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Gwangju Institute of Science and Technology Researchers Improve the Solubility of Redox Molecules for Enhanced Energy Storage Systems

*Researchers introduce hydrotropic-supporting electrolyte to enhance the aqueous solubility of organic redox molecules for electrochemical capacitors* 

Redox-enhanced electrochemical capacitors (ECs) are a sustainable alternative to the dominant battery technologies that use metal ions and corrosive chemicals. However, obtaining organic redox-active electrolytes with sufficient solubility for use in redox-enhanced ECs is challenging. Now, researchers from Korea have introduced a hydrotropic-supporting electrolyte with tailor-made organic redox-active species for ECs, which results in increased solubility of the redox-active species without sacrificing electrochemical kinetics.



Image title: Enhancing the solubility of organic redox-active molecules.

Image caption: Hydrotropes customized with sparingly soluble organic redox-active species can enhance its solubility and the redox activity of electrochemical capacitors, without compromising the electrochemical kinetics of the redox molecules, finds a new study by researchers from Korea.

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Dominant battery technologies using flammable, toxic, unsustainable, and expensive energy sources are a major contributor to climate change. Switching from fossil fuels to cleaner and environmentally friendly energy sources is thus crucial to curtail the impacts of climate change. This transition can be supported by improving the efficiency of energy storage systems for safer and stable operations, sustainability, high energy/ power density.

Research on this front has focused on molecular engineering approaches to the development of aqueous-based redox-enhanced electrochemical capacitors (redox ECs). Redox ECs are a type of advanced hybrid electric double-layer capacitors that use redox-active molecules at the electrode-electrolyte interface to increase the energy density. Owing to the use of organic redox-active electrolytes, they are known to provide a cost merit, use of earth-abundant elements, and structural tunability. However, a major challenge in their development is the lack of sufficient solubility of these species in aqueous systems, which results in a low energy density. Furthermore, previous attempts at improving their solubility have proven to be time-consuming and cost extensive.

Now, researchers from Korea have used hydrotropic-supporting electrolyte (HSE) as an approach to enhancing the solubility of the organic redox-active species. The study, led by Assistant Professor Seung Joon Yoo and Professor Sukwon Hong from Gwangju Institute of Science and Technology in Korea, was made available online on 20 April 2023 and was published in Volume 8, Issue 5 of the journal <u>ACS Energy Letters</u> on 12 May 2023.

The researchers used the process of hydrotropy, wherein a class of amphiphilic molecules are used. In this unique solubilization phenomenon, the volume of the hydrophobic component is relatively small compared to that of the surfactant, thus allowing an increase in the solubility of the sparingly soluble solute multifold. The researchers tested a range of quinones as a model species owing to their utility as a redox-active additive and an acceptable electrochemical stability.

The researchers found that using the HSE (p-toluene sulfonic acid (p-TsOH), 2naphthalenesulfonic acid (2-NpOH), and anthraquinone-2-sulfonic acid (AQS)) improved the solubility of hydroquinone (HQ) without any chemical functionalization. Importantly, they demonstrated that an increase in the solubility is proportional to the concentration of the respective HSEs.

Moreover, they designed a biredox salt, 2-[N,N,N-tris(2-hydroxyethyl)] anthracenemethanaminum-9,10-dione bromide (AQM-Br), which could participate in Faradaic reactions at both the positive and negative electrodes, and tested it in the HSE system in a concentration-dependent manner. Prof. Yoo highlights, *"The solubility of HQ in HSE was increased 7-fold, and a designer multifunctional dual-redox species (AQM-Br) was synthesized, the solubility of which was significantly enhanced from being barely soluble to* >1 *M by optimizing the HSE."* 

Furthermore, the researchers also attempted to understand the action of solubilization for both the HQ and AQM-Br electrolyte. Using the intermolecular nuclear Overhauser effect and dynamic light scattering analyses, they found that the hydrotrope solubilization for HQ/HSE was achieved through the co-solubilizer mechanism, whereas for AQM-Br/HSE, it was due to the formation of quasi-micelle nanostructures.

Explaining the potential implications of the study, Prof. Yoo concludes, "Our simple approach can be readily extended to a different class of redox species and find applicable to a wide variety of applications including redox flow batteries." In addition, our study provides

a guideline for the design of energy-dense redox-active electrolytes and an optimal selection of HSE and redox-active electrolyte pairs.

Are redox ECs the future? It certainly looks like they're here to stay!

Reference	
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## About the Gwangju Institute of Science and Technology (GIST)

The Gwangju Institute of Science and Technology (GIST) is a research-oriented university situated in Gwangju, South Korea. Founded in 1993, GIST has become one of the most prestigious schools in South Korea. The university aims to create a strong research environment to spur advancements in science and technology and to promote collaboration between international and domestic research programs. With its motto of "A Proud Creator of Future Science and Technology," GIST has consistently received one of the highest university rankings in Korea.

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# About Assistant Professor Seung Joon Yoo from Gwangju Institute of Science and Technology

Dr. Seung Joon Yoo is an Assistant Professor of Materials Science & Engineering at Gwangju Institute of Science & Technology (GIST), Korea. He obtained his Ph.D. from the University of California, Santa Barbara (UCSB) in 2014. From 2015 to 2019, he was a Postdoctoral Research Associate in the laboratory of Prof. Galen D. Stucky, University of California, Santa Barbara (UCSB). He is a recipient of the prestigious NSF Partnership for International Research and Education: Electron Chemistry and Catalysis Interfaces (PIRE-ECCI) Postdoctoral Fellowship and Graduate Fellowship. His current research interests include organic chemistry, electrochemistry, and electrochemical energy storage. His lab is the organic electrochemistry and energy materials laboratory at GIST.

## About Professor Sukwon Hong from Gwangju Institute of Science and Technology

Sukwon Hong is a Professor of Chemistry at Gwangju Institute of Science and Technology (GIST). His group is developing organometallic catalyst for asymmetric reactions, ethenolysis, CO<sub>2</sub> chemistry, and photochemistry. The Hong group at GIST is also developing functional molecules for energy conversion, such as solar cells and thermoelectricity. Before coming to GIST, Dr. Hong worked as assistant professor at the University of Florida. In 2003, He received his PhD in Chemistry from Northwestern University, USA.