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Density-Controlled Ion Transport in Amorphous Hafnium Oxide

Ion conduction in ceramic materials is crucial for the performance and reliability of various functional materials, such as gate dielectrics, electrochemical transistors, and memristors. In crystalline materials, ion conduction depends on point defects like vacancies and interstitials, making ionic conductivity directly proportional to defect concentration and mobility. However, in amorphous materials, such crystallographic point defects are undefined. Our study utilized time-of-flight secondary ion mass spectroscopy (ToF-SIMS) depth profiling to measure oxygen tracer diffusion in amorphous hafnium oxide thin films and found that sub-stoichiometric hafnium oxide ($HfO_{1.74}$) exhibits diffusivity two orders of magnitude lower than stoichiometric hafnium oxide (HfO_2). Furthermore, among stoichiometric amorphous hafnium oxide thin films, higher-density hafnium oxide film (9.9 g cm^{-3}) demonstrate lower diffusivity compared to their lower-density counterpart (6.8 g cm^{-3}). This finding indicates that traditional defect chemistry fails to describe diffusion trends in amorphous hafnium oxides. Instead, free volume and film density significantly influence oxygen diffusivity. This work underscores that density and free volume are critical design considerations for ionic conduction in amorphous materials, with implications for designing ionic conductors in various applications, including memory devices, and synaptic transistors.

Topical Area

Hard matter: quantum, electronic, semiconducting materials

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