ALD preparation of high-*k* HfO_2 thin films with enhanced energy density and efficiency for electrostatic energy storages

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S1 Raman spectrum of HfO_2 samples with different thickness.

The peak positions agree with the previous reported Raman spectra of monoclinic HfO2 powder and nanoparticles.[1, 2] These results also indicate that the monoclinic phase might be dominated in annealed HfO_2 films.

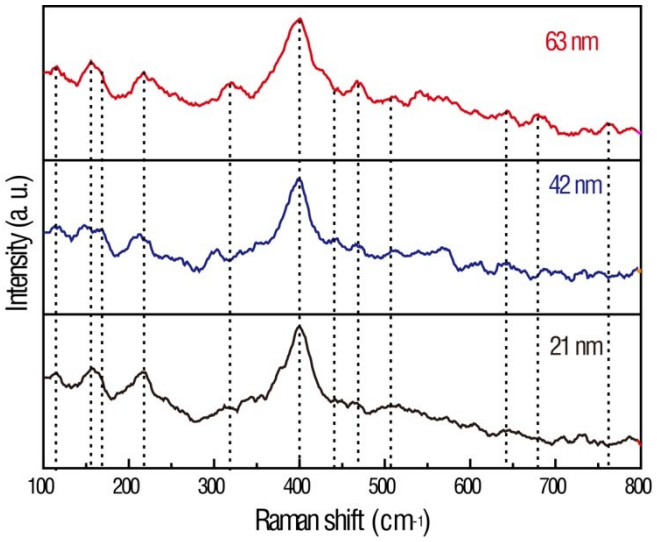

Figure S1 Raman spectrum of HfO_2 samples with different thickness

S2 Hf 4f core level and O 1s core level XPS of HfO_2

As shown in Figure S2, core-level XPS spectra conforms that the annealed film is HfO2. Calibration of core levels was done by setting the carbon 1s peak at 284.6 eV. Peaks at 16.91 eV and 18.61 eV correspond to Hf 4f7/2 and Hf 4f5/2, which is related to the Hf-O bonding in HfO2. The obtained spin-orbit splitting is 1.7 eV, which matches well with other reports.[3, 4] All these evidences confirm the formation of pure HfO_2 films. The XPS spectra of O 1s core-level of the sample is shown in the inset of Figure S2. The peak located at 530.11 eV is attributed to O-Hf in HfO_2 films which are also similar to other reports.[5, 6] The appearance of peak at 532.11 eV may be attributed to O-C bonds, which may due to the residual carbon from the organic metal precursor during the ALD process. [7]

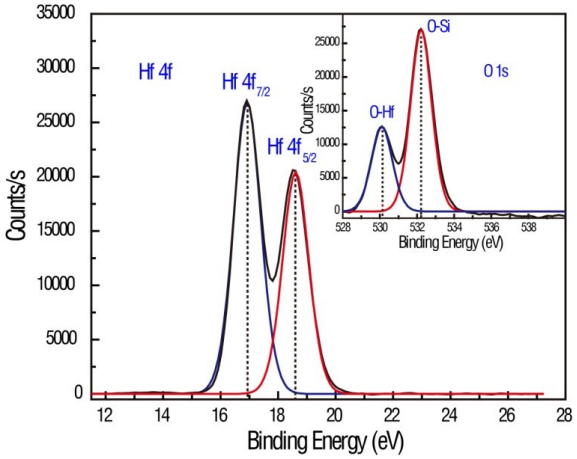

Figure S2 Hf 4f core level XPS of HfO_2 of 63nm, the inset shows the O 1s core level XPS of HfO_2

S3 AFM patterns of the HfO_2 samples with different thickness

The surface morphologies of all the samples are shown in Figure 4, which were measured by AFM. All the HfO_2 films exhibit island-like patterns with clear crystalline grain boundaries. The root-mean-square (RMS) surface roughness is about 2 nm for the annealed films with different thickness, which is obtained in tapping mode taken over a scale of 2 μm×2 μm. The relative smooth surface makes contributions to enhance the breakdown strength, which is the key parameter to remarkably improve the energy density. As increasing the thickness of the film, the grain size becomes larger. The grain size of 42 nm and 63 nm HfO2 films are almost the same. The grain size increases from about 32 nm to 60 nm when the thickness increasing from 21 nm to 42 and 63 nm.

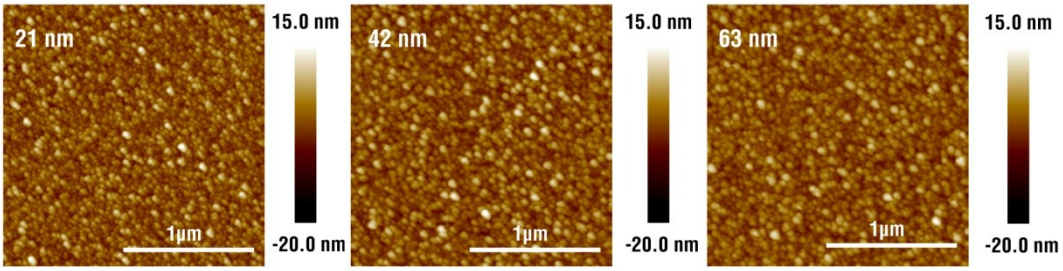

Figure S3 AFM patterns of the HfO_2 samples with different thickness

Reference

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