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Preface to Special Issue: Flexible energy storage

While the better part of the 20th century was dedicated to increasing the energy density of energy storage devices (ESDs) including both batteries and supercapacitors, the 21st century has witnessed a quick rise in integration of these devices into electronic appliances. Lithium ion batteries made complex mobile electronic devices a reality, but at present electronic appliances are guiding the design of ESDs. The nature of electronic devices has evolved into flexible/foldable displays, wearable artificial skins, robotics, sensory devices, etc. Moreover, astounding innovations are seamlessly integrating energy harvesting systems with our daily activities. But ESDs seem to have lagged behind in terms of mechanical flexibility, with barely any initiative for commercialization. At this moment, commercial flexible electronics lack compatible flexible energy storing units, resulting in undesirably thick/heavy foldable phones or other devices. This is a great reason why we need high-energy flexible ESDs and has promoted a recent surge of interest towards solving this problem. Any ESD that can sustain a substantial amount of mechanical deformation can be termed as a flexible ESD. These deformations could come in any form like bending, folding, crumpling, stretching, rolling, and compressing. The ESDs should maintain stable operation even under these harsh circumstances to be truly termed as flexible. Since ESDs are composed of several components, it is just not enough to make their electrodes flexible. The electrolytes and current collectors must also be compatible with the flexible electrodes; otherwise we run the risk of electrolyte leakage or delamination from current collectors which will lead to subpar performance. The aim should be to preserve all the goodness of traditional ESDs while making them flexible. Therefore, it is high time that special attention be given to this emerging field of flexible ESDs. This will undoubtedly bridge the gap that exists between integrated energy harvesting and consuming systems and accelerate our journey towards achieving the Internet of Things in a true sense.

Thankfully, this research direction has been advancing very fast and, to keep up with it, we present this special issue on “Flexible Energy Storage”. This issue is to draw attention towards how the pursuit of achieving flexibility in ESDs is gradually being adopted by researchers around the globe, to put forward the unconventional approaches that have been employed, to discuss the roadblocks that must be cleared, and to suggest unique perspectives that help guide future research. This issue features some of the best research innovations in this emerging field encompassing a wide variety of flexible ESDs. The starting point naturally is the incorporation of new inherently flexible materials like graphene and MXene. There has been extensive research on the synthesis of these materials and examples of work demonstrating their flexible applications are presented. Therefore, it is useful to have a comprehensive analysis of the fabrication methods that particularly focus on flexibility. These materials could also be incorporated into highly flexible systems like textiles, for instance, by self-assembly of MXene on polymer-based fibres to achieve highly conducting fabrics.

Once we have a good start by synthesis of electrodes using these new materials, the next and most crucial step to consider is their safe assembly to construct flexible ESDs, including micro-supercapacitors, hybrid ion capacitors, and new-generation batteries including printed batteries, aqueous batteries, Zn-MnO_2 batteries, sodium ion batteries, lithium metal batteries, and metal air batteries. Breakthroughs in these fields will provide us with more options to build next-generation ESDs that have tailored energy/power density, voltage output, safety, and integration ability for satisfying future demands like in wearable, all-weather electronics. Having flexibility is just the beginning, achieving ESDs free of design restrictions will allow the next level of device integration, such as solar-powered rechargeable batteries. This is the vision that we are truly looking forward to. Therefore, we have invited world-leading scientists in this field to share their results and their outlook for the future. This special issue includes all these exciting works and more.

This special issue packs 23 contributions, including 16 reviews and 7 research articles from globally reputed scientists. As mentioned earlier, electrolytes, current collectors and the overall integration of these components with energy-dense electrodes are focused to show a glimpse of what the future holds. We emphasize that we have compiled this issue considering the gradual shift towards alternative energy storage systems aiming to reduce pressure on Earth’s natural resources. For example, lithium ion batteries that have dominated the global electronics market for a substantial time are beginning to put pressure on the limited lithium resources, pushing researchers to look for alternatives that use earth-abundant resources like sodium ion batteries. Not only that, achieving greater safety is also a crucial factor and therefore developing aqueous, ionogel and solid electrolytes is important for ESDs. All these great considerations must be consolidated with robust and mechanically flexible architecture that enhance their durability. To summarize, special attention has been given to the following aspects: (i) flexible batteries and hybrid capacitors focussed on enhancing both active materials and electrolytes, (ii) applications of new-generation flexible materials and emerging composites for textile-based ESDs, (iii) integration of ESDs with consumer electronics and energy harvesting systems like solar cells on flexible substrates by means of high throughput methods like printing, (iv) innovative approaches for achieving flexibility in next generation batteries (*e.g.*, Na^+, Zn^{2+}), and (v) blurring the trade-off between flexibility and performance.

We are confident that this unique compilation spanning multiple disciplines will inspire a generation of researchers to think differently and

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ahead of the present times, keeping a steady gaze on the bigger picture of bringing together all electronics into a single flexible unit. We heartily invite everyone to take part in this initiative by exploring this special issue, reading these articles and spreading these ideas. Finally, we extend our sincere gratitude towards all the authors, reviewers, Editor-in-Chief, and editorial team for their great contributions at various stages from inception to fruition.

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