research programme: Stipendie fra udlandet

**Relations**
Publications:
Electrical Breakdown and Mechanical Ageing in Dielectric Elastomers
Project: PhD

**Mechanically invisible encapsulations**
Department of Chemical and Biochemical Engineering
Period: 15/12/2012 → 14/03/2016
Number of participants: 6
Phd Student:
Mazurek, Piotr Stanislaw (Intern)
Supervisor:
Hvilsted, Søren (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)
Examiner:
Szabo, Peter (Intern)
Benslimane, Mohamed Yahia (Ekstern)
Köllnberger, Andreas (Ekstern)

**Financing sources**
Source: Internal funding (public)
Name of research programme: Institut stipendie (DTU) Samf.

**Relations**
Publications:
Mechanically invisible encapsulations
Project: PhD

**Exploring biochemical process performance limits through topology optimization**
Department of Chemical and Biochemical Engineering
Period: 01/12/2012 → 16/06/2016
Number of participants: 7
Phd Student:
Larsson, Hilde Kristina (Intern)
Supervisor:
Gernaey, Krist V. (Intern)
Skov, Anne Ladegaard (Intern)
Main Supervisor:
Krühne, Ulrich (Intern)
Examiner:
Sin, Gürkan (Intern)
Drønen, Nils Kjetil (Intern)
Nopens, Ingmar (Ekstern)

**Financing sources**
Source: Internal funding (public)
Name of research programme: Grundforskningsfonden

**Relations**
Publications:
Modelling of Mass Transfer Phenomena in Chemical and Biochemical Reactor Systems using Computational Fluid Dynamics
**Novel clay/nanocellulose biocomposite films and coatings in the context of packaging applications**

Department of Chemical and Biochemical Engineering  
Period: 01/10/2012 → 20/09/2016  
Number of participants: 7  
Phd Student:  
Trifol Guzman, Jon (Intern)  
Supervisor:  
Daugaard, Anders Egede (Intern)  
Hassager, Ole (Intern)  
Main Supervisor:  
Szabo, Peter (Intern)  
Examiner:  
Skov, Anne Ladegaard (Intern)  
Niska, A. Kristiina Oksman (Ekstern)  
Rojas, Orlando (Ekstern)  

**Financing sources**  
Source: Internal funding (public)  
Name of research programme: Marie Curie (EU-stipendium)

**Relations**  
Publications:  
Hybrid nanocellulose/nanoclay composites for food packaging applications  
Project: PhD

**A soft and conductive PDMS-PEG block copolymer as a compliant electrode for the dielectric elastomer**

Soft conducting elastomers have been prepared from polydimethylsiloxane-polyethylene glycol (PDMS-PEG) copolymer and surfactant-stabilized multi-walled carbon nanotubes (MWCNTs). The copolymer was chain-extended with PDMS of molecular weight 17.2 kg mol-1 in order to obtain a crosslinkable PDMS with molecular weight around 20 – 30 kg mol-1. MWCNTs were treated with surfactant and sonicated for better dispersion in the polymer matrix. The conductivity and mechanical properties of conducting elastomers were thoroughly investigated including stress and strain at break. The developed conducting elastomers showed high conductivity combined with inherent softness. The high conductivity and softness, PDMS-PEG copolymers with incorporated MWCNTs hold great promises as compliant and highly stretchable electrodes for stretchable devices such as electro-mechanical transducers.

Department of Chemical and Biochemical Engineering  
The Danish Polymer Centre  
Period: 01/08/2012 → 31/01/2016  
Number of participants: 3  
Project ID: 52070  
Project participant:  
A Razak, Aliff Hisyam (Intern)  
Madsen, Frederikke Bahrt (Intern)  
Project Coordinator:  
Skov, Anne Ladegaard (Intern)  
Documents:  
MRS Adv._official_aliff

**Monolithic structures and their influence on electromechanical breakdown phenomena**

Department of Chemical and Biochemical Engineering  
Period: 01/08/2012 → 03/06/2015  
Number of participants: 6  
Phd Student:  
Hassouneh, Suzan Sager (Intern)
Synthesis and development of novel coating components for exterior wood applications based on sustainable resources

Department of Chemical and Biochemical Engineering
Period: 01/08/2012 → 22/02/2016
Number of participants: 6
Phd Student:
Nguyen, Hiep Dinh (Intern)

Supervisor:
Hvilsted, Søren (Intern)
Main Supervisor:
Daugaard, Anders Egede (Intern)
Examiner:
Skov, Anne Ladegaard (Intern)
Johansson, Mats (Ekstern)
Paulsen, Andreas Lundtang (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: 1/3 DTU-stip, 2/3 FUR/andet
Project: PhD

Relations
Publications:
Bio-based alkyds by direct enzymatic bulk polymerization
Project: PhD

Development of new materials for dielectric electroactive polymers as actuators and generators

Department of Chemical and Biochemical Engineering
Period: 15/08/2011 → 26/11/2014
Number of participants: 7
Phd Student:
Madsen, Frederikke Bahrt (Intern)

Supervisor:
Daugaard, Anders Egede (Intern)
Hvilsted, Søren (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)
Examiner:
Kiil, Søren (Intern)
Brook, Michael Adrian (Ekstern)
Graz, Ingrid Maria (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Institut stipendie (DTU) Samf.
Project: PhD
Development of stimuli-responsive polymer network
Department of Chemical and Biochemical Engineering
Period: 01/08/2011 → 17/12/2014
Number of participants: 6
Phd Student:
Ma, Baoguang (Intern)
Supervisor:
Hvilsted, Søren (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)
Examiner:
Daugaard, Anders Egede (Intern)
Hietala, Sami Heikki Olavi (Ekstern)
Paulsen, Andreas Lundtang (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Eksternt finansieret virksomhed
Project: PhD

Large Scale Processing of dielectric electro active polymers
Department of Chemical and Biochemical Engineering
Period: 01/08/2011 → 30/09/2014
Number of participants: 7
Phd Student:
Vudayagiri, Sindhu (Intern)
Supervisor:
Daugaard, Anders Egede (Intern)
Kiørboe, Lars Georg (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)
Examiner:
Szabo, Peter (Intern)
Shea, Herbert R. (Ekstern)
Sommer-Larsen, Peter (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Offentlig finansiering
Project: PhD

Mechanical characterization methods for dielectric electroactive polymers
Department of Chemical and Biochemical Engineering
Period: 01/08/2011 → 26/11/2014
Number of participants: 6
Phd Student:
Goswami, Kaustav (Intern)
Supervisor:
Daugaard, Anders Egede (Intern)
Main Supervisor:
Skov, Anne Ladegaard (Intern)
Examiner:
von Solms, Nicolas (Intern)
Benslimane, Mohamed Yahia (Ekstern)
Opris, Dorina (Ekstern)
Financing sources
Source: Internal funding (public)
Name of research programme: Institut stipendie (DTU) Samf.
Project: PhD

Loading of micro-containers for oral drug delivery
Department of Micro- and Nanotechnology
Period: 01/02/2011 → 14/05/2014
Number of participants: 7
Phd Student:
Marizza, Paolo (Intern)
Supervisor:
Keller, Stephan Sylvest (Intern)
Müllertz, Anette (Ekstern)
Main Supervisor:
Boisen, Anja (Intern)
Examiner:
Skov, Anne Ladegaard (Intern)
Rantanen, Jukka Tapio (Ekstern)
Schneider, Marc (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Institut stipendie (DTU) Samf.
Project: PhD

New Polymer Architectures for Proton Conducting Fuel Cell Membranes
Department of Chemical and Biochemical Engineering
Period: 01/03/2010 → 21/05/2013
Number of participants: 6
Phd Student:
Nielsen, Mads Møller (Intern)
Supervisor:
Jankova Atanasova, Katja (Intern)
Main Supervisor:
Hvilsted, Søren (Intern)
Examiner:
Skov, Anne Ladegaard (Intern)
Jones, Deborah (Ekstern)
Tenhu, Heikki Juhani (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Forskningsrådsfinansiering
Project: PhD

Molecular Rheology of Complex Fluids
Department of Chemical and Biochemical Engineering
Period: 01/02/2010 → 19/04/2013
Number of participants: 7
Phd Student:
Huang, Qian (Intern)
Supervisor:
Rasmussen, Henrik K. (Intern)
Skov, Anne Ladegaard (Intern)
Main Supervisor:
Hassager, Ole (Intern)
Stimuli-Adaptable Materials
Department of Chemical and Biochemical Engineering
Period: 01/05/2009 → 21/05/2013
Number of participants: 7
Phd Student: Frankær, Sarah Maria Grundahl (Intern)
Supervisor: Daugaard, Anders Egede (Intern)
Kiil, Søren (Intern)
Main Supervisor: Skov, Anne Ladegaard (Intern)
Examiner: Hvilsted, Søren (Intern)
Hansen, Kristoffer Karsten (Intern)
Hietala, Sami (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Marie Curie (EU-stipendium)
Project: PhD

From Nanostructure to Micro Morphology in Polymers
Department of Mechanical Engineering
Period: 01/04/2008 → 30/11/2011
Number of participants: 7
Phd Student: Yu, Kaijia (Intern)
Supervisor: Hansen, Hans Nørgaard (Intern)
Skov, Anne Ladegaard (Intern)
Main Supervisor: Rasmussen, Henrik K. (Intern)
Examiner: Szabo, Peter (Ekstern)
Stading, Mats (Ekstern)
Svaneborg, Carsten (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Institut stipendie (DTU)
Project: PhD

Department of Chemical and Biochemical Engineering
Period: 15/07/2007 → 29/09/2010
Number of participants: 6
Phd Student:
Conte, Elisa (Intern)
Supervisor:
Abildskov, Jens (Intern)
Main Supervisor:
Gani, Rafiqul (Intern)
Examiner:
Skov, Anne Ladegaard (Intern)
Jiménez-González, Concepción (Ekstern)
Kate, Antoon J. B. ten (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: DTU-lønnet stipendie
Project: PhD

Physics of Adhesion
Department of Chemical and Biochemical Engineering
Period: 15/11/2006 → 17/03/2010
Number of participants: 7
Phd Student:
Keller, Mette Krog (Intern)
Supervisor:
Bach, Anders (Ekstern)
Skov, Anne Ladegaard (Intern)
Main Supervisor:
Hassager, Ole (Intern)
Examiner:
Szabo, Peter (Ekstern)
Creton, Costantino (Ekstern)
Venerus, David C. (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: 1/3 DTU-stip, 2/3 FUR/andet
Project: PhD

Dielektriske elastomer aktuatorer
Department of Chemical and Biochemical Engineering
Period: 01/08/2001 → 23/08/2004
Number of participants: 5
Phd Student:
Skov, Anne Ladegaard (Intern)
Supervisor:
Sommer-Larsen, Peter (Intern)
Main Supervisor:
Hassager, Ole (Intern)
Examiner:
Keunings, Roland (Ekstern)
Ndoni, Sokol (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Forskningsrådsfinansiering
Project: PhD

Activities:
Material design and processing – hard competition or great synergy?
Period: 19 Sep 2012
Anne Ladegaard Skov (Lecturer)
Department of Chemical and Biochemical Engineering
The Danish Polymer Centre

Related event

Materiality of Light: Designing With Light And Energy Flows
19/09/2012 → 21/09/2012
Copenhagen, Denmark
Activity: Talks and presentations › Conference presentations