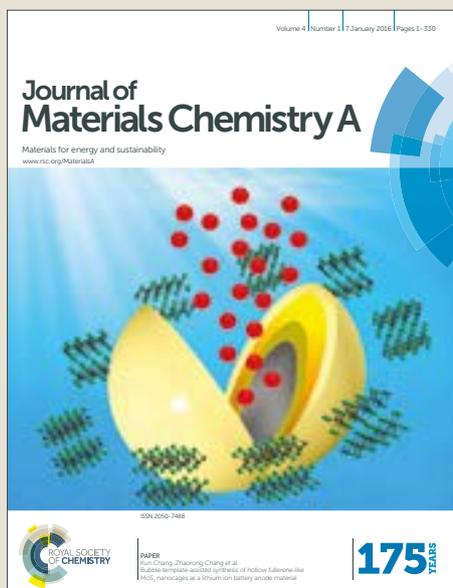


# Journal of Materials Chemistry A

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## Journal of Materials Chemistry A

## Communication

## Shear-aligned Graphene Oxide Laminate/Pebax Ultrathin Composite Hollow Fiber Membranes Using a Facile Dip-coating Approach

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Graphene oxide (GO) has been considered as a promising candidate for molecular separation because of its capability to form highly efficient gas flow intergalleries. However, a major challenge is the lack of a facile, scalable, low-cost membrane fabrication approach, especially for the hollow fiber composite membrane with a thin selective layer. By using a facile dip-coating technique, we prepared a shear-aligned GO/Pebax composite hollow fiber membrane on a porous polymeric support. The incorporation of the aligned GO laminates significantly improved the original Pebax (polyethylene oxide – polyamide block copolymer) membrane permeance without compromising the CO<sub>2</sub>/N<sub>2</sub> selectivity, and the composite membrane had good operational stability and enhanced mechanical strength. We further investigate the formation mechanism of the shear-aligned GO structure and discovered the control of the withdraw speed and liquid film thickness was a key factor. This dip-coating technique offers significant opportunities to exploit the GO-based membrane for industrial CO<sub>2</sub> capture.

### Introduction

Graphene oxide (GO) is a two-dimensional material with distinct structural, chemical and electrical properties. It offers a wide-ranging opportunity for developing a new family of molecular separation membranes.<sup>1</sup> The separation mechanisms of GO-based membranes arise from 1) pores on GO sheets; 2) in-plane slit-like pores; and 3) intergalleries between GO sheets. Precise control of the perforation and slit-like pores still remains a technical challenge. Therefore, most researchers apply physical or chemical approaches to regulating the intergalleries for the separation of small molecules.<sup>2</sup> Currently, the preparation of pure GO membranes is mostly based on the deposition of GO onto porous supports with external forces (e.g. pressure, centrifugal or capillary forces).<sup>3, 4</sup> However, it has been challenging to precisely tune the laminate arrangements especially over large surface areas. This leads to random plane-to-plane stacking and non-selective defects. Recently, a centrifugation-filtration synergetic approach was applied to precisely manipulate the GO intergallery channels. The resultant GO membrane had highly ordered laminate structure for ultrafast gas sieving.<sup>2</sup> However, this fabrication approach requires simultaneous manipulation of spin coating and filtration, and thus making the

scale-up difficult. In another approach, the GO suspension was firstly concentrated to a discotic nematic phase, then casting-aligned to form an anisotropic membrane.<sup>5, 6</sup> However, for the pure GO membranes, there are still concerns about their structural stability, especially in an aqueous environment. For example, the water-induced swelling effect can change the densely-packed GO laminate structures and subsequently compromise the molecular sieving capabilities. As a result, it is preferable to fabricate GO/polymeric mixed matrix membranes.

Polymeric membranes currently dominate the gas separation market due to the ease of processing and low cost.<sup>7-9</sup> So far, only a few successful attempts have been reported to blend GO into a polymer matrix for efficient gas separation membrane fabrication. Shen et al.<sup>10</sup> utilized the favorable molecular interactions between GO and polymer to assemble GO into well-defined GO laminates within flat sheet membrane matrix. The molecular sieving interlayer spacing and straight diffusion pathways facilitated the efficient gas transport through the membrane. In another line of research, Berean et al.<sup>11</sup> blended GO into polydimethylsiloxane (PDMS) membrane matrix. Due to the relatively weak interaction between GO and polymer, the interfacial voids provided fast gas transport channels. However, most existing GO/polymer mixed matrix gas separation membranes are relatively thick flat sheet membranes (>50 μm). The formation of thin (~1 μm), continuous GO/polymer film on hollow fiber support is preferable to promote the practical gas permeance for practical application. Considering the dimension of GO sheets can be tens of microns, the randomly oriented GO nanosheets within the thin polymeric film are prone to create non-selective defects and compromise the gas separation efficiency. At

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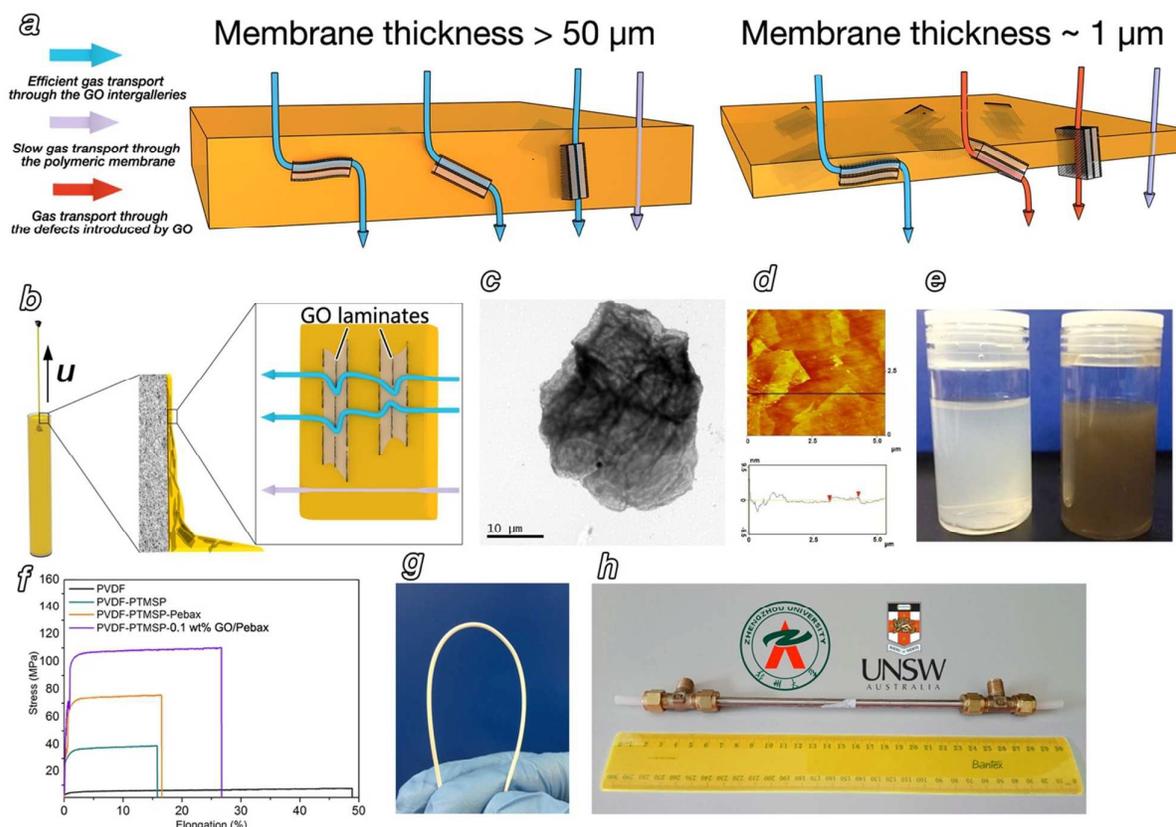


Figure 1. Preparation of GO/polymer membrane for gas separation. a) Schematic representation of the thick and thin mixed matrix membrane. b) Formation of the thin selective layer via the dip-coating process. c,d) TEM and AFM of the GO nanosheets. e) Optical image of the Pebox solution (left) and the GO/Pebox solution (right). f) Tensile testing of the membrane. g,h) Optical image of the prepared membrane and membrane module.

the same time, it is still challenging to control the orientation of GO laminates within the thin GO/polymer layer (Figure 1a).

Dip coating is a facile approach to introducing a composite membrane with a thin selective layer. It allows easier control and scale-up, and can be integrated with the existing membrane fabrication apparatus for industrial production.<sup>12, 13</sup> We have previously prepared a composite hollow fiber gas separation membrane via the dip-coating technique.<sup>14</sup> A thin, continuous Pebax-1657 (Figure S1) selective layer was deposited onto polyvinylidene fluoride (PVDF) membrane surface. We postulated that the surface tension, capillary effect, and gravity during the dip-coating process would facilitate the GO sheets alignment within the thin selective layer (Figure 1b). Hence, we explored the formation of a thin GO/Pebox composite layer on a hollow-fiber PVDF membrane with a pore size of 0.05 μm.

## Results and discussion

GO nanosheets in this work were prepared via the modified Hummers' method.<sup>15</sup> This approach generates an oxidized graphite material that can be sonicated to form a well-dispersed stable aqueous GO suspension. The presence of hydroxyl, carboxyl, carbonyl groups on GO is confirmed by the FT-IR results (Figure S2).

The feature diffraction peak of GO appears at  $10.4^\circ$ , indicating the increase of interlayer distance to 0.85 nm compared with the graphite ( $2\theta=26.6^\circ$ , d-spacing 0.34 nm). This can be attributed to the presence of functional groups, especially the carboxyl groups with intrinsic repulsive electrostatic forces (Figure S3).<sup>2, 16</sup> The in-house prepared GO has a dimension of ca. 1-2 μm up to 20-30 μm with a thickness of ca. 1-1.2 nm (Figure 1 c-d) and is in good agreement with previous studies.<sup>2, 15</sup> Seeking to understand the effect of GO concentration, 0.05, 0.1 and 0.2 wt % of GO were dispersed into 3 wt % Pebax solution (solvent: water/ethanol=30/70). The resultant suspension solutions are homogeneous based on visual observation, and they are stable at room temperature due to the formation of hydrogen bonds between GO and Pebax (Figure 1e),<sup>10</sup> and the stable suspension facilitates the formation of an even coating layer on the supporting membrane surface. The virgin PVDF and coated PVDF membranes show elastic behavior at a low elongation rate, and the incorporation of GO shows a striking improvement of Young's modulus for the composite membrane, which originated from both the mechanical strength of GO and the good interfacial compatibility between GO and Pebax polymers (Figure 1f). This result also indicates the composite membrane has good flexibility and enhanced mechanical properties (Figure 1g-h). Such a property

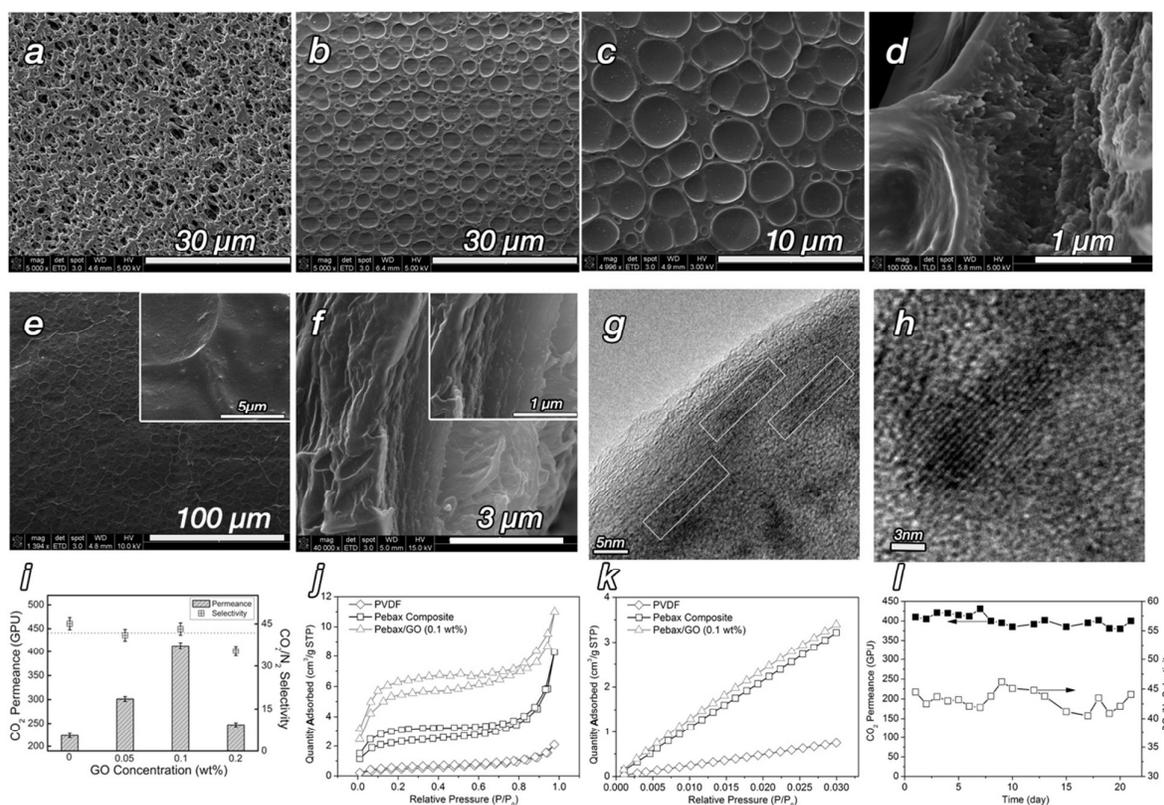


Figure 2 Performance of the GO/Pebax hollow fiber membrane. SEM images of a) original PVDF membrane, b) PTMSP coated membrane, c) pure Pebax coated membrane, d) cross-section of pure Pebax coated membrane, e) GO/Pebax coated membrane, f) cross-section of GO/Pebax coated membrane (insert: enlarged image). g,h) cross-section TEM images of the GO/Pebax membrane (with highlighted GO). i) Membrane performance with different GO concentration. j,k) N<sub>2</sub> and CO<sub>2</sub> adsorption isotherms. l) Long-term performance of the membrane.

is preferable for large-scale manufacture of the composite membrane.

In order to avoid the penetration of coating layer into the pores, the PVDF membrane was initially coated with poly[1-(trimethylsilyl)-1-propyne] (PTMSP, Figure S1) as a highly permeable gutter layer. After the PTMSP coating, the membrane surface is smoother yet still has some micron-scale shallow pits possibly due to the condensation of water vapour during the drying process (Figure 2a-c). The thickness of the PTMSP gutter layer is approximately 5 μm (Figure S4). Further two cycles of Pebax coating increased the thickness by around 1 μm, suggesting the thickness of the Pebax (or GO/Pebax) selective layer is in the order of 1 μm. However, it should be noted that it is difficult to accurately determine the thickness of Pebax layer due to the absence of a clear PTMSP/Pebax interface (the selective layer and the gutter layer are interfused together) (Figure S4). It can be attributed to the re-dissolution of PTMSP during the Pebax coating process (water/ethanol solvent). Such an observation is in agreement with other dip-coated multi-layer composite membranes that the exact thickness of the selective layer is difficult to characterize.<sup>17,18</sup> In this work, we also attempt to investigate the effect of GO content on the selective layer thickness and the results suggest membranes with different GO content have similar overall thickness (±0.2 micron), as the GO concentrations applied in this work is relatively low (up to 0.2 wt %).

After the addition of GO/Pebax coating, the membrane surface is still largely smooth, with only a few minor wrinkles (Figure 2e) indicating that GO laminates are mostly parallel to the membrane surface. This shear aligned multi-layer GO laminates are confirmed by the cross-sectional SEM and TEM images (Figure 2f-h). Due to the confinement effects of the Pebax chains and the hydrogen bonding with surrounding polymers, GO nanosheets form stacked laminates with d-space of ~0.7 nm (intergallery distance of ~0.35 nm considering the thickness of GO, in between the molecular kinetic diameters of CO<sub>2</sub> (0.33 nm) and N<sub>2</sub> (0.36 nm)). It should be noted that the d-space is determined from the TEM images as the GO peak in the composite membrane XRD result is too weak to be determined. The intergallery distance is calculated by deducting the GO laminate thickness from the d-space value. Such structure is in line with previous research and beneficial for size exclusion gas separation.<sup>19,20</sup>

Figure 2i shows the incorporation of GO within Pebax significantly improves the CO<sub>2</sub> permeance (up to 90 %) without losing the selectivity. However, further increase of the GO concentration (to 0.2 wt%) leads to the decrease of both permeance and selectivity. When the GO content is relatively high in the coating solution, the non-permeable 2D layers can disrupt the solvent evaporation and subsequently perturb the alignment of GO, which results in

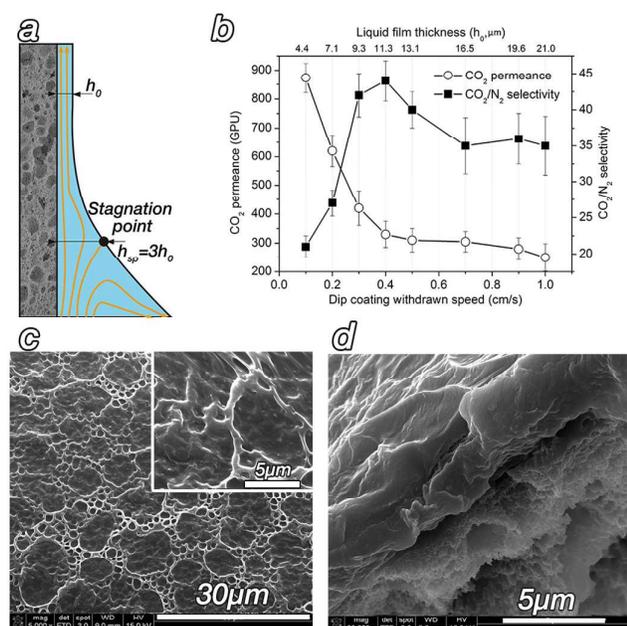


Figure 3. Formation of the parallel-aligned GO laminates. a) Schematic diagram of the liquid film and meniscus. b) Membrane performance with different withdrawn speed (0.1 wt% GO). c, d) SEM image of the membrane prepared with 1 cm/s withdrawn speed

more tortuous gas transport pathway and defects within the thin GO/polymer layer. The gas permeation through the membrane is determined by its solubility and diffusivity and both highly selective chemical affinities and molecular sieving effects are preferable for fast and selective gas separation.<sup>21, 22</sup> We then conducted gas adsorption test to further explore the function of GO in gas separation. The incorporation of GO improves the nitrogen adsorption for the composite membrane due to the large surface area of GO. But it only marginally increases the  $\text{CO}_2$  uptake. Due to the presence of functional groups such as  $-\text{COOH}$  and  $-\text{OH}$  on GO surface, it usually exhibits preferential adsorption of polar gasses such as  $\text{CO}_2$ .<sup>3, 23, 24</sup> However, the formation of hydrogen bonds between GO and Pebax limits the preferential adsorption of  $\text{CO}_2$  onto the GO laminates.<sup>25, 26</sup> In addition, it can also rigidify the Pebax polymeric chains and reduce the gas permeability. This observation suggests the improvement in  $\text{CO}_2$  permeation did not originate from the solution/diffusion mechanism for the Pebax/GO membrane. Instead, considering the intergallery distance of  $\sim 0.35$  nm for the GO laminates which is larger than  $\text{CO}_2$  kinetic diameter (0.33 nm), we postulated that by introducing size-selective channels the parallel-aligned GO laminates create highly efficient gas transport channels without leading to extra defects within the thin composite layer. The presence of the GO laminates would provide more rapid pathways compared to the relatively slower diffusion through the bulk Pebax polymer.<sup>24, 27-30</sup> All these aspects lead to the improved membrane performance after the incorporation of GO even the gas transport pathways are more tortuous. We further examined the long-term operational stability of the composite membrane and the result suggests good stability for the membrane (Figure 2i). The aging of PTMSP can lead to the loss of permeance overtime.<sup>31</sup> However, no clear loss of permeance is observed in this

work. The GO/Pebax layer partially fused within the PTMSP gutter layer, suggesting the intertwining of their polymer chains, which stabilize the non-equilibrium PTMSP structure. On the other hand, we also conducted the dip-coating of pure GO (0.1 wt% in ethanol/water solvent) onto the PTMSP pre-coated membrane, and the gas permeation results shows it has negligible change before and after the pure GO coating ( $\text{CO}_2$  3,570 GPU and  $\text{CO}_2/\text{N}_2$  selectivity of 4.5 for PTMSP gutter layer coated membrane vs.  $\text{CO}_2$  3,250 GPU and  $\text{CO}_2/\text{N}_2$  selectivity of 4.6 for GO/PTMSP coated membrane), indicating the GO coating layer is not continuous. This observation can be attributed to the weak affinity of GO to PTMSP, which leads to GO detachment during the repeated coating process.

The shear-induced alignment of nanowires, carbon nanotubes and stripes on substrate surface has been reported previously.<sup>32-34</sup> But this has not been demonstrated with the multi-layer GO laminates for molecular sieving. During the dip coating process, the viscous forces near the surface entrain the adjacent fluid upward. The entrainment effect forms a meniscus and the material above the stagnant point are carried into the liquid film on the substrate surface (Figure 3a). The thickness of the liquid layer is defined by the withdrawn speed, liquid viscosity, and surface tension. The capillary number  $\text{Ca}_0$  in this work was lower than 1 (within the Newtonian regime, Section 2.3 in Supporting information), thus the liquid film thickness can be calculated by the Landau-Levich equation.<sup>35</sup> The liquid film thickness with 0.1 wt% GO solution and 0.3 cm/s withdrawn speed is  $\sim 9$   $\mu\text{m}$  (Figure S5), which is comparable to the lateral GO dimensions in this work. As a result, the meniscus acts to align the GO laminates during the dip-coating process, mainly via the strong deposition force from receding meniscus by gravity. In order to confirm this speculation, we further prepared a series of membranes with different withdraw speeds. Figure 3b depicts that for the membrane containing 0.1 wt% GO, with the increase of withdrawn speed, the  $\text{CO}_2$  permeance decreases, which can be explained by the thicker liquid film and eventually thicker selective layer. However, at low withdraw speed ( $< 0.3$  cm/s) the very thin coating may introduce some defects. On the other hand, high withdraw speed also leads to the loss of selectivity. As shown in the SEM images, the GO laminates lose the parallel structure under high withdrawn speeds (Figure 3c-d). This corrugated surface structure can introduce extra defects between randomly orientated GO laminates. The higher withdraw speed increased the liquid film thickness. In this work, the speed of 1.0 cm/s results in a liquid film of  $\sim 22$   $\mu\text{m}$ , which is comparable to or higher than the average GO dimension, allowing the random orientation of GO sheets within the liquid layer. On the other hand, thicker liquid requires a longer time for solvent evaporation. The constant convection force towards the liquid surface during the prolonged evaporation process has been discovered to lead to the heterogeneous GO structures.<sup>36</sup> We also investigated the effect of dip-coating withdrawn speed for membrane containing a relatively high content of GO (0.2 wt%). The result (Figure S6) depicts the aggregation of GO can disrupt the densely packed GO structure and degrade the gas separation performance, suggesting the concentration of GO plays an important role in forming the alignment structure.

The performances of composite hollow fiber membranes are summarized in Table 1. Normally, lower membrane selectivity is anticipated with the ultrathin composite membranes compared with homogeneous dense membranes.<sup>37</sup> Compared with the literature values, the composite membranes containing aligned GO have the highest CO<sub>2</sub>/N<sub>2</sub> selectivity due to the alignment of GO interlayer galleries, as well as the good compatibility between GO nanosheets and the polymeric matrix. Even though the PDMS/PAN composite membranes exhibited a much higher CO<sub>2</sub> permeance but the selectivity was significantly lower than the composite membrane in this work.<sup>38</sup> The GO/Pebax composite membrane also outperformed our previous work of ZIF-8/Pebax-based composite membranes, where the incorporation of ZIF-8 nanoparticles into the thin Pebax selective layer creates extra defects.<sup>37</sup>

Table 1 Comparison of hollow fiber composite membranes for CO<sub>2</sub>/N<sub>2</sub> separation

Membrane type	Testing T/P (°C/MPa)	Permeance (GPU)		Selectivity CO <sub>2</sub> /N <sub>2</sub>	Ref.
		N <sub>2</sub>	CO <sub>2</sub>		
Matrimid/PES	25/0.39	0.76	60	39	<sup>39</sup>
PIM-1/Matrimid	25/0.1	9.2	235	25.5	<sup>40</sup>
6FDA-DAM-DABA	30/0.23	21.7	520	24	<sup>41</sup>
Pebax /PSf	25/0.2	2	61	30	<sup>42</sup>
PDMS/PAN	25/0.2	112	1211	10.8	<sup>38</sup>
Pebax/ZIF-8/PVDF	25/0.2	10.9	345	31.7	<sup>37</sup>
Pebax/GO/PVDF	25/0.2	9.6	413.3	43.2	This work

## Conclusions

In conclusion, we report here that shear-aligned GO laminates can be achieved within a thin polymer layer by a facile dip coating process. Control of liquid film thickness appears crucial to achieving alignment of GO laminates within the coating. The incorporated GO laminates provide highly efficient gas transport pathway without introducing extra defects to the thin layer. In general, this technique provides a feasible pathway for large-scale production of GO-polymer composite membrane for industrial CO<sub>2</sub> separation, and potentially offers an opportunity to exploit the unique properties of regulating the structure of other similar 2D materials within a thin composite layer to realize their unique properties in separation, sensing and catalysis.

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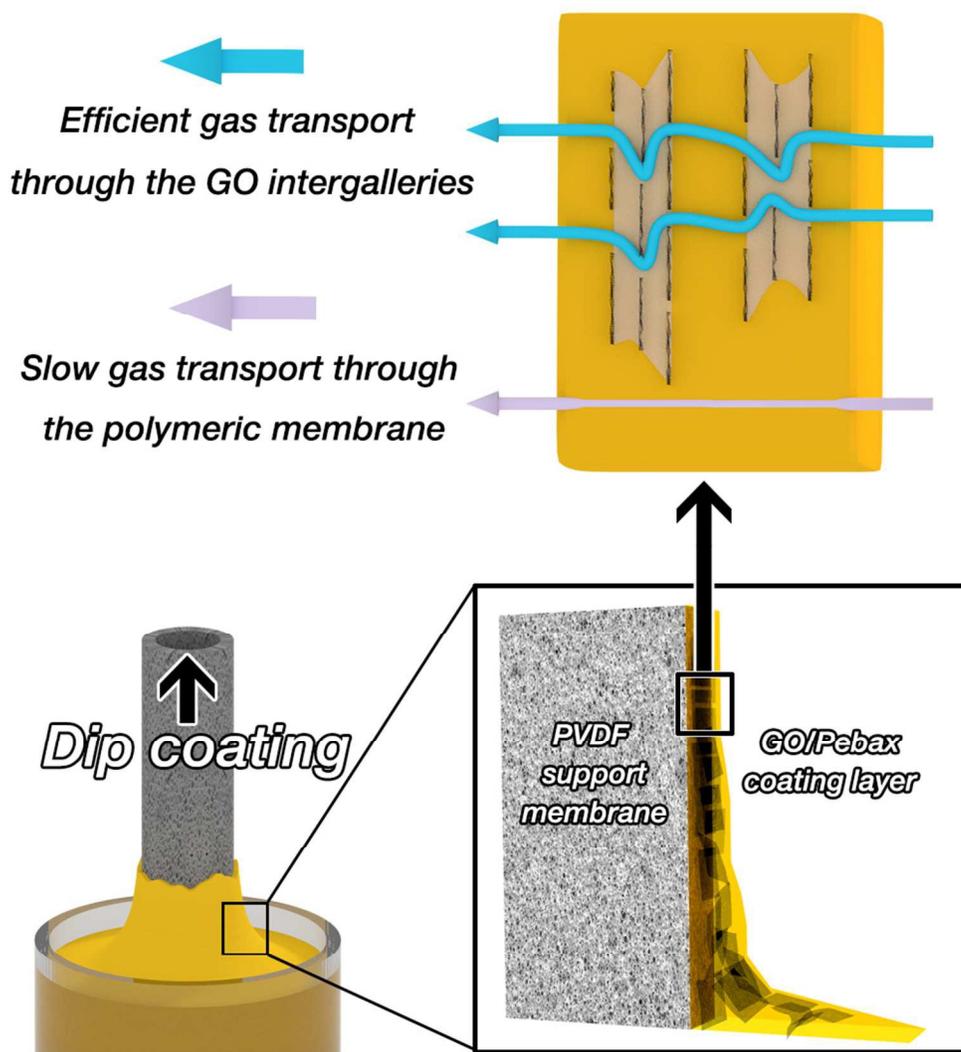
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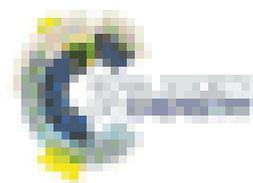
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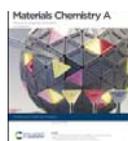
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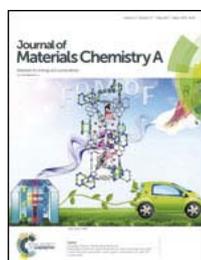
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## Journal of Materials Chemistry A

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### Review Article

## Rechargeable zinc-air batteries: a promising way to green energy

Peng Gu, Mingbo Zheng, Qunxing Zhao, Xiao Xiao, Huaiguo Xue and Huan Pang

Rechargeable zinc-air batteries show great potential in applications such as electric vehicles and wearable devices, especially for the flexible design. And the challenges and functional materials for each component are provided and discussed from air electrode, solid-state electrolyte to zinc anode, with perspectives of research directions.

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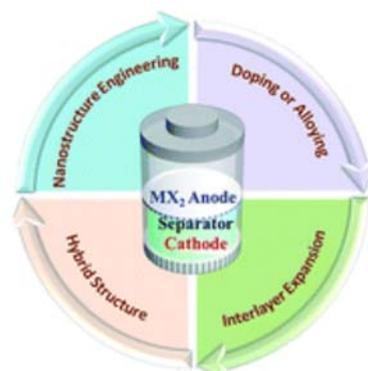
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#### Review Article

### Recent progress in layered metal dichalcogenide nanostructures as electrodes for high-performance sodium-ion batteries

Wenpei Kang, Yuyu Wang and Jun Xu

The sodium storage performance of layered metal dichalcogenide anodes enhanced through nanostructure engineering, crystal structure modulation, doping/alloying and composite design is systematically reviewed.



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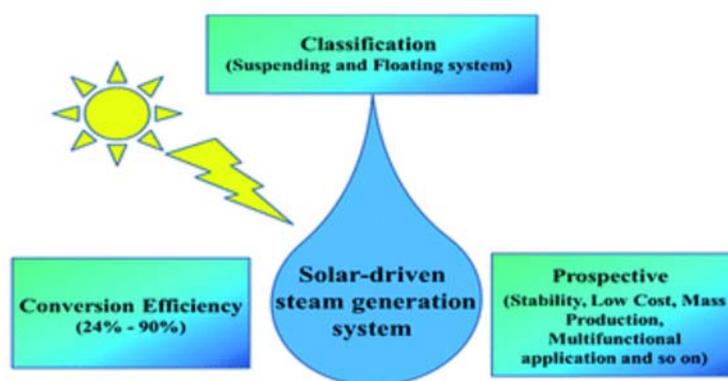
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Review Article

## The emergence of solar thermal utilization: solar-driven steam generation

Ziyang Deng, Jianhua Zhou, Lei Miao, Chengyan Liu, Ying Peng, Lixian Sun and Sakae Tanemura

This work mainly reviewed the classification, development, influencing factors and future prospects of solar-driven steam generation systems.



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The article was first published on 25 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7691-7709

<https://doi.org/10.1039/C7TA01361B>

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Review Article

## In pursuit of catalytic cathodes for lithium-oxygen batteries

Ali Eftekhari and Balaji Ramanujam

Li-O<sub>2</sub> batteries are among the most promising electrochemical energy storage systems, which have attracted significant attention over the past five years due to their potential to satisfy new demands such as powering electric vehicles.

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*J. Mater. Chem. A*, 2017, **5**, 7710-7731

<https://doi.org/10.1039/C7TA01124E>

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**Communication**

**Shear-aligned graphene oxide laminate/Pebax ultrathin composite hollow fiber membranes using a facile dip-coating approach**

Yatao Zhang, Qin Shen, Jingwei Hou, Putu Doddy Sutrisna and Vicki Chen

Graphene oxide (GO) has been considered as a promising candidate for molecular separation because of its capability to form highly efficient gas flow intergalleries within an ultrathin composite membrane.

The article was first published on 01 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7732-7737

<https://doi.org/10.1039/C6TA10395B>

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**Communication**

**A new Na[(FSO<sub>2</sub>)(*n*-C<sub>4</sub>F<sub>9</sub>SO<sub>2</sub>)N]-based polymer electrolyte for solid-state sodium batteries**

Qiang Ma, Juanjuan Liu, Xingguo Qi, Xiaohui Rong, Yuanjun Shao, Wenfang Feng, Jin Nie, Yong-Sheng Hu, Hong Li, Xuejie Huang, Liqun Chen and Zhibin Zhou

The NaFNFSI-based SPE can deliver the excellent interfacial stability with Na metal and good cycling performances for the Na|SPE|NaCu<sub>1/9</sub>Ni<sub>2/9</sub>Fe<sub>1/3</sub>Mn<sub>1/3</sub>O<sub>2</sub> cell.

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<https://doi.org/10.1039/C7TA01820G>

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**Communication**

**Experimental and theoretical insights into sustained water splitting with an electrodeposited nanoporous nickel hydroxide@nickel film as an electrocatalyst**

Zhicai Xing, Linfeng Gan, Jin Wang and Xiurong Yang

Electrodeposited nanoporous Ni(OH)<sub>2</sub>@Ni film acts as a Janus electrocatalyst for the hydrogen evolution and oxygen evolution reaction, and overall water splitting, exhibiting superior activities and exceptional long-term stability. Theoretical calculations show that the synergistic electrocatalytic effects of Ni(OH)<sub>2</sub> and Ni are responsible for the high activities.



The article was first published on 29 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7744-7748

<https://doi.org/10.1039/C7TA01907F>

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**Communication**

**Potassium-chemical synthesis of 3D graphene from CO<sub>2</sub> and its excellent performance in HTM-free perovskite solar cells**

Wei Wei, Baoyun Hu, Fangming Jin, Zhenzi Jing, Yuexiang Li, Andres Alberto García Blanco, Dario J. Stacchiola and Yun Hang Hu

The conversion of greenhouse gas CO<sub>2</sub> into novel materials is the most promising approach to solve greenhouse gas issues.



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Communication

**Atomic-scale topochemical preparation of crystalline Fe<sup>3+</sup>-doped β-Ni(OH)<sub>2</sub> for an ultrahigh-rate oxygen evolution reaction**

Kaiyue Zhu, Huanying Liu, Mingrun Li, Xuning Li, Junhu Wang, Xuefeng Zhu and Weishen Yang

Fe<sup>3+</sup>-doped β-Ni(OH)<sub>2</sub>, prepared *via* an atomic-scale topochemical transformation route, exhibits much higher oxygen evolution activity than the state-of-the-art IrO<sub>2</sub>.



The article was first published on 28 Mar 2017

**J. Mater. Chem. A**, 2017, **5**, 7753-7758

<https://doi.org/10.1039/C7TA01408B>

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Communication

## Microwave-synthesized tin oxide nanocrystals for low-temperature solution-processed planar junction organo-halide perovskite solar cells

Mutalifu Abulikemu, Marios Neophytou, Jérémy M. Barbé, Max L. Tietze, Abdulrahman El Labban, Dalaver H. Anjum, Aram Amassian, Iain McCulloch and Silvano Del Gobbo

Tin oxide nanoparticles prepared by microwave-assisted non-aqueous sol-gel were used to cast an electron transporting layer for organohalide perovskite solar cells showing, in average, high efficiencies thanks to the good optoelectronic properties of the material.



The article was first published on 25 Mar 2017

**J. Mater. Chem. A**, 2017, **5**, 7759-7763

<https://doi.org/10.1039/C7TA00975E>

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Communication

## Nano-structured hybrid molybdenum carbides/nitrides generated *in situ* for HER applications

Rajinder Kumar, Ritu Rai, Seema Gautam, Abir De Sarkar, Nidhi Tiwari, Shambhu Nath Jha, Dibyendu Bhattacharyya, Ashok K. Ganguli and Vivek Bagchi

A highly efficient, low-cost (precious-metal-free), highly stable nanohybrid electrocatalyst containing carbon-supported molybdenum carbide and nitride nanoparticles of size ranging from 8 to 12 nm exhibit excellent HER catalytic activity.



The article was first published on 21 Mar 2017

**J. Mater. Chem. A**, 2017, **5**, 7764-7768

<https://doi.org/10.1039/C7TA01815K>

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Communication

**A general approach to synthesise ultrathin NiM (M = Fe, Co, Mn) hydroxide nanosheets as high-performance low-cost electrocatalysts for overall water splitting**

Xiuhui Sun, Qi Shao, Yecan Pi, Jun Guo and Xiaoqing Huang

A general approach to ultrathin NiM (M = Fe, Co, Mn) hydroxide nanosheets as efficient electrocatalysts for overall water splitting.



The article was first published on 30 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7769-7775

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Communication

**Simultaneous enhancement of the molecular planarity and the solubility of non-fullerene acceptors: effect of aliphatic side-chain substitution on the photovoltaic performance**

Zhe Zhang, Miao Li, Yahui Liu, Jicheng Zhang, Shiyu Feng, Xinjun Xu, Jinsheng Song and Zhishan Bo

By incorporating aliphatic side chains and adjusting their lengths, non-fullerene acceptors can obtain an improved morphology and photovoltaic performance.



The article was first published on 03 Apr 2017

*J. Mater. Chem. A*, 2017, **5**, 7776-7783

<https://doi.org/10.1039/C7TA02141K>

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## metal-free electrocatalysts for overall water-splitting

Xin Yue, Shangli Huang, Junjie Cai, Yanshuo Jin and Pei Kang Shen

N and F dual doped porous graphene nanosheets (NFPGNS).



The article was first published on 04 Apr 2017

**J. Mater. Chem. A**, 2017, **5**, 7784-7790

<https://doi.org/10.1039/C7TA01957B>

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### Communication

## One-step CVD synthesis of carbon framework wrapped Co<sub>2</sub>P as a flexible electrocatalyst for efficient hydrogen evolution

Cui Ye, Min Qiang Wang, Guo Chen, Yang Hui Deng, Ling Jie Li, Hong Qun Luo and Nian Bing Li

Carbon framework wrapped cobalt phosphide on carbon cloth (Co<sub>2</sub>P@C/CC) was synthesized *via* one-step CVD as a flexible electrocatalyst for the HER.



The article was first published on 04 Apr 2017

**J. Mater. Chem. A**, 2017, **5**, 7791-7795

<https://doi.org/10.1039/C7TA00592J>

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### Communication

## Crystallographic orientation propagation in metal halide perovskite thin films

Alexander Z. Chen, Benjamin J. Foley, Jennifer H. Ma, Matthew R. Alpert, J. Scott Niezgoda and Joshua J. Choi

Preferential crystallographic orientation can propagate through metal halide perovskite thin

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The article was first published on 11 Apr 2017

**J. Mater. Chem. A**, 2017, **5**, 7796-7800

<https://doi.org/10.1039/C7TA02203D>

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#### Communication

### A fluorinated dialkoxide-based magnesium-ion electrolyte

Jake T. Herb, Carl A. Nist-Lund and Craig B. Arnold

A magnesium-ion battery electrolyte that results in high conductivity, 100% coulombic efficiency, and low overpotentials is presented. The fluorinated dialkoxide-based electrolyte is synthesized *via* a green route without the use of any metal alkyls.



The article was first published on 12 Apr 2017

**J. Mater. Chem. A**, 2017, **5**, 7801-7805

<https://doi.org/10.1039/C7TA01578J>

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#### Communication

### *In situ* formation of a 3D core/shell structured Ni<sub>3</sub>N@Ni–Bi nanosheet array: an efficient non-noble-metal bifunctional electrocatalyst toward full water splitting under near-neutral conditions

Lisi Xie, Fengli Qu, Zhiang Liu, Xiang Ren, Shuai Hao, Ruixiang Ge, Gu Du, Abdullah M. Asiri, Xuping Sun and Liang Chen

An *in situ* electrochemically developed core/shell structured Ni<sub>3</sub>N@Ni–Bi nanosheet array behaves as an efficient bifunctional catalyst for overall water splitting.

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The article was first published on 10 Apr 2017

*J. Mater. Chem. A*, 2017, **5**, 7806-7810

<https://doi.org/10.1039/C7TA02333B>

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#### Communication

### Molecular engineering of face-on oriented dopant-free hole transporting material for perovskite solar cells with 19% PCE

Kasparas Rakstys, Sanghyun Paek, Peng Gao, Paul Gratia, Tomasz Marszalek, Giulia Grancini, Kyung Taek Cho, Kristijonas Genevicius, Vygtintas Jankauskas, Wojciech Pisula and Mohammad Khaja Nazeeruddin

Dopant-free HTM KR321 showed highly ordered characteristic face-on organization leading to increased vertical charge transport and PCE over 19% in PSC with improved stability.



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*J. Mater. Chem. A*, 2017, **5**, 7811-7815

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#### Paper

### Rationally tuning host-guest interactions to free hydroxide ions within intertrimerically cuprophilic metal-organic frameworks for high OH<sup>-</sup> conductivity

Ziyin Li, Zhangjing Zhang, Yingxiang Ye, Kaicong Cai, Fenfen Du, Heng Zeng, Jin Tao, Quanjie Lin, Ying Zheng and Shengchang Xiang

Rationally tuning host-guest interactions to free hydroxide ions within intertrimerically cuprophilic MOF for high OH<sup>-</sup> conductivity.

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**J. Mater. Chem. A**, 2017, **5**, 7816-7824

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**Paper**

**MnO<sub>2</sub>/MnCo<sub>2</sub>O<sub>4</sub>/Ni heterostructure with quadruple hierarchy: a bifunctional electrode architecture for overall urea oxidation**

Changlong Xiao, Shuni Li, Xinyi Zhang and Douglas R. MacFarlane

A bifunctional core-shell structured MnO<sub>2</sub>/MnCo<sub>2</sub>O<sub>4</sub>/Ni electrode with quadruple-hierarchy has been developed for highly efficient overall urea electrolysis.



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**J. Mater. Chem. A**, 2017, **5**, 7825-7832

<https://doi.org/10.1039/C7TA00980A>

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**Paper**

**Highly efficient photocatalytic hydrogen production from pure water via a photoactive metal-organic framework and its PDMS@MOF**

Pengyan Wu, Min Jiang, Yang Li, Yanhong Liu and Jian Wang

Photoactive MOF modified with a thin PDMS layer exhibits high stability and ultrahigh photocatalytic efficiency for H<sub>2</sub> production in water.



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**Paper**

## Elucidating the diffusion pathway of protons in ammonium polyphosphate: a potential electrolyte for intermediate temperature fuel cells

Chunwen Sun, Carlos Alberto López and José Antonio Alonso

The diffusion pathway of protons in ammonium polyphosphate is revealed.



The article was first published on 28 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7839-7844

<https://doi.org/10.1039/C7TA01404J>

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 **Paper**

## Electronic and defect properties of $(\text{CH}_3\text{NH}_3)_2\text{Pb}(\text{SCN})_2\text{I}_2$ analogues for photovoltaic applications

Alex M. Ganose, Christopher N. Savory and David O. Scanlon

In this report, we study a range of 2-D perovskite-like compounds  $(\text{CH}_3\text{NH}_3)_2\text{MPs}_2\text{X}_2$  ( $\text{M} = \text{Sn}, \text{Pb}$ ;  $\text{Ps} = \text{OCN}, \text{SCN}, \text{SeCN}$  and  $\text{X} = \text{Cl}, \text{Br}, \text{I}$ ), finding suitable band gaps, band alignments and defect behaviour for photovoltaic top cells in tandem devices.



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*J. Mater. Chem. A*, 2017, **5**, 7845-7853

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**Paper**

## Phosphorus and oxygen dual-doped graphene as superior anode material for room-temperature potassium-ion batteries

Guangyao Ma, Kangsheng Huang, Jia-Sai Ma, Zhicheng Ju, Zheng Xing and Quan-chao Zhuang

The intercalation of potassium ions into graphitic carbon materials has been demonstrated to be feasible while the electrochemical performance of the potassium-ion battery (PIB) is still unsatisfactory.



The article was first published on 11 Apr 2017

*J. Mater. Chem. A*, 2017, **5**, 7854-7861

<https://doi.org/10.1039/C7TA01108C>

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**Paper**

## Fabrication of PEDOT films *via* a facile method and their application in Pt-free dye-sensitized solar cells

Yin-Chang Li, Song-Ru Jia, Zhi-Yan Liu, Xue-Qin Liu, Yang Wang, Ya Cao, Xiao-Qin Hu, Cheng-Long Peng and Zhen Li

PEDOT films with network-like structures aggregated by plenty of nanoparticles demonstrated good photovoltaic properties.



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The article was first published on 28 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7862-7868

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Paper

## Ruthenium-nickel-nickel hydroxide nanoparticles for room temperature catalytic hydrogenation

Lihua Zhu, Shiyao Shan, Valeri Petkov, Weiwei Hu, Anna Kroner, Jinbao Zheng, Changlin Yu, Nuowei Zhang, Yunhua Li, Rafael Luque, Chuan-Jian Zhong, Hengqiang Ye, Zhiqing Yang and Bing H. Chen

Ru nanoclusters on Ni/Ni(OH)<sub>2</sub> nanoparticles supported on carbon (Ru/Ni/Ni(OH)<sub>2</sub>/C) were successfully prepared and characterized. Ru/Ni/Ni(OH)<sub>2</sub>/C exhibited an unprecedented catalytic activity for naphthalene hydrogenation at room temperature due to synergetic catalysis.



The article was first published on 27 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7869-7875

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**Paper**

## High permselectivity hyperbranched polyester/polyamide ultrathin films with nanoscale heterogeneity

Xin Kong, Ze-Lin Qiu, Chun-Er Lin, You-Zhi Song, Bao-Ku Zhu, Li-Ping Zhu and Xiu-Zhen Wei

Incorporating hyperbranched polyesters into a cross-linked polyamide matrix by interfacial polymerization to construct an ultrathin film with high permselectivity performance.



The article was first published on 21 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7876-7884

<https://doi.org/10.1039/C7TA00246G>

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**Paper**

## FRET-guided surging of cyanobacterial photosystems improves and stabilizes current in photosynthetic microbial fuel cell

Sharbani Kaushik, Mrinal K. Sarma and Pranab Goswami

A quantum dot-based nano-biocomposite supports FRET to photosystems, biofilm growth and electron relay on the anode in a cyanobacteria-based fuel cell.



The article was first published on 24 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7885-7895

<https://doi.org/10.1039/C7TA01137G>

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L. dos Santos-Gómez, J. M. Porras-Vázquez, E. R. Losilla and D. Marrero-López

A  $\text{PrBaCo}_2\text{O}_{5+\delta}$  layered perovskite cathode prepared by an easy and economic spray-pyrolysis deposition method exhibits high performance and stability for low-temperature SOFCs.



The article was first published on 28 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7896-7904

<https://doi.org/10.1039/C6TA10946B>

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**Paper**

**Improved carriers injection capacity in perovskite solar cells by introducing A-site interstitial defects**

Pengjun Zhao, Wenping Yin, Minwoo Kim, Manhyung Han, Young Jae Song, Tae Kyu Ahn and Hyun Suk Jung

A new strategy to enhance perovskite solar cell performance by introducing potassium cation ( $\text{K}^+$ ) as interstitial site defects.



The article was first published on 25 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7905-7911

<https://doi.org/10.1039/C7TA01203A>

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**Paper**

**Redox chemistry of  $\text{CaMnO}_3$  and  $\text{Ca}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$  oxygen storage perovskites**

B. Bulfin, J. Vieten, D. E. Starr, A. Azarpira, C. Zachäus, M. Hävecker, K. Skorupska, M. Schmücker, M. Roeb and C. Sattler

Investigation of the redox chemistry and thermodynamics of the perovskites  $\text{CaMnO}_3$  and



The article was first published on 20 Mar 2017

**J. Mater. Chem. A**, 2017, **5**, 7912-7919

<https://doi.org/10.1039/C7TA00822H>

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**Paper**

**Novel thin-film composite membranes *via* manipulating the synergistic interaction of dopamine and *m*-phenylenediamine for highly efficient forward osmosis desalination**

Lina Xu, Jia Xu, Baotian Shan, Xiulin Wang and Congjie Gao

Novel thin-film composite (TFC) membranes were elaborately designed and fabricated *via* the incorporation of mussel-inspired dopamine (DA) into an *m*-phenylenediamine (MPD) aqueous solution on a macroporous substrate.



The article was first published on 27 Mar 2017

**J. Mater. Chem. A**, 2017, **5**, 7920-7932

<https://doi.org/10.1039/C7TA00492C>

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**Paper**

**Polyaniline nanoflowers grown on vibration-isolator-mimetic polyurethane nanofibers for flexible supercapacitors with prolonged cycle life**

Ali Khosrozadeh, Mohammad Ali Darabi, Quan Wang and Malcolm Xing

Employing a polyurethane nanofibrous membrane as a cushiony support for the deposition of flower-like polyaniline greatly improves the cycling stability of polyaniline pseudocapacitors.

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*J. Mater. Chem. A*, 2017, **5**, 7933-7943

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**Paper**

## Nitrogen-doped porous graphene as a highly efficient cathodic electrocatalyst for aqueous organic redox flow battery application

Jianguo Cao, Zhentao Zhu, Juan Xu, Meng Tao and Zhidong Chen

The redox flow battery (RFB) is considered one of the most attractive energy storage technologies because of its high efficiency, long service life and great safety.



The article was first published on 28 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7944-7951

<https://doi.org/10.1039/C7TA00873B>

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**Paper**

## Effects of adhesives on the electrochemical performance of monodisperse $\text{LiMn}_{0.8}\text{Fe}_{0.2}\text{PO}_4/\text{C}$ microspheres as cathode materials for high power lithium-ion batteries

Jianlong Li, Mingwu Xiang, Yan Wang, Jinhua Wu, Hang Zhao and Heng Liu

A facile co-precipitation approach combined with spray-drying and high-temperature calcinations was developed to synthesize  $\text{LiMn}_{0.8}\text{Fe}_{0.2}\text{PO}_4/\text{C}$  microspheres on a large scale.



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**Paper**

**Photoswitching storage of guest molecules in metal-organic framework for photoswitchable catalysis: exceptional product, ultrahigh photocontrol, and photomodulated size selectivity**

Le Le Gong, Wan Ting Yao, Zhi Qiang Liu, An Min Zheng, Jian Qiang Li, Xue Feng Feng, Lu Fang Ma, Chang Sheng Yan, Ming Biao Luo and Feng Luo

We present a novel MOF catalyst, which shows both ultrahigh photocontrol in catalytic activity and selectivity.



The article was first published on 28 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7961-7967

<https://doi.org/10.1039/C7TA01388D>

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**Paper**

## High area-specific capacitance of Co(OH)<sub>2</sub>/hierarchical nickel/nickel foam supercapacitors and its increase with cycling

Zheyin Yu, Zhenxiang Cheng, Xiaolin Wang, Shi Xue Dou and Xiangyang Kong

Supercapacitors are important energy storage systems due to their high power densities compared to batteries, giving them unique applications.



The article was first published on 28 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7968-7978

<https://doi.org/10.1039/C7TA00719A>

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**Paper**

## An efficient microbial fuel cell using a CNT-RTIL based nanocomposite

Leila Mahrokh, Hedayatollah Ghourchian, Kenneth H. Neelson and Mohammad Mahrokh

A woven carbon fiber filament coated with a nanocomposite consisting of amine functionalized multi-walled carbon nanotubes and a room temperature ionic liquid was used as the anode and cathode.



The article was first published on 15 Mar 2017

*J. Mater. Chem. A*, 2017, **5**, 7979-7991

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## desulfurization

Shuting Du, Qiming Sun, Ning Wang, Xiaoxin Chen, Mingjun Jia and Jihong Yu

Hierarchical TS-1 zeolites with abundant intracrystalline mesopores are synthesized and show excellent catalytic performance in oxidizing bulky organic compounds.



The article was first published on 12 Apr 2017

*J. Mater. Chem. A*, 2017, **5**, 7992-7998

<https://doi.org/10.1039/C6TA10044A>

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### Paper

## Effective calcium doping at the B-site of BaFeO<sub>3-δ</sub> perovskite: towards low-cost and high-performance oxygen permeation membranes

Yao Lu, Hailei Zhao, Kui Li, Xuefei Du, Yanhui Ma, Xiwang Chang, Ning Chen, Kun Zheng and Konrad Świerczek

A highly efficient and cost-effective strategy for doping Ca at the B-site of BaFeO<sub>3-δ</sub> was demonstrated to enhance oxygen permeability.



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### A green and robust solid catalyst facilitating the magnesium sulfite oxidation in the magnesia desulfurization process

Lidong Wang, Tiejue Qi, Siyu Wu, Shihan Zhang, Dan Qi and Huining Xiao

Effect of cobalt loading on the dispersity and catalysis performance.



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### The role of dissolution in the synthesis of high-activity organic nanocatalysts in a wet chemical reaction

L. J. Liu, Y. D. Lai, H. H. Li, L. T. Kang, J. J. Liu, Z. M. Cao and J. N. Yao

By combining nucleation/growth with dissolution of nanocrystals in a simple wet chemical reaction without capping agent under ambient conditions, a high-activity organic nanophotocatalyst (ZnTPP-H<sub>2</sub>O) was synthesized by simply adjusting the reaction time.

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### Microwave heating and the fast ADOR process for preparing zeolites

Marta Navarro, Samuel A. Morris, Álvaro Mayoral, Jiří Čejka and Russell E. Morris

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### Light-induced generation of free radicals by fullerene derivatives: an important degradation pathway in organic photovoltaics?

L. N. Inasaridze, A. I. Shames, I. V. Martynov, B. Li, A. V. Mumyatov, D. K. Susarova, E. A. Katz and P. A. Troshin

Photochemical degradation of fullerene derivatives producing persistent radical species represents one of the key failure mechanisms of organic solar cells.

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#### Paper

### Effective charge-discriminated group separation of metal ions under highly acidic conditions using nanodiamond-pillared graphene oxide membrane

Lei Wang, Xinghua Guo, Kecheng Cao, Bo Li, Yang Li, Meicheng Zhang, Rui Wen, Xing Li, Shoujian Li and Lijian Ma

Precise charge-discriminated group separation of coexisting cations in nitric acid solution was achieved by the as-prepared nanodiamond-pillared graphene oxide membrane.



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*J. Mater. Chem. A*, 2017, **5**, 8051-8061

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### Quick one-pot synthesis of amorphous carbon-coated cobalt-ferrite twin elliptical frustums for enhanced lithium storage capability

Yang Xiang, Hu Wu, Kelvin H. L. Zhang, Mike Coto, Teng Zhao, Sheng Chen, Bitao Dong, Shiyao Lu, Amr Abdelkader, Yuzhen Guo, Yanfeng Zhang, Shujiang Ding, Kai Xi and Guoxin Gao

A novel hierarchical hybrid nanostructure of carbon-coated  $\text{CoFe}_2\text{O}_4$  twin elliptical frustums ( $\text{CoFe}_2\text{O}_4@\text{C}$  TEFs) has been developed with excellent performance in lithium ion batteries.



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## Ultrahigh yield of hydrogen peroxide on graphite felt cathode modified with electrochemically exfoliated graphene

Weilu Yang, Minghua Zhou, Jingju Cai, Liang Liang, Gengbo Ren and Lili Jiang

The development of an efficient oxygen reduction reaction (ORR) cathode for hydrogen peroxide production represents an important challenge in the field of electrochemical processes and is highly demanded for chemical industries and environmental remediation applications.



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## Highly dispersed Co-based Fischer-Tropsch synthesis catalysts from metal-organic frameworks

Bin Qiu, Ce Yang, Wenhan Guo, Yao Xu, Zibin Liang, Ding Ma and Ruqiang Zou

Ultra-small Co nanoparticles embedded in hierarchically porous carbon were made *in situ* from metal-organic frameworks and used as catalysts in the Fischer-Tropsch synthesis.



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**A self-assembled 3D urchin-like  $\text{Ti}_{0.8}\text{Sn}_{0.2}\text{O}_2$ -rGO hybrid nanostructure as an anode material for high-rate and long cycle life Li-ion batteries**

Yutao Dong, Dan Li, Chengwei Gao, Yushan Liu and Jianmin Zhang

Self-assembled 3D urchin-like  $\text{Ti}_{0.8}\text{Sn}_{0.2}\text{O}_2$ -rGO was fabricated by a one-step hydrothermal process as an anode material for high-rate and long cycle life LIBs.



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**Reusable  $\text{Co}_x\text{Ni}_{1-x}$  dye adsorbents as supercapacitor electrode materials**

Haiming Sun, Xijia Yang, Lishu Zhang, Lijun Zhao and Jianshe Lian

A strategy to fabricate an electrode material by sintering a dye sludge consisting of a CoNi adsorbent with Congo Red under a  $\text{N}_2$  atmosphere.



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## Synergistic effect of ceria on the structure and hydrogen evolution activity of nickel nanoparticles grown on reduced graphene oxide

Mohammad Zhiani and Saeedeh Kamali

A facile chemical reduction procedure for the synthesis of nickel nanoparticles on a ceria-reduced graphene oxide composite (ceria-rGO) was reported and the rule of the ceria in HER was discussed.



The article was first published on 13 Apr 2017

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## Improving the photo-oxidative capability of BiOBr via crystal facet engineering

Xuelian Wu, Yun Hau Ng, Liang Wang, Yi Du, Shi Xue Dou, Rose Amal and Jason Scott

An increased presence of the {010} facet improved the performance of BiOBr for both water photooxidation and formic acid photodegradation.



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A chrysanthemum-inspired hierarchical  $\alpha$ - $\text{MoC}_{1-x}$  hybrid represents an excellent host material for  $\text{Li}^+$  ion storage.



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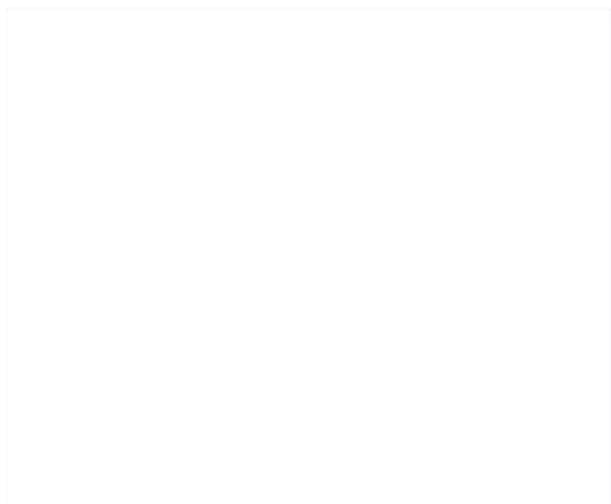
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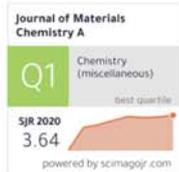
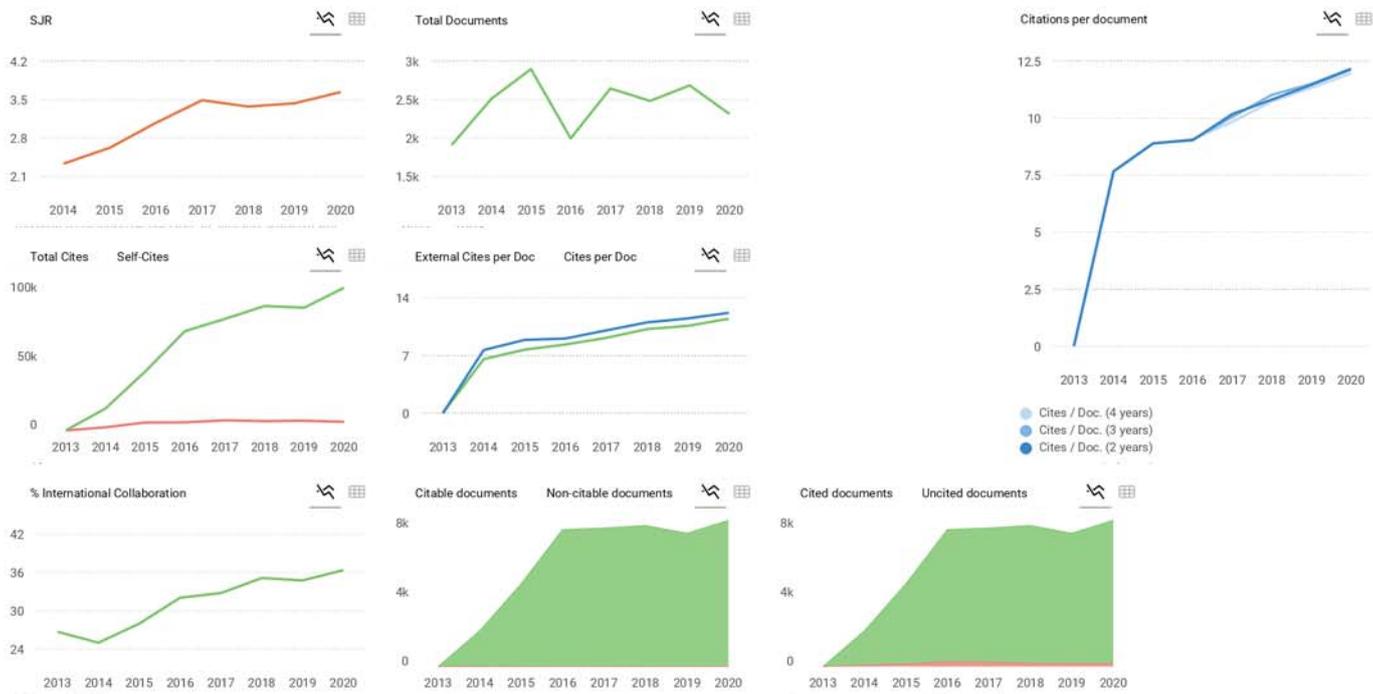

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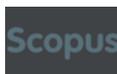
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