

ENERGY STORAGE FOR A GREEN FUTURE: Novel Dibenzamide Porous Organic Polymer Shows Exceptional Performance in Lithium-ion Batteries

Polymers are widely used in the electrodes of lithium-ion batteries (LIBs) as they can hold together its components and improve the battery's mechanical stability and flexibility. In particular, porous organic polymers (POPs) have garnered significant attention over the last decade as cathode materials in LIBs due to their structural diversity and the presence of different surface chemical groups (i.e., functional groups) that can be tuned to improve the cathode characteristics. However, most POP-based electrodes face limitations in either lithium storage capacity or stability at higher charging and discharging (cycling) rates.

To overcome these challenges, a team comprising two researchers from the Research Institute for Sustainable Energy (RISE), TCG Centres for Research and Education in Science and Technology (TCG-CREST)—along with researchers from the Indian Institute of Technology (IIT) Indore, the Indian Institute of Science Education and Research (IISER) Kolkata, and JEOL Ltd., Japan—has designed a novel bifurcated dibenzamide (DBA) linkage for POPs. This innovative linkage significantly enhances the lithium storage capacity of POP-based cathodes while maintaining superior cycling stability. Their findings have been published in the [Journal of the American Chemical Society](#).

According to Dr. Abhik Banerjee of RISE, *"The novelty of our approach is in the formation of a bifurcated linkage from the condensation reaction between aryl amide, such as terephthalamide (Tam), and aryl aldehyde, such as terephthalaldehyde (TA). This linkage, termed DBA, has an affinity for lithium, i.e., it has lithiophilic functionalities. By virtue of this DBA linkage, we were able to design a simple, high capacity, long-life polybenzamide-based cathode with an excellent cycling rate performance."*

To begin with, the researchers synthesised four different DBAs using different aryl aldehydes. They then employed proton nuclear magnetic resonance spectroscopy (^1H NMR) to confirm lithiation—an indicator of the lithiophilicity of the DBA linkages. *"During lithiation, hydrogen atoms in an organic compound (such as DBA) are replaced with lithium atoms. The opposite process is termed delithiation,"* explains Himanshi Singh, who is currently pursuing an apprenticeship at RISE and was part of the research team. *"We saw that all four variants of DBA were lithiophilic. As a next step, we extended this synthesis to design POPs with DBA linkages."*

For the synthesis of these POPs, the research team used Tam and various ditopic aryl aldehydes, including TA. Ditopic here means that there are two sites on the compound where another molecule can bond. A condensation reaction between the two led to the formation of different porous polybenzamide polymers. The researchers then applied these different polybenzamide POPs as cathode materials for LIBs, and analysed their capacity and cycling stability. *"We found that the DBA linkages increase the diffusion and access of Li-ions to reaction sites, thereby facilitating reversible*

electrochemical reactions. This allows polybenzamide POPs to achieve high specific capacities as LIB cathode materials,” notes Dr. Banerjee.

Among the different polybenzamide POPs synthesised by the researchers, the polybenzamide formed by the condensation of TAm and TA stood out for its exceptional qualities. The TAm–TA cathode was able to maintain 63% of its initial capacity at the demanding rate of 10C compared to its capacity at 1C. The battery was also able to maintain its performance over 3000 cycles at 1.3C and over 6000 cycles at high rates of 5C to 40C with a Coulombic efficiency—the ratio of the discharge capacity to charge capacity—of 99.99%, indicating its durability and longevity. *“We also tested the TAm–TA POP in full-cell assemblies as both cathode and anode and found that it could deliver excellent specific capacity,”* adds Ms. Singh. *“These experiments together suggest the high application potential for TAm–TA POP in real-life LIBs.”*

Additionally, the research team analysed the lithiation and delithiation mechanisms of the POPs using *ex situ* Fourier transform infrared spectroscopy (FT-IR) and x-ray photoemission spectroscopy. Their findings indicated that during the discharging process, each DBA group gains three pairs of lithium ions and electrons into their carbonyl (C=O) and amide (N-H) functionalities. These three pairs are reversibly released during charging to regenerate the original polybenzamide framework. The researchers further performed density functional theory (DFT) simulations that supported this finding. As Dr. Banerjee mentions, *“Understanding the lithiation and delithiation mechanism of DBA will aid in efforts to further tune the design of these polymer frameworks and improve their performance in LIBS.”*

Today, LIBs are considered a central pillar to future efforts towards sustainability. As the world shifts away from fossil fuel-based energy towards green energy sources, there will be an increasing need for high-capacity, long-life batteries to support this transition and ensure the continuous supply of energy. The innovative approach outlined in this research not only solves long-standing challenges associated with POP-based electrodes, but also opens new avenues for research and development in energy storage. *“The enhanced performance of the novel DBA-based POPs designed in our study will usher in a new era of high-performance LIBs,”* concludes Dr. Banerjee.

Congratulations to the team for their stellar contributions to the field of energy storage!