Effect of compression on X-ray intensity associated with nuclear decay of Hafnium

P. Das[1], G. Mandal[1], A. Kumar[2], B.S. Tomar[2]
[1]*Variable Energy Cyclotron Centre,1/AF Bidhannagar, Kolkata-700064*
[2]*Radio Analytical ChemistryDivision, Bhabha Atomic Research Centre, Mumbai - 400085, INDIA*
** email: parnika@vecc.gov.in*

Introduction

The K_β/K_α X-ray intensity has been easily measurable quatity with relatively high precision and has been studied extensively for K-X-ray emission by radioactive decay, photo-ionization (PI) and charge particle decay [1]. It is well established that the intensity ratio is dependent on the electronic environment of the atom which emits X-rays. Relativistic self-consistent field calculations were performed by Scofield [2] and experimental results are observed to be in agreement. However, Hansen et al. [3] measured K-Xray intensity ratio following K-capture decay of radioactive nuclei and observed considerable difference between K_β/K_α ratio by electron capture (EC) and those by X-ray or electron bombardment. In case of 3d transition elements, it was pointed that the reason for the difference is due to the 3d excess electron in EC and the strong shake off process in PI. So, the technique of X-ray intensity ratio measurements gives information regarding the electronic environment around the X-ray emitting atom depending on the excitation process. In this work, the technique has been employed to study the change in the electronic environment around the Hafnium nuclei under compression.

Experimental Work

In this experiment nat HfO_2 powder was irradiated by thermal neutrons using Dhruva facility and left for a long period of cooling (about 90 days). The sample was annealed and small amount of irradiated sample spectra was taken using a HPGe detector. Only gamma lines observed confirmed the presence of active ^{175}Hf ($T_{1/2}$=70 days) and ^{181}Hf ($T_{1/2}$=42.39 days). A point source was prepared and a spectrum was obtained using a LEPS detector, shown in Fig 1. Calibration was done using a Eu source with 41.542 keV X-ray and 121 keV γ-ray. The K_{alpha} and K_{beta} lines of Hf are shown in Fig 1 inset.

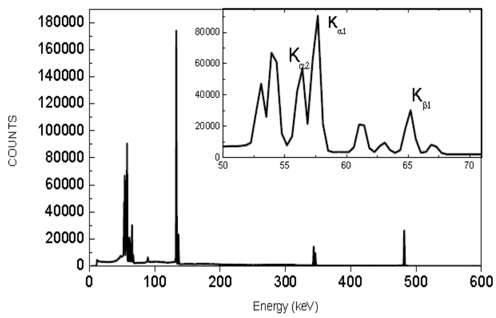

Fig. 1 Spectrum of a point source of an annealed neutron irradiated HfO_2

The irradiated HfO_2 annealed was then loaded in a Diamond Anvil Cell (DAC) (Almax Industries, Belgium) with a diamond culet size of 400 µm. Samples are loaded inside the central hole (120 µm diameter) of the stainless steel gasket, pre-indented to thickness of 64 µm. Silicone oil is used as the pressure transmitting medium. The sample was studied at 18 GPa, 33 GPa and 45 GPa using LEPS. Each spectrum was recorded for 1 hour keeping same geometry. The pressure was measured using a RUBY LUX spectrometer from the shift in ruby fluorescence obtained from ruby crystals loaded in DAC along with the sample under study.

We also performed an experiment at Elettra synchrotron facility to study the structural evolution of HfO_2 under compression and X-ray diffraction measurements were done using DAC mentioned above.

Results

The spectra obtained using LEPS were fitted the K_α peaks with a two peak fitting and K_β with a single peak and determined the ratio of $K_{\alpha1}/K_{\alpha2}$ and $K_\beta/K_{\alpha1}$. Variation of these parameters with pressure is shown in Fig 2. We find analyzing XRD data that there is a phase transition of HfO_2 which monoclinic at ambient pressure. At 18 GPa, 33 GPa and 45 GPa HfO2 has orthorhombic structure and observed volume is 128.18 Å, 123.04 Å, 120.33 Å respectively.

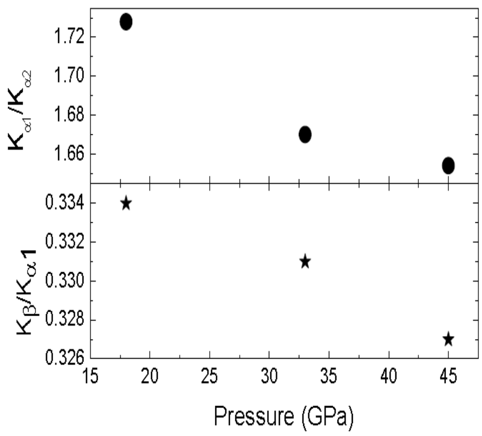

Fig. 2 (a) Variation $K_{\alpha1}/K_{\alpha2}$ with Pressure
(b) Variation $K_\beta/K_{\alpha1}$ with Pressure

Conclusion

From our preliminary analysis we find that there is volume compression in HfO_2 lattice. We also observe that there is a decreasing trend in the ratio of $K_{\alpha1}/K_{\alpha2}$ and $K_\beta/K_{\alpha1.}$ Since the data was recorded for 1 hour in close succession, we may conclude that the excitation mechanism remained same. We find that the area under 482 keV were equal within one standard deviation considering statistical error. Our data analysis and determination of error on the parameters is in progress.

The technique has a potential to be used as a pressure calibrant in high pressure studies.

References

[1] Takeshi Mukoyama et al., Advances in Quantum Chemistry, Vol37, pp 309.

[2] J. H. Scofield, Phys. Rev. A**9**, 1041 (1974).

[3] J. S. Hansen et al. Nucl. Phys. **A142**, 604 1970