Graphene inclusion controlling conductivity and gas sorption of metal-organic framework -
DTU Orbit (11/11/2019)

A general approach to prepare composite films of metal-organic frameworks and graphene has been developed. Films of copper(ii)-based HKUST-1 and HKUST-1/graphene composites were grown solvothermally on glassy carbon electrodes. The films were chemically tethered to the substrate by diazonium electrografting resulting in a large electrode coverage and good stability in solution for electrochemical studies. HKUST-1 has poor electrical conductivity, but we demonstrate that the addition of graphene to HKUST-1 partially restores the electrochemical activity of the electrodes. The enhanced activity, however, does not result in copper(iii) to copper(i) reduction in HKUST-1 at negative potentials. The materials were characterised in-depth: microscopy and grazing incidence X-ray diffraction demonstrate uniform films of crystalline HKUST-1, and Raman spectroscopy reveals that graphene is homogeneously distributed in the films. Gas sorption studies show that both HKUST-1 and HKUST-1/graphene have a large CO₂/N₂ selectivity, but the composite has a lower surface area and CO₂ adsorption capacity in comparison with HKUST-1, while CO₂ binds stronger to the composite at low pressures. Electron paramagnetic resonance spectroscopy reveals that both monomeric and dimeric copper units are present in the materials, and that the two materials behave differently upon hydration, i.e. HKUST-1/graphene reacts slower by interaction with water. The changed gas/vapour sorption properties and the improved electrochemical activity are two independent consequences of combining graphene with HKUST-1.

General information
Publication status: Published
Organisations: Department of Chemistry, Centre for Catalysis and Sustainable Chemistry, Organic Chemistry, Aarhus University
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Pages: 13921-13932
Publication date: 2018
Peer-reviewed: Yes

Publication information
Journal: R S C Advances
Volume: 8
Issue number: 25
ISSN (Print): 2046-2069
Ratings:
BFI (2018): BFI-level 1
Scopus rating (2018): CiteScore 3.16 SJR 0.807 SNIP 0.785
Web of Science (2018): Impact factor 3.049
Web of Science (2018): Indexed yes
Original language: English
Electronic versions:
c8ra02439a.pdf
DOIs:
10.1039/c8ra02439a

Bibliographical note
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Source: FindIt
Source ID: 2418734783
Research output: Contribution to journal › Journal article – Annual report year: 2018 › Research › peer-review