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Computational Study on Advanced Materials for Organic Radical Battery Cathodes

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Project Summary
The objectives of this research are to understand organic radical materials for rechargeable battery and design novel materials based on the understandings. First-principles quantum calculations will be used to perform the proposed research. The achievement of the proposed research will advance scientific knowledge for organic radical battery materials and structural batteries.

Li ion batteries have attracted a great attention as leading technologies for the next generation energy storage in various applications from consumer electronics to electric vehicles. Currently, common format of Li ion batteries is based on inorganic materials, in particular transition metal oxides for cathode and graphite for anode. However, these materials are suffering from the slow charging/discharging. Many efforts have been made to rectify these issues, however they also accompany the compromises, such as sacrifice of the specific energy, manufacturing cost increase, unavailable mass production, etc. More transformative technology is demanded for developing rechargeable batteries that are free from those problems.

Organic radical battery (ORB) can be a promising candidate for that purpose. It uses organic radical compounds, mostly nitroxide-based polymers. The use of organic polymers brings several advantages to ORB compared to conventional inorganic Li ion batteries. They are more environmentally friendly than metal-based batteries and enables design of flexible batteries that can lead to structural batteries. As this type of battery is relatively new (first suggested in year 2005), many mechanisms still remain to be clarified and more research efforts are demanded to better understand ORB materials.

Nitroxide-based polymers are known to function as electrodes of ORB. The voltage and specific capacity (two most important quantities of ORB) are determined by the chemical bonds in these materials. Among various nitroxide-based polymers, poly(2,2,6,6-tetramethyl-1-piperidinyloxy-4-yl methacrylate) (PTMA) has been a well-known cathode material for ORB. PTMA is a polymethacrylate (PMMA) with (2,2,6,6-tetramethylpiperdinioxyloxy) radical (TEMPO) attached, as illustrated in Figure 1(a). The nitroxide radical in TEMPO radical, as shown in Figure 1(b), is the most common subunit used in ORBs. It can be oxidized to an oxoammonium cation or reduced to an aminoxyl anion, as illustrated in Figure 1(c). As the charge transfer in this redox is mediated by electrons

![Figure 1](image-url)
that are much faster than ions in conventional metal-based cathodes, enabling fast charge/discharge of ORBs. The change of thermodynamic energy during the redox will generate the open cell voltage (OCV) of ORB. Interestingly, the OCV of ORB is reported to be as high as the one from metal-based cathodes, while it has been known the electron transfer usually occurs at low voltage.

In the proposed research, PTMA will be investigated for their electrochemical and structural properties and the mechanism of polymer construction. The radical dependence of polymer cathodes will be also examined by high-throughput calculations on other radicals than TEMPO, especially high voltage radicals: e.g. anisole-family compounds, 4,4’dimethoxybiphenyl, poly(2.5-dimercapto-1,3,4-thiadiazole), di-tert-butyl-1,4-bis(2-methoxyethoxy)benzene, polyaniline, etc. Successful achievement of the proposed research will lead to the development of new novel radical materials that can be employed in applications such as flexible batteries, fast charging batteries, and structural batteries.

**Student Prerequisites**
Background in physics, chemistry, and materials science, with focus on crystals, molecules, chemical bonding, electrochemistry. Experience with the Linux systems is preferred but not required; the Linux techniques will be trained during the research program.

**Student Duties**
*Data production* by performing first-principles calculations under the mentor’s guidance. The student will be given computational allocations in the mentor’s local computer cluster and national supercomputer centers to perform the calculations. Basic level of analysis on the produced data, to characterize the chemical bonding and identify the stable structure of given materials, are also expected. These results can lead to a discovery of new material for structural batteries. By performing these duties, the student will be familiar with atomistic simulations and quantum calculations, which are becoming a dominant trend in computational materials science, as well as electrochemical energy storage technologies including ORB. At the end of the program, the student will be also given an opportunity to publish a journal paper, which will be useful to demonstrate the student’s research ability and helpful for the future career.

**Mentor Supervision and Interaction**
In the first two-weeks, the student will be trained to have required theoretical background and learn computational techniques through lessons from the mentor; 4 meetings are planned (it will be adjusted depending on the student’s progress). For the rest of the period, the student will perform the research as well as learn higher levels of theory and techniques, as needed. The student will report his/her progress directly to the mentor in weekly meeting. Performance of the research will be assessed based on the amount of data production and the quality of data analysis.