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Silicone elastomer map: Design the ideal elastomer

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ABSTRACT

Soft, stretchable and light-weight transducers are most sought after for research on advanced applications like stretchable electronics, soft robotics and energy harvesters. Stretchable electronics require elastomers that have high elongation at break, high dielectric permittivity and high breakdown strength. Commercial silicone elastomer formulations often do not encompass all the necessary properties required to function effectively as stretchable transducers but they are used out of familiarity. In this study, most commonly used commercial silicone formulations are formulated with different stoichiometry and also blends of these formulations are made in order to manipulate their resulting properties. The properties of these blends like ultimate stress and strain, Young's modulus, dielectric permittivity and breakdown strength are investigated and mapped to identify those that have the best suited properties for fabricating soft stretchable devices. On a research level, Sylgard 184, Sylgard 186, Ecoflex 00-50, Ecoflex 00-30 and Ecoflex 00-10 are widely used for fabricating such soft devices and hence they will be worked upon in this study. The elastomers obtained using the methods of mixing illustrated here can act as a starting point for conceptualizing the feasibility of a product on research level.

1. INTRODUCTION

An unprecedented interest in transitioning from hard to soft machinery has propelled extensive research in the field of flexible and stretchable transducers [1][2]. Advantages of soft, stretchable, lightweight electronics that can withstand the wear and tear of movement in all degrees of freedom[3][4][5] are plenty compared to the conventional rigid technology. Particularly, for developing futuristic applications, for example: lifelike prosthetics and robots, intrinsic wearable biomedical devices, flexible digital computing devices, camouflage coatings and wave/solar/wind energy harvesters the aforementioned properties are key. Active research is taking place recently for developing robust and fully functional soft robotics[6], stretchable and soft electronics [7] wearable digital computing devices[7] flexible circuits and spatial solar panels [8], stretchable transparent conductors [9], printable elastic conductors[10], wearable, flexible devices for memories and computing[11], pressure sensitive sensor skin emulating human skin[12]–[19]. Such a myriad of applications requires a versatile elastomer that gives the best performance efficiency to these devices. Although, a variety of commercial formulations are available with superior mechanical properties in various grades they are not ideal for all the applications[20]. Lack of readily available elastomer formulations with customized properties for individual applications is one of the main constraining factors for producing stretchable devices with optimum performance efficiency. Although Sylgard 184 from Dow is very popular in research community for its versatile properties (low viscosity, high optical transmittance and durability[21], [22]), it has limited extensibility, high Young's modulus (Y) and moderate dielectric properties, which is a disadvantage for developing stretchable electronics, especially, high-voltage applications[23]. Sylgard 184 is used for a wide range of research activity, e.g. to develop shape memory composites[24], compliant graphite electrodes[25], microfluidic devices [26], super stretch strain sensor[27], micro contact pressure sensor [28] and tactile sensor[29].

An alternative to Sylgard 184 is a low-viscosity Ecoflex silicone elastomer kit by Smooth-On which is often considered too soft for many applications. Ecoflex is also being used for developing innovative applications in the form of soft matter sensor[30], capacitive strain sensor[31], strain sensors for prosthetics[32], hyper elastic pressure sensors[33], radio frequency sensors[34], sensors for wearable electronics[35], flexible ultrasonic imaging membranes [36], wearable

pressure sensors based on piezo capacitance[37], wearable sensors for biomedical, healthcare[38], [39]. Though individually every formulation has a few remarkable properties, none of them have the ideal properties required to function as dielectric elastomer. Mechanical and electrical properties of these elastomers can be manipulated to a large degree either by changing the stoichiometry of the formulations or by blending two formulations with each other. A mixture of two formulations with contrasting properties, often results in a blend that is moderately reinforced with the properties of both formulations. Exploiting the synergy between two formulations with diverse properties is a proficient way to fabricate innovative silicone devices on research level. In the previous studies by Russo, S. et al. (2016)[40] and Park, S. et al. (2018)[41], the properties of the blends of some commercial formulations were investigated and it was concluded that by blending Dragon Skin with Sylgard 184, the extensibility and tear resistance of Sylgard 184 was considerably improved while retaining its capacity to plasma bond.

In the current work, popular commercial silicone formulations (Sylgard 184, Sylgard 186 and Ecoflex 00-50, Ecoflex 00-30, Ecoflex 00-10) are mixed at different stoichiometric ratios and are also blended with each other in order to tweak their properties. The formulations and blends are investigated to find the best suited formulation for elastomeric transducers. Optically transparent formulations are chosen for our study as they are sought after for many applications [9][42] especially for wearable invisible sensors. Ecoflex silicones are chosen because of their low viscosity and low Young's modulus. Furthermore, the properties of the blends and mixtures are mapped to guide the silicone research community to pick and choose a formulation that best suits the intended application.

2. EXPERIMENTAL

2.1 Materials

Five different two-part (A and B) commercial silicone elastomers were used. Sylgard 184 and 186 silicone elastomers were provided by Dow, Germany. Silicone rubbers Ecoflex 00-50, 00-30 and 00-10 were provided by FormX, Netherlands.

2.2 Sample preparation

Sample compositions are shown in Table 1. Parts A and B of every silicone formulation were weighed according to the mixing ratio and mixed in a SpeedMixer DAC 150 FVZ-K for 2 minutes at 2000 rpm. No additional degassing step is required while using a speed mixer. The manufacturer recommended mixing ratios of Sylgard 184 and 186 of 10:1 (part A: part B) and for Ecoflex 00-50, 00-30 and 00-10 the mixing ratio is 1:1 (part A: part B). Depending on the experiment type, mixed compositions were processed in different ways. All measurements were carried out at room temperature and room humidity. No solvent or flow control agent was used.

To make films, formulations were doctor bladed on acetate/polyester support film, provided by Pütz GmbH + Co. Folien KG, Germany. Blade gap was 400 μm and 200 μm , respectively. All formulations were cured at 80 $^{\circ}\text{C}$ for 2 hours. After curing, film thicknesses were 300 $\mu\text{m} \pm 10\%$ and 100 $\mu\text{m} \pm 20\%$, respectively as the film thicknesses usually vary from the blade thickness [20]. 1 mm $\pm 20\%$ thick samples were prepared in a metal mold and 6.4 mm or thicker samples were prepared in a metal cup, both type of samples were cured at 80 $^{\circ}\text{C}$ for 2 hours.

2.3 Mechanical properties

Stress-strain measurements were carried out with Instron 3345 tensile tester equipped with a 500 N load cell. The effective samples dimensions were 5 cm x 2 cm x 300 μm . The applied crosshead velocity was 5 cm/min.

2.4 Breakdown strength

Breakdown tests were performed on an in-house-built instrument following the international standards (IEC 60243-1 (1998) and IEC 60243-2 (2001)) [43]. 100 $\mu\text{m} \pm 20\%$ thick film samples were placed on a plastic support during breakdown testing. The plastic frames with silicone film were placed into the breakdown instrument between two semi-spherical electrodes made of stainless steel. Subsequently, 100 V/s voltage ramp was applied until polymer film broke down. Breakdown strengths were calculated from Weibull analysis[44].

2.5 Dielectric permittivity

Novocontrol Broadband Dielectric Spectrometer, Germany was used to investigate dielectric properties. Disc samples (1 mm \pm 20% thick, 25 mm diameter) were placed between two gold electrodes. Samples were tested at room temperature in the frequency range from 10⁻¹ to 10⁶ Hz at 1 V.

Table 1: Sample list with corresponding sample names and mixing ratios.

No.	Sample name	Part I ^a	Part I by weight	Part II ^a	Part II by weight
1.	Sylgard 184 5:1	Sylgard 184 (5:1)	-	-	-
2.	Sylgard 184 10:1	Sylgard 184 (10:1)	-	-	-
3.	Sylgard 184 15:1	Sylgard 184 (15:1)	-	-	-
4.	Sylgard 184 20:1	Sylgard 184 (20:1)	-	-	-
5.	Sylgard 186 5:1	Sylgard 186 (5:1)	-	-	-
6.	Sylgard 186 10:1	Sylgard 186 (10:1)	-	-	-
7.	Sylgard MIX 1:3	Sylgard 184 (10:1)	1	Sylgard 186 (10:1)	3
8.	Sylgard MIX 1:1	Sylgard 184 (10:1)	1	Sylgard 186 (10:1)	1
9.	Sylgard MIX 3:1	Sylgard 184 (10:1)	3	Sylgard 186 (10:1)	1
10.	Ecoflex 00-50	Ecoflex 00-50	-	-	-
11.	Eco MIX 00-50 1:3	Sylgard 184 (10:1)	1	Ecoflex 00-50	3
12.	Eco MIX 00-50 1:1	Sylgard 184 (10:1)	1	Ecoflex 00-50	1
13.	Eco MIX 00-50 3:1	Sylgard 184 (10:1)	3	Ecoflex 00-50	1
14.	Ecoflex 00-30	Ecoflex 00-30	-	-	-
15.	Eco MIX 00-30 1:3	Sylgard 184 (10:1)	1	Ecoflex 00-30	3
16.	Eco MIX 00-30 1:1	Sylgard 184 (10:1)	1	Ecoflex 00-30	1
17.	Eco MIX 00-30 3:1	Sylgard 184 (10:1)	3	Ecoflex 00-30	1
18.	Ecoflex 00-10	Ecoflex 00-10	-	-	-
19.	Eco MIX 00-10 1:3	Sylgard 184 (10:1)	1	Ecoflex 00-10	3
20.	Eco MIX 00-10 1:1	Sylgard 184 (10:1)	1	Ecoflex 00-10	1
21.	Eco MIX 00-10 3:1	Sylgard 184 (10:1)	3	Ecoflex 00-10	1

^a Mixing weight ratio of part A and part B (A:B) of the silicone kits in brackets

3. RESULTS and DISCUSSION

The ultimate stress (σ_{\max} (MPa)), elongation at break ($\epsilon_{\text{tensile}}$ (%)), and breakdown strength (E_{BD} (V/ μm)) of all the formulations are plotted with their respective Y (MPa) in Figure 1. All the properties are also tabulated (see Table 1). The figure of merit for a dielectric elastomer actuator ($F_{\text{om(DEA)}}$) and the figure of merit for a dielectric elastomer generator ($F_{\text{om(DEG)}}$) are calculated and plotted in Figure 2. The properties of formulations which are relevant for stretchable electronics are discussed further below.

3.1 Mechanical properties/elasticity

The ultimate tensile strengths of all formulations are plotted with their respective Y in Figure 1 (a). For soft and stretchable devices, extensibility (Figure 1b) is a parameter of utmost importance[45]. Of all the formulations, Sylgard 184 has the least extensibility. By varying the mixing ratio of Sylgard 184 (Part A: Part B = 20:1) the extensibility is doubled (135% to 296%). SylgardMIX (1:3) shows improved extensibility (135% to 365%) over Sylgard 184. Though, Ecoflex 00-50, Ecoflex™ 00-30 have the highest extensibility of ~800% their blends with Sylgard 184 (Eco MIX 00-50 and Eco MIX 00-30) show a rather limited extensibility of up to only ~360%. The addition of a soft formulation to a hard one results in intermediate properties, as expected. The properties of the formulations indicate that blending is a quick and a surefire approach to make reliable soft networks with mechanical integrity as opposed to varying the stoichiometry, which, if done inappropriately, often leads to formation of weak networks [20].

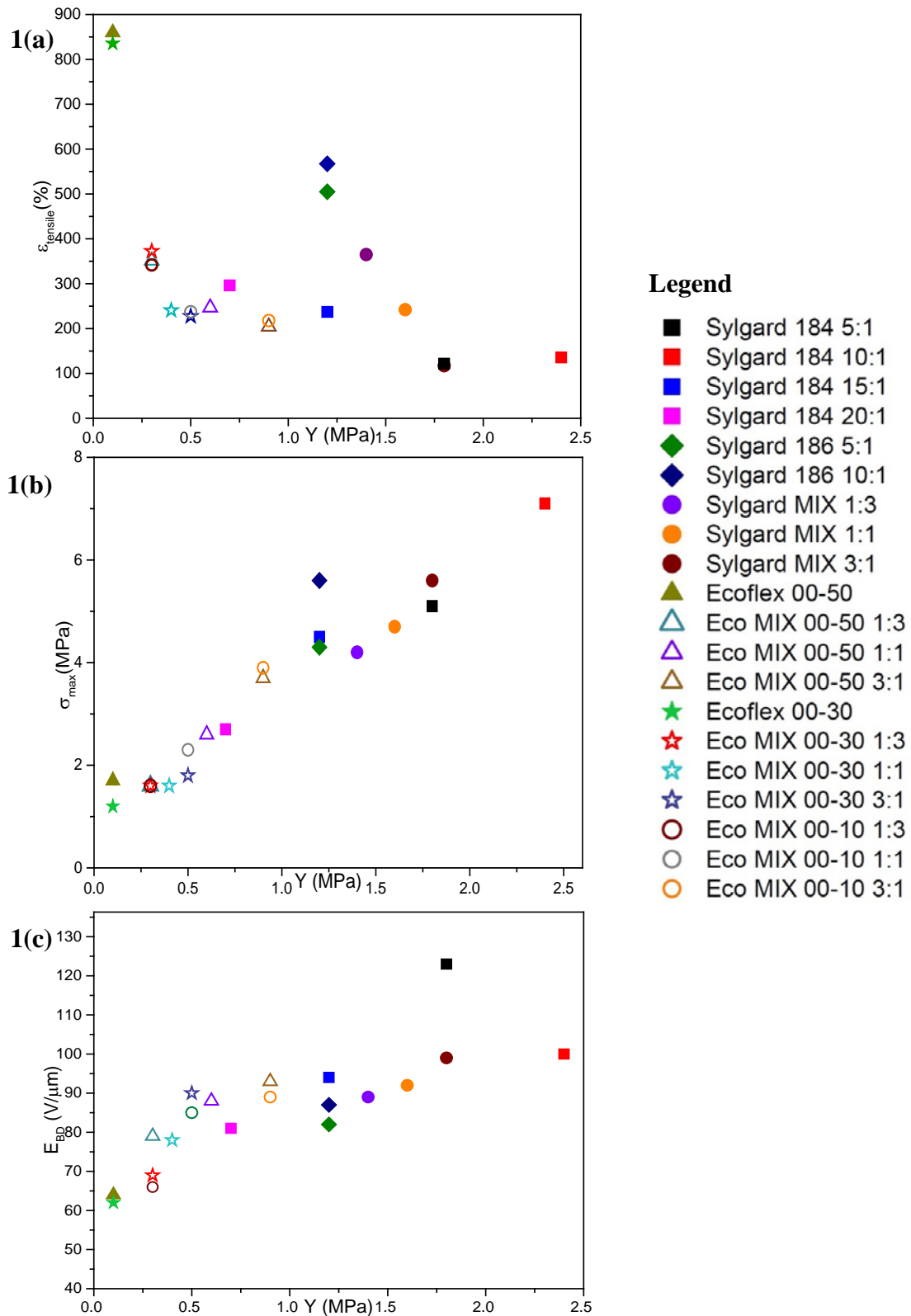


Figure 1: Various properties of investigated silicone elastomers plotted against the Young's modulus (Y). a) Ultimate stress Vs Y, b) Tensile strain vs. Y, c) Electrical breakdown vs. Y.

3.2 Electrical properties

When Sylgard 184 is mixed in the ratio of 5:1, the breakdown strength is improved from 100 to 123 V/μm (Figure 1(c)) compared to Sylgard 184 mixed in the ratio of 10:1. When Sylgard 186 is mixed in the ratio of 5:1, the dielectric permittivity improves from 2.8 to 3.2 compared to Sylgard 186 mixed in the ratio of 10:1. The blends resulting from the amalgamation of formulations with diverse properties, i.e. Eco MIX 00-50, Eco MIX 00-30, Eco MIX 00-10 benefit from being moderately reinforced by the high dielectric permittivity of Ecoflex™ and high breakdown strength of Sylgard 184. Compared to Sylgard 184; Eco MIX 00-50, Eco MIX 00-30 and Eco MIX 00-10 show improved dielectric permittivity and a slightly degraded breakdown strength. Researchers, can quickly and efficiently introduce better dielectric permittivity and breakdown strength into silicone formulations by fabricating such blends instead of using high dielectric constant fillers (e.g. titanium dioxide) which require tedious processing[46][47]. Furthermore, it was observed that all the investigated elastomers broke down electrically in similar ways by creating silica as a solid residual [48][49].

3.3 Figure of Merit

The $F_{om}(DEA)$ [50] and $F_{om}(DEG)$ [51] are designed to facilitate an easy evaluation of the performance of the elastomers as dielectric elastomer transducers. Mathematically, $F_{om}(DEA)$ and $F_{om}(DEG)$ are expressed as shown in equation(2) and equation(3).

$$F_{om}(DEA) = \frac{3\varepsilon'\varepsilon_0 E_{BD}^2}{Y} \quad (2)$$

$$F_{om}(DEG) = \frac{\varepsilon'\varepsilon_0 E_{BD}^2}{2\varphi} \quad (3)$$

Table 2: Properties of the different silicone formulations

Sample name	Mechanical properties			Dielectric properties	
	Y [MPa]	$\varepsilon_{tensile}$ [%]	σ_{max} [MPa]	ε' (1Hz)	E_{BD} [V/μm]
Sylgard 184 5:1	1,8	121,8	5,1	2,9	123
Sylgard 184 10:1	2,4	135,6	7,1	3,0	100
Sylgard 184 15:1	1,2	237,0	4,5	3,0	94
Sylgard 184 20:1	0,7	296,3	2,7	3,1	81
Sylgard 186 5:1	1,2	504,9	4,3	3,2	82
Sylgard 186 10:1	1,2	567,2	5,6	2,8	87
Sylgard MIX 1:3	1,4	365,0	4,2	2,8	89
Sylgard MIX 1:1	1,6	242,1	4,7	3,0	92
Sylgard MIX 3:1	1,8	117,1	5,6	3,2	99
Ecoflex™ 00-50	0,1	859,7	1,7	3,4	64
Eco MIX 00-50 1:3	0,3	351,4	1,6	3,3	79
Eco MIX 00-50 1:1	0,6	246,8	2,6	3,1	88
Eco MIX 00-50 3:1	0,9	204,4	3,7	3,1	93
Ecoflex™ 00-30	0,1	835,4	1,2	3,8	62
Eco MIX 00-30 1:3	0,3	372,6	1,6	3,2	69
Eco MIX 00-30 1:1	0,4	240,7	1,6	3,6	78
Eco MIX 00-30 3:1	0,5	227,6	1,8	3,2	90
Ecoflex™ 00-10	-	-	-	-	38
Eco MIX 00-10 1:3	0,3	341,6	1,6	4,0	66
Eco MIX 00-10 1:1	0,5	237,5	2,3	3,5	85
Eco MIX 00-10 3:1	0,9	217,9	3,9	3,4	89

Here, E_{BD} is the electrical field at which electrical breakdown occurs and ϕ is the strain energy function of the elastomer, ϵ_0 is the permittivity of free space ($8.854 \times 10^{-12} \text{ F}\cdot\text{m}^{-1}$) and ϵ' is the relative permittivity of our formulations. $F_{om}(\text{DEG})$ is comprehensive only when the strain energy function of the elastomer is considered. The strain energy density (ϕ) for our elastomers is calculated from their hysteresis curves[52] in the reverse extension cycle (15% to 0% strain) where the displacement is within the linear stress strain regime. Based on the F_{om} , (Figure 2(a) and 2(b)), Eco MIX 00-50 (1:3), Ecoflex 00-30, Eco MIX 00-10 (1:1) are the best formulations to function as stretchable transducers/electronics and by far exceed the benchmark Sylgard 184. In fact, all the formulations demonstrate an equal or better $F_{om}(\text{DEA})$ and $F_{om}(\text{DEG})$ than the rigid Sylgard 184. Although, $F_{om}(\text{DEA})$ and $F_{om}(\text{DEG})$ have a strong dependency on the breakdown strength of the formulations, the influence of Y and ϕ cannot be undermined as is evident from our calculations. Another issue to consider when transitioning from research to commercialization is the life-time. Research within this crucial topic is emerging and holds great promise for the DEG technology[53]–[58].

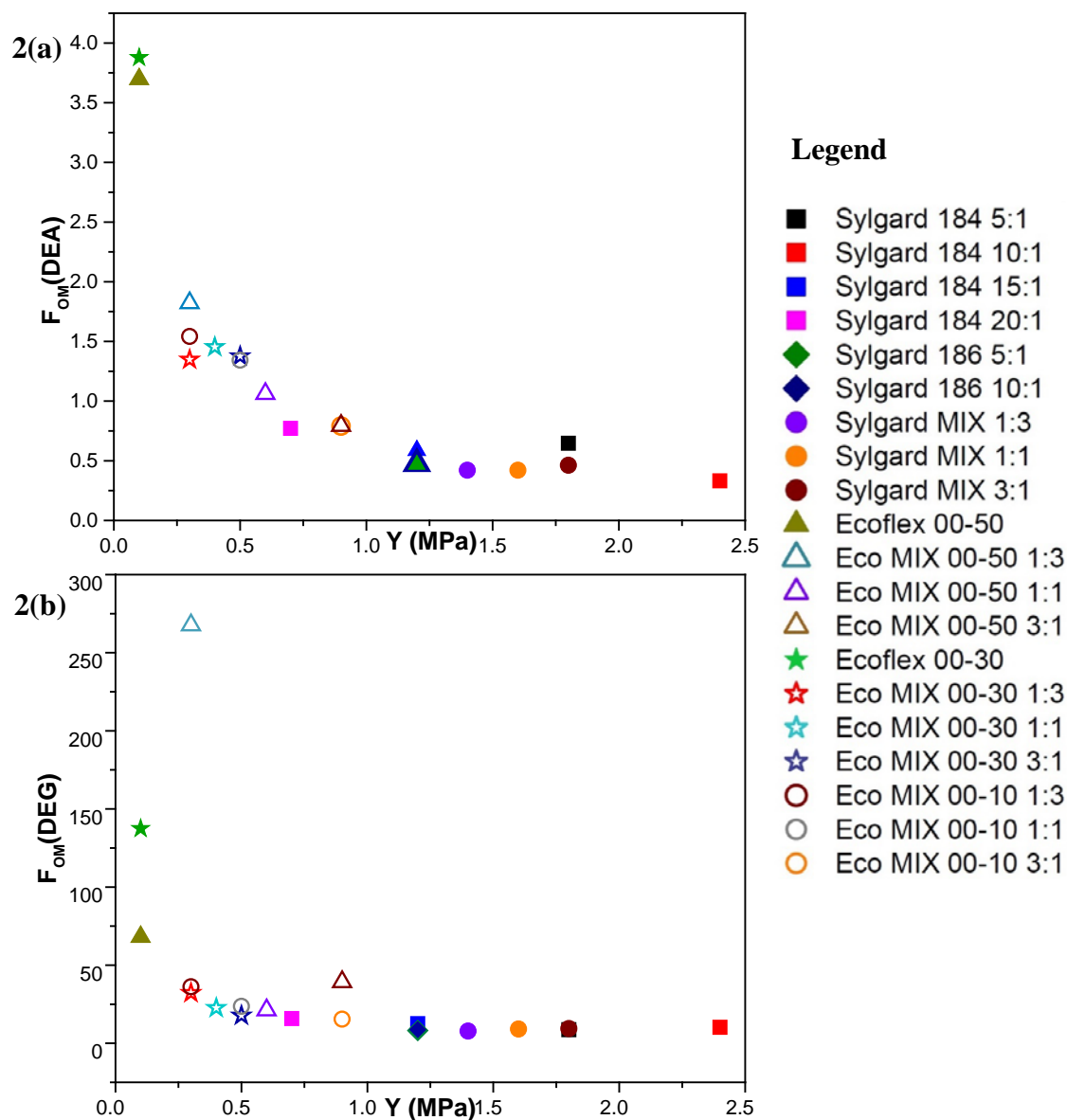


Figure 2: Figure of merit for an actuator ($F_{om}(\text{DEA})$) (a) and a generator ($F_{om}(\text{DEG})$) (b). For the calculations ϵ' measured at 1 Hz and ϕ measured from 15 % to 1 % strain were used.

4. CONCLUSIONS

We formulate 21 different recipes by combining some commonly used commercial silicone formulations (Sylgard 186, Sylgard 184 and Ecoflex 00-50, 00-30 and 00-10) at different stoichiometric ratios and also by blending two different formulations with each other. By doing so, we establish that the properties can be manipulated suitably. Furthermore, the electrical and mechanical properties of these blends are investigated and mapped with an objective to find those that are suitable for the fabrication of soft and stretchable transducers. The electrical and mechanical properties of the formulations are mapped and their figure of merit is also calculated. By combining Ecoflex 00-50 and Sylgard 184, a highly extensible elastomer blend with optimum dielectric permittivity and breakdown strength is obtained which qualifies as a good candidate for fabricating transducers based on the figure of merit. This synergy of two formulations with different properties can act to our advantage by providing a blend that is suitably reinforced with desired properties. These formulations can act as a good starting point for developing advanced hi-tech, versatile and stretchable devices.

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