Investigation of Rare Earth (RE = La, Dy, and Er) doping of HfO_2 by perturbed angular correlation spectroscopy

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The hyperfine interactions at ^{181}Hf(^{181}Ta) probe nuclei in HfO2 samples doped with 5% rare-earth (RE = La, Dy, and Er) elements were measured by perturbed angular correlation (PAC) spectroscopy. Hafnium dioxide has been extensively studied as a potential alternative to silicon dioxide as dielectric gate material in the silicon based complementary-metal-oxide-semiconductors (CMOS) technology due to its high dielectric constant and relatively high thermal stability with respect to silicon. The addition of RE to HfO2 can suppress oxygen vacancies and promote a change in the crystal structure increasing the dielectric permitivity. Samples were prepared using the sol-gel chemical method, starting from very pure metals. The resulting powder of each sample was annealed in air at 550 ^{o} C for 12 h. The crystal structures of samples were checked by X-ray diffraction. Result revealed only a single phase corresponding to the monoclinic phase P 21/C of HfO_2 for all samples. PAC spectra measured at room temperature were fitted with two electric quadrupole interactions with different environments for all samples including a pure HfO_2. The major components (70-75%) were characterized by quadrupole frequencies $\nu_Q \sim 790$ MHz with a distribution around 6% and asymmetry parameter $\eta =$ 0.34 for Dy- and Er-doped samples. La-doped sample showed a slightly lower frequency ($\nu_Q = 727$ MHz) with larger asymmetry parameter ($\eta =$ 0.43) for the major component. The major components are ascribed to probe nuclei at regular monoclinic sites of HfO_2. The quadrupole frequency ν_Q and asymmetry parameter η for Dy- and Er-doped samples agree well with those values reported in the literature [1]. The minor components were characterized by $\nu_Q \sim$ 760 MHz with a distribution around 8% and asymmetry parameter η = 0.75 for Dy- and Er-doped samples and ν_Q = 783 MHz and η = 0.61 for La-doped HfO_2. These values are quite different from those reported for pure sample [1], assigned to probe nuclei near defects, and probably they are due to probe nuclei close to RE dopants, which reduce the presence of defects.

[1] M. Forker, P. de la Presa, W. Hoffbauer, S. Schlabach, M. Bruns, and D. V. Szabó, Phys. Rev. B 77, 054108 (2008)