THE UNIVERSITY OF TENNESSEE KNOXVILLE
DEPARTMENT OF CHEMISTRY

Department of Chemistry
University of Tennessee
Knoxville, Tennessee 37996-1600
Phone: (865) 974-3392
musfeldt@utk.edu

November 17, 2022

Dear Dr. Lenders,

Enclosed, please find our manuscript *"Structural phase purification of* HfO_2*:Y through pressure cycling"* which we are submitting for consideration at Advanced Materials.

As you probably know, amorphous hafnia is widely employed as a gate dielectric in DRAM chips. The recent prediction that flat bands in this material have the potential to support single atom memory combined with the discovery that single crystals can be stabilized using high temperature laser floating zone + rapid cooling in the form of HfO_2:x%Y (x = 0, 7, 11, 12, 20%) opens a number of exciting opportunities to explore the potential of this system. Bottlenecks to the full adaptation of these materials are (i) the presence of yttrium stabilizer along with (ii) the high growth temperatures ($\geq 3000\ ^o$C). The former will damage the flat bands of the polar orthorhombic phase. The latter makes co-processing with silicon more challenging.

In this work, we report a way forward: pressure cycling of HfO_2:x%Y to create pure phase materials such as antipolar orthorhombic hafnia as well as elusive phases such as tetragonal hafnia at room temperature. We test four different pressure pathways and use synchrotron-based infrared absorption and Raman scattering spectroscopies + complementary lattice dynamics calculations to demonstrate the presence and metastable character of the various phases. Analysis of the relative energetics also reveals opportunities to combine pressure and temperature to extend this strategy. We anticipate that novel pressure methods to create and kinetically stabilize pure phase hafnia to advance the development of high performance devices that take advantage of flat band physics are likely to be of great interest to the readership of Advanced Materials.

We look forward to the review of this paper.

Sincerely,

Janice L. Musfeldt
Ziegler Professor of Chemistry and Physics