Researchers develop organic, air-stable, highly conducting neutral molecular crystal with unique electronic properties

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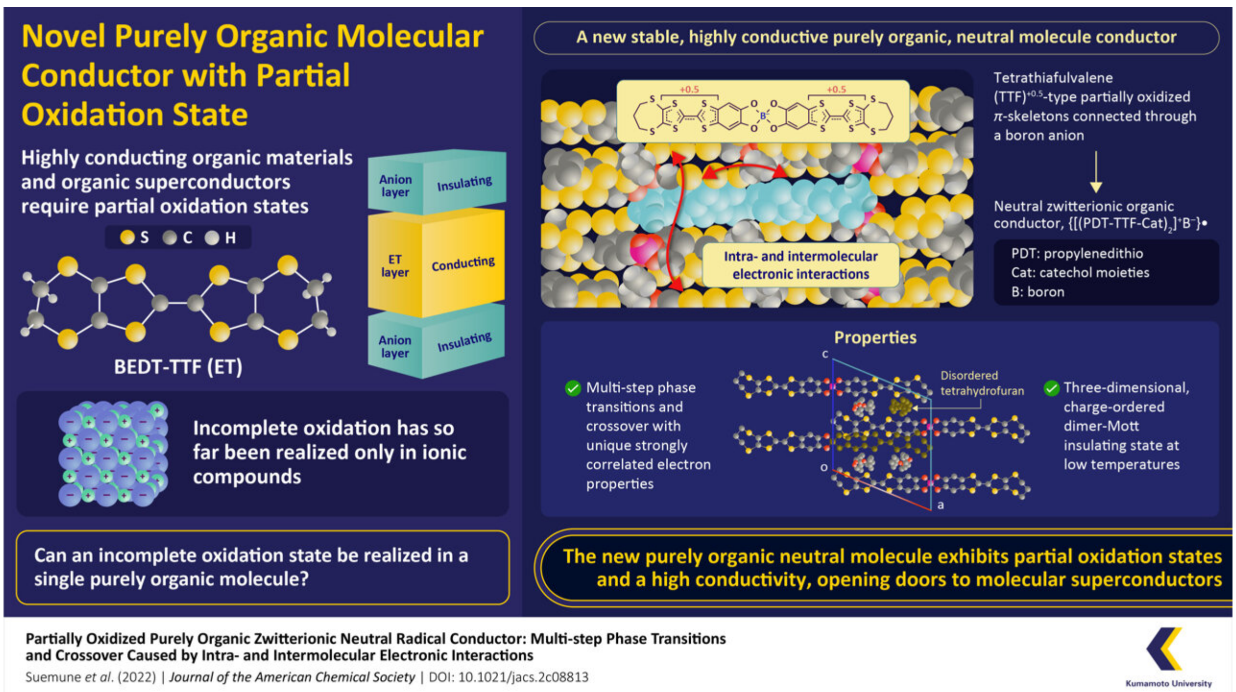

An unprecedented partial oxidation state realized in a single purely organic neutral molecule by a team of researchers in Japan could provide a platform for the development of novel organic superconductors and strongly correlated electron systems, with potential applications in future superconducting technology. Credit: Kumamoto University

In an unprecedented feat, researchers from Japan have developed an organic, air-stable, highly conducting neutral molecular crystal with unique electronic properties.

Typical organic substances such as paper, sugar, and naphthalene are composed of electrically neutral molecules that are poor conductors of electricity. Since the advent of superconductivity, there has been a renewed interest in highly conducting organic materials.

It has been demonstrated that the key to excellent electrical conductivity is a combination of an electron-donating molecule and an electron-accepting molecule, known as a charge transfer complex that is characterized by an incomplete oxidation state. Such an incomplete or partial oxidation state has been realized only in ionic compounds so far.

However, superconducting thin films with useful features like excellent solubility and workability require single organic small molecules with a partial oxidation state. Equivalently, the development of small organic molecules with both positively and negatively charged groups, commonly known as zwitterions, could potentially revolutionize superconducting technology.

To this end, a team of researchers from Japan, led by Dr. Akira Ueda from Kumamoto University, has now developed a single purely organic neutral molecule with an incomplete oxidation state for the first time. In a recent article made available online on November 22, 2022 and published in the *Journal of the American Chemical Society* on December 7, 2022, the researchers presented the details of the development of this purely organic air-stable neutral "zwitterionic" radical conductor.

"It has been long believed that combining two (or more) types of ionic molecules is essential for achieving partial oxidation state in organic materials. Our research group has now overturned this common belief with our original molecular design. For the first time, we have succeeded in creating an 'incomplete oxidation state' with a purely organic neutral molecule," explains Dr. Ueda, the corresponding author of the article.

The researchers connected two tetrathiafulvalene $(TTF)^{+0.5}$-type partially oxidized π-skeletons through a negatively charged boron (B) ion to realize an unprecedented partial oxidation state in a single, purely organic, neutral zwitterionic molecule. The new molecule, namely a propylenedithio (PDT)-substituted derivative, $\{[(PDT\text{-}TTF\text{-}Cat)_2]^+B^-\}\bullet$, uses a boron anion B^- as the linker to two partially oxidized $TTF^{+0.5}$ skeletons through catechol (Cat) parts. This design facilitates intra- and intermolecular electronic interactions, the key to achieving incomplete oxidation states.

The team confirmed the formation of partially oxidized state by measuring the electrical resistivity and magnetic susceptibility of the molecules in the solid state, and using X-ray diffraction for its characterization. They further demonstrated that the partially oxidized state led to multi-step phase transitions and crossover, which provided unique strongly correlated electron properties to the neutral molecular solid. This manifested as a three-dimensional charge-ordered dimer-Mott insulating state at low temperatures.

Overall, the results indicate that the newly developed neutral molecular crystal could serve as a promising platform for studying molecular superconductors and strongly correlated molecular electronic systems. "We expect that the new development will lead to novel functional organic substances and materials," concludes Dr. Ueda.

More information: Taro Suemune et al, Partially Oxidized Purely Organic Zwitterionic Neutral Radical Conductor: Multi-step Phase Transitions and Crossover Caused by Intra- and Intermolecular Electronic Interactions, *Journal of the American Chemical Society* (2022). DOI: 10.1021/jacs.2c08813

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Provided by Kumamoto University

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