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Editorial Overview: Batteries and Supercapacitors (2023)
Energy storage, the unrepentant attention-seeker

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Electrochemical energy storage (batteries and supercapacitors) can be likened to the proverbial spoilt child who keeps seeking attention: every breakthrough comes with more challenges. Having rewarded the big uncle in the family (Li-ion battery) with a Nobel prize for excellent behaviour in driving green transportation and portable electronics, one would have thought that the scientific community would have understood most of the underlying chemistries but, alas, we are wrong!

In this theme collection, we have 22 current opinions on various energy storage systems: lithium-ion batteries, metal-sulfur batteries, aqueous metal-ion batteries (potassium-ion batteries, magnesium-ion batteries, zinc-ion batteries), rechargeable metal (Zn & Al)-air batteries, redox-flow batteries, and supercapacitors. The theme touched on various aspects of these energy storage systems, from anode to cathode and electrolytes, and the need for top-level characterisation (synchrotron-based techniques).

The focus on lithium-ion batteries (LIBs) has turned toward obtaining high-energy cathode materials that would allow the development and commercialisation of electric vehicles that can travel longer distances (>600 km) than the current average of about 350 km on a single charge. To realise this ambitious dream, next-generation high-energy nickel-rich nickel manganese cobalt (NR-NMC) layered oxides have been proposed by the research community. However, the application of the NR-NMC cathode materials has been haunted by surface instability, structural failure and thermal instability leading to capacity fading. As described by Kebede (Article 101261), some of the proposed solutions include doping and coating of the NR-NMC with appropriate compounds. It should be highlighted here that the trend in high-energy cathode materials for LIBs is shifting toward the cobalt-free, manganese- or nickel-rich layered oxides [1,2]. One anticipates growth in this area as efforts are geared toward cost reduction and widespread utilisation of electric vehicles.

Despite the importance of an anode material for achieving high areal capacity for LIB, it usually receives less attention than its cathode counterpart. In this theme, Balogun and co-workers (Article 101260) looked at the prospects of the self-supportive carbon—and transition metal-based materials for the development of high-areal-capacity-based self-supportive electrodes for LIBs. Ezema and co-workers (Article 101250) reviewed the progress being today on the use of green synthesis for transition metal-based oxides as anode for LIBs.

As part of the ongoing efforts at developing the next-generation low-cost and safe batteries (especially the sodium-ion batteries, SIBs), Palaniyandy and co-workers (Article 101217) reviewed the mechanisms of mixed-ion cathode materials for aqueous and non-aqueous lithium/sodium-ion batteries, while Chen and co-workers (Article 101200) reviewed the underlying principles of carbon-modification for improving the electrochemical performance of the popular NASICON family, the $Na_3V_2(PO_4)_3$ cathode materials for SIBs. Aside from SIBs, two important metal-ion batteries in this theme are the K-ion batteries using the P3-type layered oxide cathode framework by Barpanda and co-workers (Article 101216, also see our cover page), and Mg-ion batteries using MnO_2 polymorphs by Shimokawa and co-workers (Article 101209). The authors proposed that the defective spinel-type $ZnMnO_3$ is a promising cathode candidate for developing rechargeable magnesium-ion batteries, even at elevated temperatures (150 °C).

In the race for developing the low-cost and high energy-dense 'beyond-lithium-batteries', the metal-sulfur batteries seem to receive the loudest crowd-cheering. In this theme, =Choudhury and co-workers (Article 101217) discussed the need to focus on inverse vulcanisation as a facile method to obtain sulfur-based polymers as alternatives to conventional sulfur in lithium—sulfur batteries (LSB). The authors compared normal inverse vulcanization with the more recent catalytic and photo-induced inverse vulcanisation processes. An emerging family member is the aluminium—sulfur battery (ASB) which is likely to leapfrog from the knowledge of LSB to make impact. For the first time, Deka & Dutta (Article 101222) reviewed this new technology, focusing on the recent advances in the use of carbon materials as sulfur-hosts for ASB.

Supercapacitors continue to make impact despite the intense survivability competition with some new battery technologies in terms of power density. Although supercapacitors are inherently the device of choice for power density, while batteries are for energy density, current research efforts seem to be directed for both to be applied in devices that requires possess high power and high energy densities. In this theme, Abbas and co-workers (Article 101249) reviewed the latest research progress with a focus on the critical role that nanostructured carbon pores play in supercapacitors, especially the trade-off between energy and power. Ezema and co-workers (Article 101239) reviewed the progress being made in the use of redox-active materials, V_2O_5-based electroactive materials, in enhancing the energy density of supercapacitors. Ola and co-workers (Article 101243) reviewed a usually overlooked aspect of solar-supercapacitor, called photo-supercapacitor (PSC). In the authors' opinion, there is an urgent to standardise the characterisation parameters and definitions, with the inclusion of such methods as the cyclic voltammetry and galvanostatic charge—discharge analysis to avoid confusion.

Redox-flow batteries (RFBs), especially vanadium-based flow batteries, have long been recognised as the most plausible batteries for stationary applications. Mitigating cross-contamination via bipolar redox-active materials and bipolar membranes. However, the long-term cycling performance of the RBFs, arising from the cross-contamination of the catholyte/anolyte, has continued to hamper their widescale commercialisation. In his opinion, Chen (Article 101188) believes that this technological challenge of cross-contamination 'can ultimately' be mitigated by using bipolar redox-active materials that allow the use of the same electrolyte in the two half-cells.

One of the huge concerns about batteries is safety. Most (if not all) research activities in this field have been focused on the electrolytes. In this theme, three groups shared their opinions on electrolytes: Martins (Article 101241) looked on the latest advances at improving liquid electrolytes for Li-ion and Li metal batteries, Kim and co-workers (Article 101251) focused on sulfide-based all solid-state batteries (ASSBs), while Pang and co-workers (Article 101285) reviewed the need to develop solvating electrolytes for the next generation lithium-sulfur batteries.

Zinc-based batteries are receiving increasing attention because of their high energy and power densities, environmentally benign, and safe to use compared to the lithium-ion batteries. Fabio and co-workers (Article 101230) discussed the challenges of efficient utilisation of the zinc anode, also known as the depth of discharge (DoD), which continue to hamper the commercialisation of rechargeable aqueous zinc-ion batteries (ZIBs). For practical application of ZIBs, the authors challenged the community to focus discharging at high DoD values ($\geq$50%), high current densities (>2 $mAcm^{-2}$) and capacity limits (>2 $mAhcm^{-2}$) and making the economic and environmental sustainability of the chosen strategies a priority.

Metal-air batteries (MABs) are next generation battery technologies driven by bifunctional oxygen electrocatalysis, i.e., oxygen reduction reaction (ORR) and oxygen evolution reaction (OER). MABs are known for their high specific energy and power densities, low-cost and environmentally benign. The performance of the bifunctional electrocatalysts is entirely dependent on the rational design and defect-engineering of both the metal catalysts and their carbon supports. Ipadeola et al. (Article 101198) reviewed the recent trends in the adoption of porous transition metal-based nanostructured electrocatalysts as bifunctional electrocatalysts for rechargeable aluminium-air batteries (RAABs). Chen and co-workers (Article 101197) revisited the rational design and defect-engineering of carbon-supported single and dual atom catalysts for bifunctional oxygen electrocatalysis in alkaline media. From their work, we are reminded about the need to strike a balance on the extent to which defects are introduced for optimum performance. This is critical considering that although defects can promote the desorption of OH* for ORR and facilitated deprotonation of OH* for OER, excessive defects ultimately lead to reduced graphitisation which then compromises the electrical conductivity. In the authors' opinion, the best design strategy is always to develop 'advanced carbon substrates with both high graphitisation and abundant defects to support the metal sites'. Zhang and co-workers (Article 101206) agreed with Chen and co-workers (Article 101197) but extended the discussion to rechargeable zinc-air batteries (RZABs) and showing that 'carbon skeletons with additional heteroatom, edge defects, or curved nanostructure may additionally regulate the electronic structure of active sites' for improved ORR and OER performance in RZABs.

As expected, RZABs dominated the MABs in this theme; Zhang and co-workers (Article 101206) on single atom catalysts; Tammeveski and co-workers (Article 101229) on transition metal-based catalysts, and Haruna et al. (Article 101264) on high-entropy material-based catalysts. This should be expected as RZAB has been described as a viable future battery technology that can complement LIBs. In this theme, however, it was the bifunctional electrocatalysts that dominated the opinion rather than the zinc anode that playing critical limiting roles to the performance of the RZAB cell. This is perhaps should not be surprisingly because many researchers do not seem to fully appreciate the importance of zinc utilisation (i.e., DoD) for real practical application [3,4].

Rolison and co-workers [3] have advised the community on the need for researchers to consider '*translating materials-Level performance into device-relevant metrics for zinc-based batteries*'. According to the authors [3,4], for RZABs to compete with LIBs, they must be discharged at a DoD of 20% [3], or 11.7 $mAhcm^{-2}$, or areal specific energy density of 35 $mWhcm^{-2}{}_{geo}$ at technologically relevant current density of 10 $mAcm^{-2}$ [4]. Disappointingly, of the 100 research articles investigated by the authors, only 8 of them met this important threshold. In my opinion, this is an excellent finding that underscores the need for future work in RZABs, and one hopes to see improvements in the years ahead.

To observe and understand the underlying mechanisms of the performance and challenges of energy storage systems require top-level characterisation, synchrotron-based techniques. In this theme, Barrett and Rodella (Article 101242) reviewed the latest developments in *operando* and *in-situ* characterisation of energy storage materials using synchrotron radiation. The importance of synchrotron facilities, in terms of real-time monitoring of the electrochemical events in energy storage, cannot be over-emphasised. Although still limited in access, there has been an increased utilisation of synchrotron methods in energy storage. Without a doubt, any future breakthrough in energy storage will come from *operando* and *in-situ* characterisation studies.

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