Pushing limits of Mixed Glass Former Effect in Lithium Borophosphate glasses

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The mixed glass former effect (MGFE) offers a promising strategy for tailoring the structure and functional properties of amorphous solids. In this study, we explore its limits through systematic compositional tuning of lithium borophosphate glasses within the $40Li_2O$-zWO_3-[(60-z)/4*($3P_2O_5$-$1B_2O_3$)] system. By progressively substituting borate and phosphate units with tungsten oxide while maintaining a fixed P_2O_5:B_2O_3 ratio, we investigate how such dual substitution influences structural evolution and ionic conductivity.

Comprehensive characterization using density and thermal analysis, Raman spectroscopy, and advanced 1D and 2D MAS NMR techniques reveals a gradual transformation from a phosphate-borate to a tungstate-dominated glass network. This shift is accompanied by significant structural depolymerisation, including the formation of isolated phosphate and borate units, emergence of W-O-W linkages, and the growth of mixed tungstate species such as Q^0, Q^1, and B-O-W environments.

Impedance spectroscopy measurements show a remarkable three-order-of-magnitude increase in room-temperature DC conductivity (from 10^{-9} to 10^{-6} Ω^{-1}*cm^{-1}) across the compositional series. This enhancement is directly linked to the evolving glass structure, which facilitates lithium ion mobility through the disruption of rigid phosphate and borophosphate networks and the formation of more accessible conduction pathways.

Our findings confirm that strategic control over glass former composition enables continuous and predictable optimization of ionic transport. These results not only extend the conceptual framework of the MGFE but also demonstrate a viable path for developing high-performance solid electrolytes based on borophosphate glasses.