Electronic Supplementary Material (ESI) for RSC Advances.

Electronic Supplementary Information:

Tunable high-κ $Zr_xAl_{1-x}O_y$ thin film dielectrics from all-inorganic aqueous precursor solutions

K. N. Woods, E. C. Waddington, C. A. Crump, E. A. Bryan, T. S. Gleckler, M. R. Nellist, B. A. Duell, D. P. Nguyen, S. W. Boettcher and C. J. Page*

Department of Chemistry and Biochemistry, Materials Science Institute, University of Oregon, Eugene, OR 97403, USA

*E-mail: cpage@uoregon.edu

Figure S1: Fourier transform infrared spectroscopy (FTIR) for ZAO films annealed at 300 and 400 °C

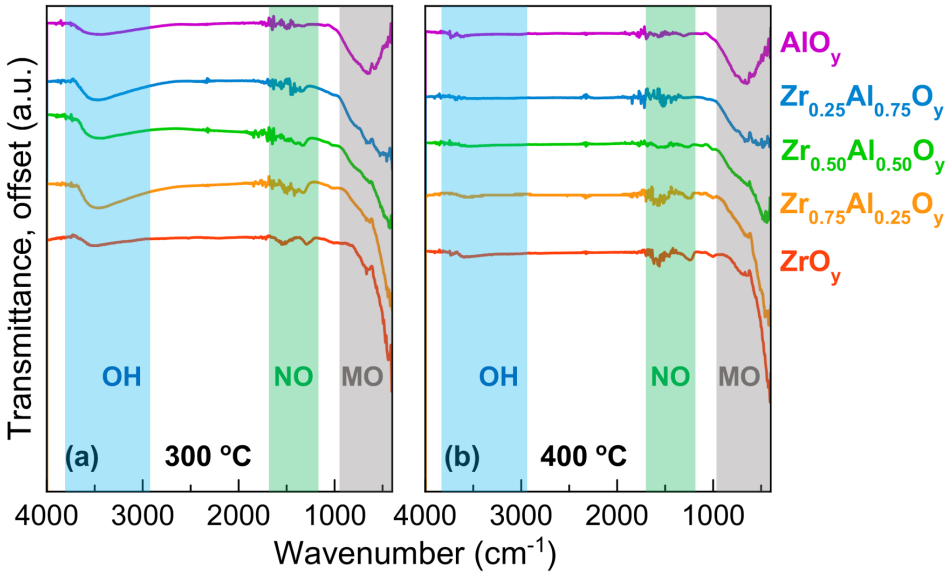

Fig. S1 FTIR spectra of two-coat ZAO films annealed at (a) 300 °C and (b) 400 °C.

Figure S2: X-ray reflectivity (XRR) spectra and best fit models for ZAO films annealed at 500 °C

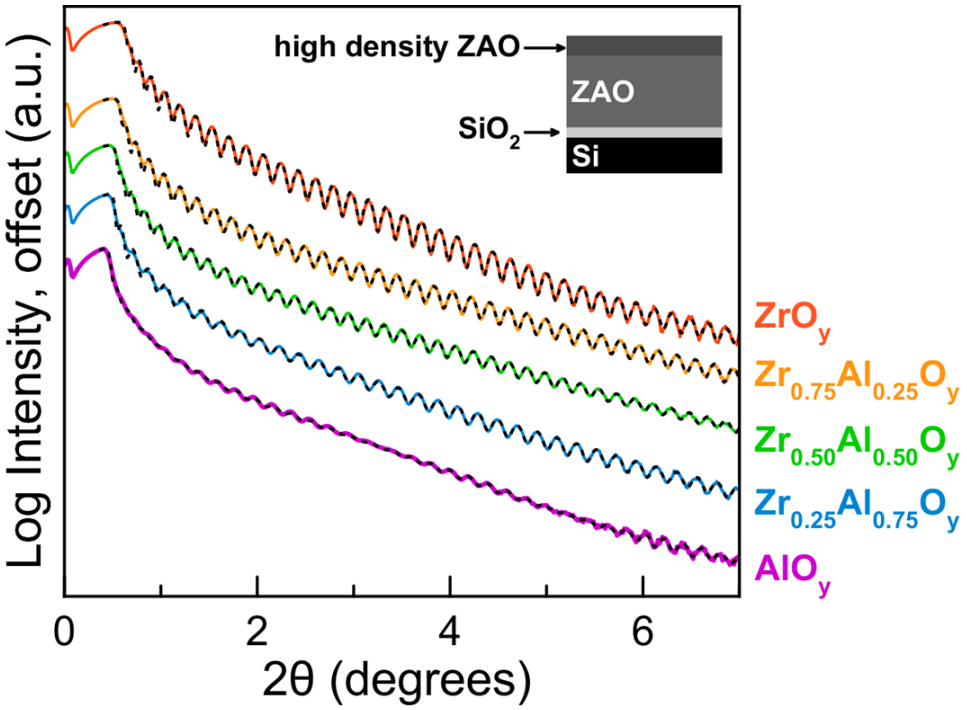

Fig. S2 Representative XRR spectra for single-coat ZAO films annealed at 500 °C (solid, colored) and overlaid best fit models (dashed, black). Best fit models were generated using a two-layer model consisting of a thin (~1-3 nm) capping layer over an underlying bulk layer of different density, consistent with models employed in previous XRR studies on aqueous-derived metal oxides.[1,2]

Figure S3: Film thickness and density of ZAO films extracted from XRR best fit models

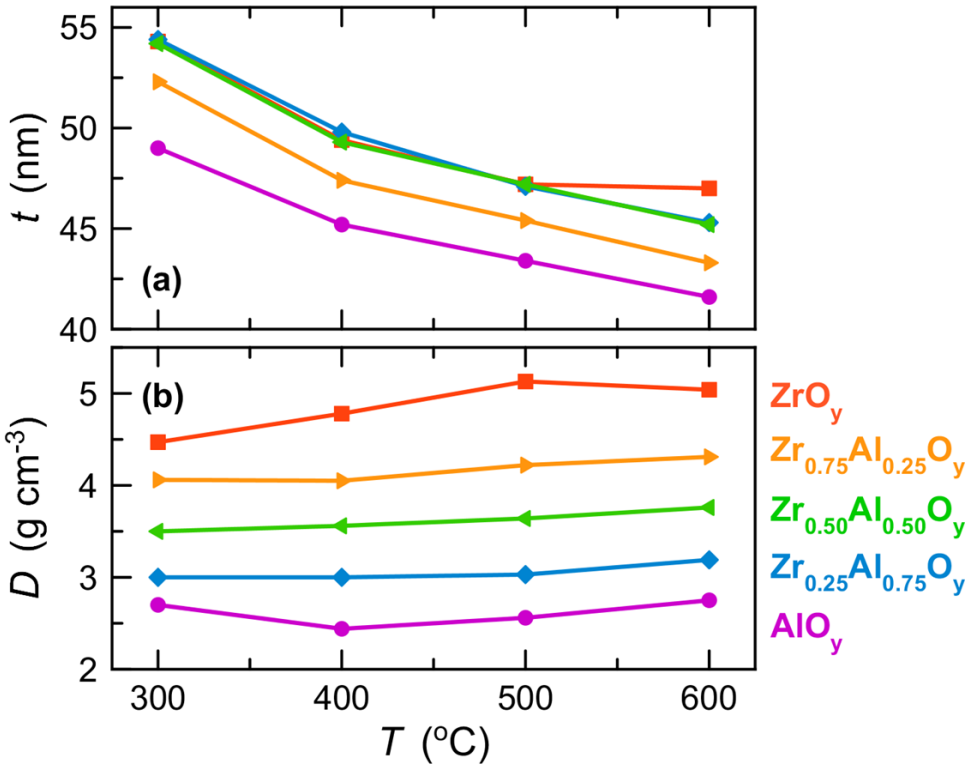

Fig. S3 (a) Film thickness [*t*] and (b) density [*D*] of single-coat ZAO films extracted from XRR best fit, two-layer models. Small differences in film thickness between compositions are attributed to small differences in the humidity of the spin-coating chamber during deposition.[3] Density values are calculated from a weighted average of the capping and bulk layer densities.

Figure S4: Dielectric constant dispersion

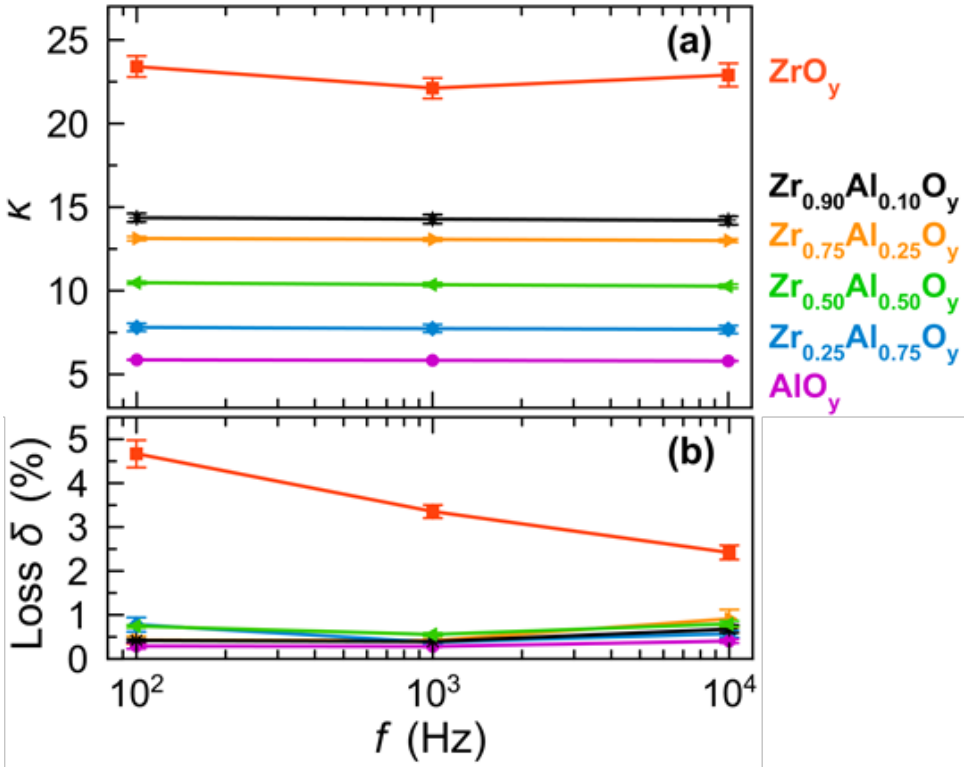

Fig. S4 (a) Dielectric constants [κ] as a function of frequency [f] for ZAO-based MIS devices and (b) corresponding loss tangents [loss δ] as a function of f. MIS devices were fabricated from two-coat (~100 nm) ZAO films annealed at 500 °C. Error bars were determined using measurements taken from three separate batches of MIS devices.

References

1 K. C. Fairley, D. R. Merrill, K. N. Woods, J. Ditto, C. Xu, R. P. Oleksak, T. Gustafsson, D. W. Johnson, E. L. Garfunkel, G. S. Herman, D. C. Johnson and C. J. Page, *ACS Appl. Mater. Interfaces*, 2016, **8**, 667–672.

2 K. N. Woods, T.-H. Chiang, P. N. Plassmeyer, M. G. Kast, A. C. Lygo, A. K. Grealish, S. W. Boettcher and C. J. Page, *ACS Appl. Mater. Interfaces*, 2017, **9**, 10897–10903.

3 P. N. Plassmeyer, G. Mitchson, K. N. Woods, D. C. Johnson and C. J. Page, *Chem. Mater.*, 2017, **29**, 2921–2926.