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### Research Seminar

When: September 9, 2020

Time: 2:30 PM

Location: Microsoft TEAMS

# Investigation into the ionic conductivity properties of Ruddlesden-Popper Oxides ( $n = 2$ and $3$ )

## Abstract

Various strategies for enhancing the ionic transport as a function of aliovalent substitution and defects have been investigated through design and synthesis of a series of doubly and triply layered Ruddlesden-Popper Oxides (RP). Several classes of these materials have been examined belonging to the series with general formula  $A_{n-1}A'B_nO_{3n+1}$  ( $A = \text{La}$ ,  $A' = \text{Li}$ ,  $B = \text{Ti/Nb/Ta}$ ,  $n = 2$  or  $3$ ). In RP materials,  $n$  defines the number of octahedral layers within a stack, separated by  $A'$  cations, with  $A$  (typically an alkaline-earth or lanthanide ion) and  $B$ -site (transition metal) cations occupying twelve and six coordinated sites, respectively. Owing to the availability of spaces between stacks in the crystal structure, these family of materials have the potential to be good solid electrolyte for lithium-ion batteries. This work presents the findings of diverse approaches to systematically enhance the ionic conductivity of these materials. Representative doubly layered materials designed through the shortening of lithium hopping distances, demonstrated significantly higher mobility of lithium ions within the structure. The impact of defects in the form of vacancies at various sites within the structure further impacted the ionic conductivities in these materials. Experimental investigations also highlight the cooperative effect of defect creation in both inter- and intra-layer sites (i.e.,  $A$  and  $A'$ ) on ionic transport phenomena through triply layered materials with RP structure. Powder X-ray diffraction, neutron diffraction, DFT, variable-temperature electrochemical impedance, dielectric and modulus studies offered an understanding into the conduction and diffusion mechanism of ion mobility. For instance, DFT calculations on the doubly layered  $\text{Li}_2\text{LaMTiO}_7$  ( $M = \text{Ta}$  or  $\text{Nb}$ ) RPs disclose the orientation of lithium diffusion pathways and the energy barriers which are directly linked to the arrangement of constituent atoms in these materials. These calculations indicate that designing materials with shorter Li-Li hopping distance and defects in the Li-layer can successfully enhance the lithium-ion conductivity in RP oxides. These systematic studies elucidate important aspects of RP oxide properties and can have significant impact on material-design strategies for enhancing the ionic conductivity of layered oxides.

## References

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