

Preface: Forum on New Materials and Approaches for Electrochemical Storage

This issue of ACS Applied Materials & Interfaces features a Forum on new materials and approaches for electrochemical energy storage. Li-ion batteries, supercapacitors, and technologies beyond Li-ion (e.g., Li–S, metal–air, and Mg-based batteries), currently represent an exciting area in applied materials research. This Forum is intended to provide the readership with articles that highlight the recent progress that has been made. These 12 papers demonstrate the broad interdisciplinary work that is needed to understand these complex devices and describe the challenges associated with finding new materials for the cathode, anode, and electrolytes in Li-ion and Li–S batteries.

The importance of materials for cathodes is addressed in five articles. Tarascon et al. (DOI: 10.1021/am405579h) provide an interesting discussion on the “secondary inductive effect” that lithium ions can have in determining the open circuit potential of polyanionic compounds containing an iron-based redox center. The article from Khalifah et al. (DOI: 10.1021/am500860a) presents an informative study on the crystal structure and electrochemical performance of thin film and bulk LiCoBO_3 . Spinel LiMn_2O_4 has received significant attention because of its high energy density and low cost. The contribution from Bartlett et al. (DOI: 10.1021/am500671e) describes the mechanism of capacity fading in LiMn_2O_4 related to oxygen nonstoichiometry and mechanical degradation of the electrode, showing an interesting observation of the structural collapse along the (111) planes after repeated cycles.

Conversion-based fluorides and oxyfluorides also have promise because of their high capacities and the high reduction potentials from strongly ionic metal-fluoride bonds. The article from Amatucci et al. (DOI: 10.1021/am500538b) describes an interesting use of the potentiostatic intermittent titration technique (PITT) to gain insight into the conversion reaction mechanisms of iron fluoride and oxyfluorides. PITT measurements allow one to isolate the true hysteresis on lithiation and delithiation from other kinetic effects such as nucleation overpotential.

Organic materials represent an interesting direction for cathodes in Li-ion batteries because their constituent elements are abundant, and their rich chemistry allows the tuning of structures and functional groups with different redox properties. This approach is illustrated by an article from Poizot et al. (DOI: 10.1021/am405470p) which describes the differences in voltage between the para- and ortho-position of the quinone/hydroquinone moiety in lithiated enolate-based materials.

Two contributions to the Forum describe the importance of anode materials. Alloys have large theoretical capacities (e.g., $\text{Li}_{4.4}\text{Si}$, 4200 mAh/g; and $\text{Li}_{4.4}\text{Sn}$, 994 mAh/g), but suffer from large volume changes during cycling. Both articles use intermetallics composed of an electrochemically active and inactive element. The article by Keles and Amine et al. (DOI: 10.1021/am405994b) reports the use of oblique angle deposition (OAD) to prepare nanocolumnar structured Cu–Sn thin films. OAD enables direct deposition of well-aligned

inclined nanocolumns on the current collector, eliminating the need for binder or conductive additives to get homogeneously distributed porosities, ensuring the direct transportation of the electrons at the same time as fast Li^+ diffusion. The article from Manthiram et al. (DOI: 10.1021/am500448f) describes the cycling performance of a $\text{FeSb}_2\text{–Al}_2\text{O}_3\text{–C}$ nanocomposite synthesized by the high-energy mechanical milling method. This work used both an inert metal (Fe) and a ceramic (Al_2O_3) to form the reinforcing matrix for supporting the active electrode material (Sb).

Electrolytes are crucial for advanced batteries, and must have high stability/safety, fast ion transport, and low cost. This direction is illustrated by two contributions. Yamada et al. (DOI: 10.1021/am5001163) describe the use of ethylene-carbonate-free superconcentrated electrolytes to address the solvent cointercalation issue in graphite negative electrodes. This strategy can expand the use of graphite electrodes with various solvents other than ethylene carbonate for high-voltage batteries. The article by Melot et al. (DOI: 10.1021/am4060194) presents interesting findings on lithium conducting garnets that are attracting significant interest as solid-state electrolytes. This study looks at the effect of the M^{2+} cation size on the structural and ionic transport properties in $\text{Li}_6\text{MLa}_2\text{Ta}_2\text{O}_{12}$.

Li-ion technology is limited by its inherent energy densities and cost. Therefore, batteries beyond Li-ion are being actively explored around the world. Examples include Li–S and metal–air batteries. Three contributions to the Forum highlight new development in lithium–sulfur batteries. An article contributed by Seshadri et al. (DOI: 10.1021/am405025n) describes a surface modified mesoporous carbon as a cathode material in Li–S batteries. The surface functionalization is believed to enhance the affinity between polysulfide and the carbon host. The article by Nazar et al. (DOI: 10.1021/am500632b) reports the synthesis of a graphene/carbon black/S composite for Li–S batteries. Scrosati et al. (DOI: 10.1021/am4057166) report a lithium-ion sulfur battery using a lithium sulfide cathode and silicon-based anode. The results demonstrate the applicability of the system as an energy storage system characterized by the employment of alternative materials, with expected high safety level and low-cost.

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Notes

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