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Energy Spotlight

Advances in QD Ink, Li−S Batteries, and Gas-Diffusion Electrodes

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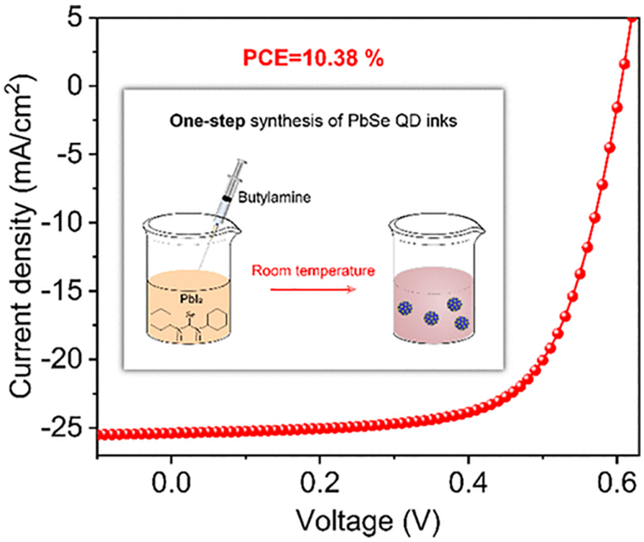
In this Energy Spotlight we present three highlights of recent articles published in *ACS Energy Letters*. The advances in utilizing semiconductor quantum dot inks for solar cell processing, electrochemical behavior of Li−S batteries under lean electrolyte conditions, and flooding possibilities in gas-diffusion electrodes are presented.

■ PbSe QUANTUM DOT SOLAR CELLS BASED ON DIRECTLY SYNTHESIZED SEMICONDUCTIVE INKS

Yang Liu, Fei Li, Guozheng Shi, Zeke Liu, Xiaofang Lin, Yao Shi, Yifan Chen, Xing Meng, You Lv, Wei Deng, Xiangqiang Pan, and Wanli Ma

ACS Energy Lett. **2020**, *5* (12), 3797−3803 (Letter)
DOI: 10.1021/acsenergylett.0c02011



For the widespread practical application of quantum dots (QDs) in electronics and optoelectronics, such as photodetectors, photovoltaics, and light-emitting diodes, QD inks that can be directly processed into functional QD films via industry-compatible manufacturing techniques such as inkjet printing are a “must-have”. Although it is very common to fabricate the QD film by spin coating and then performing solid-phase ligand exchange to gain decent film conductivity in the laboratory for exploratory R&D research, such QD film formation techniques are not suitable for large-area mass production and are thus not attractive for practical use. Ideally, in addition to showing satisfactory rheological properties, the QD inks should be based on well-passivated QDs free of defects that can act as charge carrier traps, have long-term stability over a wide range of QD concentrations, and involve only environmentally friendly solvents. To meet these requirements, a key factor is ligand engineering. As for the preparation of the QD inks, they have been typically obtained by first synthesizing high-quality QDs capped with long-chain organic ligands at elevated temperature and then conducting ligand exchange in solution with shorter ligands. The process involves multiple steps, which is time-consuming, leads to materials loss, and increases cost.

Liu *et al.* address these challenges by developing a one-step, direct synthesis procedure for PbSe QD inks at room temperature. The trick lies in the swift injection of butyl amine into the mixture of PbI_2 and selenourea derivatives, leading to the release of highly reactive HSe^- that reacts with PbI_2 to rapidly produce PbSe QDs well-passivated by lead halide complexes. As a result, the PbSe QD inks not only show excellent stability but also lead to >10% power conversion efficiency for the related solar cell devices. This simple, low-cost method is expected to facilitate the application of PbSe QDs. More importantly, the same strategy can be applied for synthesizing PbS, Ag_2S, and CdS QD inks, as previously reported by the same group (*Nature Communications*, **2019**, *10*, 5136). The method seems universal, which may be further explored for the synthesis of a variety of less toxic QD inks.
Dongling Ma

■ ROLE OF THE CARBON-BASED GAS DIFFUSION LAYER ON FLOODING IN A GAS DIFFUSION ELECTRODE CELL FOR ELECTROCHEMICAL CO_2 REDUCTION

Kailun Yang, Recep Kas, Wilson A. Smith, and Thomas Burdyny

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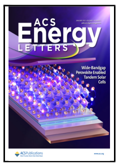

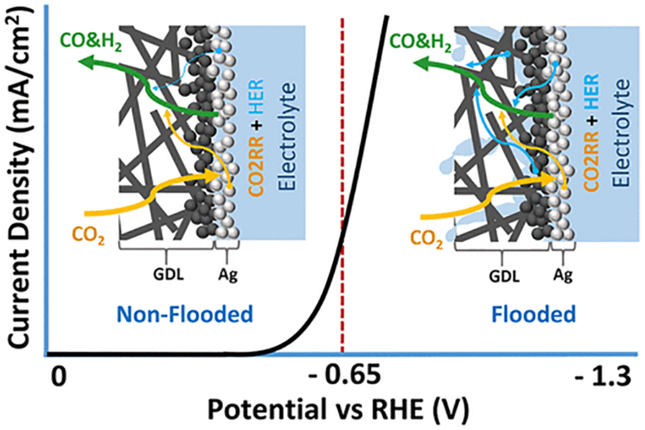

Continuous-flow electrolyzer cells employing gas diffusion electrodes (GDEs) transformed electrochemical CO_2 conversion R&D by enabling high current density operation. As a carryover from the fuel cell community, mostly carbon-based gas diffusion layers (GDLs) are used on the cathode side to distribute the CO_2 to the catalyst. They function reasonably well, but they have not been optimized for the electrochemical CO_2 reduction reaction (CO2RR). One specific drawback is flooding, when a fraction of the pores within the originally hydrophobic GDL become filled by a liquid. The penetration of water into the GDL blocks CO_2 from reaching the active site on the catalyst surface and also facilitates salt precipitation, which causes further failure by blocking the pore permanently. Despite some initial indications on the associated challenges, there has been no systematic study of this topic.

In their new study, Burdyny *et al.* demonstrate that the high overpotential required during electrochemical CO2RR initiates hydrogen evolution (HER) on the carbon GDL support. These potentials impact the wetting characteristics of the hydrophobic GDL, resulting in flooding that is independent of CO2RR. By performing a set of systematic experiments, they attempted to decouple the role of the catalyst and the GDL in both CO2RR and HER. Interestingly, the Ag/GDL and bare GDL had similar onset potentials and activity for HER (in the low-overpotential regime), indicating little difference in their electrochemical activity. It was hypothesized that water penetration may be due to electrowetting effects of the exposed carbon particles, rather than a change in the structure of the microporous layer. Electrowetting reduces the solid−liquid interfacial tension between the carbon and electrolyte and would result in a smaller contact angle as the applied potentials become more negative. It was concluded that by operating in a suitable potential range, CO_2 electrolyzers can reach a longer lifetime before flooding occurs. Such stability would greatly improve both the usability of GDLs for testing CO_2 electrolysis catalysts and operation, as well as enabling stability for industrial application. This Letter will inspire researchers to analyze flooding possibilities in their electrolyzer cells and to design GDEs and operational conditions which can mitigate this issue.

Csaba Janáky

■ EVOKING HIGH-DONOR-NUMBER-ASSISTED AND ORGANOSULFUR-MEDIATED CONVERSION IN LITHIUM−SULFUR BATTERIES

Abhay Gupta, Amruth Bhargav, and Arumugam Manthiram

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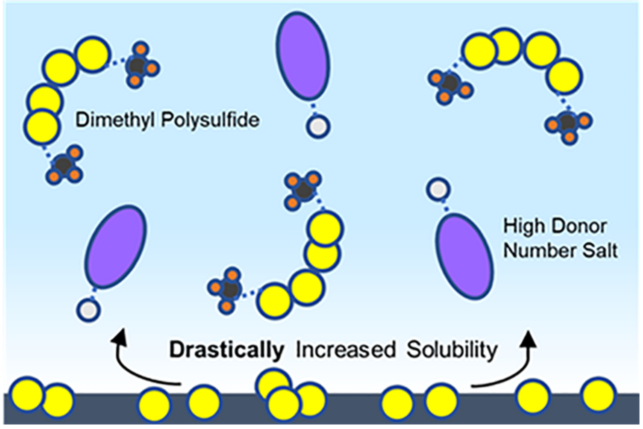

Lithium−sulfur (Li−S) batteries present an intriguing solution-mediated reaction paradigm that can bypass the sluggish solid-state intercalation kinetics in conventional lithium-ion batteries. The electrochemical response of these battery systems is predicated on an intricate combination of electrolyte transport interactions, speciation modes, and precipitate growth dynamics. While the Li−S chemistry offers exciting theoretical promise, the pathway toward achieving the desired practical performance remains challenging.

In this issue of *ACS Energy Letters*, Manthiram and coworkers present a novel approach to understand and enhance the electrochemical behavior of Li−S batteries under lean electrolyte conditions. The authors design an innovative strategy to combine the merits of high donor number salts and organosulfur-mediated discharge via the incorporation of methyl trifluoroacetate (CH_3TFA) to the Li−S electrolyte. The CH_3TFA additive results in *in situ* formation of lithium trifluoroacetate that fosters favorable solution coordination behavior and reduces polarization over cycling. In addition, an *in situ* derivatization to dimethyl polysulfides significantly improves the solubility of intermediate species, prevents clustering behavior, and alleviates cathode passivation to enhance the solution-mediated discharge behavior at lean electrolyte conditions. Built upon the intrinsic feature of active material dissolution in the Li−S chemistry, the proposed methodology synergistically activates advantageous coordination and solubility of polysulfides to unlock the electrochemical performance potential of such systems. This study sheds light on a new type of additives that can pave the way for methodical molecular engineering of intermediate species and mechanistically demystifies the solution-mediated reaction pathway toward unravelling the high specific energy promise of Li−S batteries.

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Notes

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