



Ion beam induced cubic to monoclinic phase transformation of nanocrystalline yttria



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ABSTRACT

Sol gel derived nanocrystalline yttria pellets are irradiated with 120 MeV Ag⁹⁺ ions for fluence in the range 1×10^{12} – 3×10^{13} ions cm⁻². Pristine and irradiated samples are characterized by X-ray diffraction (XRD), transmission electron microscopy (TEM) and Raman spectroscopy. XRD pattern of pristine Y₂O₃ nanocrystal reveal cubic structure. A new XRD peak at 30.36° is observed in pellet irradiated with 1×10^{13} ions cm⁻². The peak at 30.36° is corresponding to (40 $\bar{2}$) plane of monoclinic phase. The diffraction intensity of (40 $\bar{2}$) plane increases with Ag⁹⁺ ion fluence. Raman spectrum of pristine pellet show bands corresponding to cubic phase. And, ion irradiated sample show new peaks at 410, 514 and 641 cm⁻¹ corresponding monoclinic phase. HR-TEM and SAED pattern of ion irradiated sample confirmed the presence of monoclinic phase. Hence, it is confirmed that, 120 MeV Ag⁹⁺ ions induce phase transformation in nanocrystalline Y₂O₃.

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1. Introduction

Recently nanocrystalline yttrium oxide (Y₂O₃) attracted extensive research interest due to its unique optical, electrical, chemical and thermal properties. Y₂O₃ finds wide usage in a many luminescent host materials, medical diagnostics as well as biological, industrial and research fields [1–3]. Cubic phase of Y₂O₃ exhibit wide transparent range from UV (220 nm) to infrared (~8 μm) region, it is optically isotropic and hard, having high refractive index (~1.92). It possess high corrosion resistivity, high radiation stability, high melting point (~2723 K), large band gap (5.72 eV) and low phonon energy (~380 cm⁻¹) which leads to very narrow emission and enhanced quantum efficiency [4,5].

Y₂O₃ exist in cubic (C-type), monoclinic (B-type) and hexagonal (A-type) structures. C-type structure is stable at room temperature and normal pressure. Under high temperature or pressure, the C-type will be compressed and leading to reduction in the Y–O bond length. Thus, the cubic structure will lose stability and atoms will rearrange to form a higher density phase [6]. Hence, it is important to study the phase formation of different polymorphs in nano size regimes in particular cubic to monoclinic phase transition.

Swift heavy ions (SHI) have been explored by researchers in different ways in the field of material science. The energy of the ion, ion fluence and ion species greatly affect the structural properties of materials. SHI penetrates deep into the target material; lose their energy predominately through inelastic collisions with the target electrons. The resulting intense electronic excitation can produce a narrow trail of permanent damage along the ion path called ion track [7,8]. Also, it produces point defects and defect clusters. In the recent years there has been a tremendous interest in the studies of phase transformation under SHI. A few researchers reported the phase transformation of single crystals, thin films and polycrystalline Y₂O₃ under energetic heavy ion irradiation [9,10]. In the present work, 120 MeV Ag⁹⁺ ion induced phase transformation of sol-gel synthesized nanocrystalline yttrium oxide is reported.

2. Materials and methods

Nanocrystalline yttrium oxide is synthesized by the sol gel technique using yttrium (III) nitrate hexahydrate (Y(NO₃)₃·6H₂O – Aldrich chemicals), citric acid anhydrous GR (C₆H₈O₇) and 25% GR ammonia solution (NH₃) (Merck specialties private limited). The ratio of citric acid/Y³⁺ is equal to 2. The detailed experimental procedure was discussed elsewhere [11]. Pellets (5 mm diameter and 1 mm thick) of Y₂O₃ are prepared by applying a pressure of

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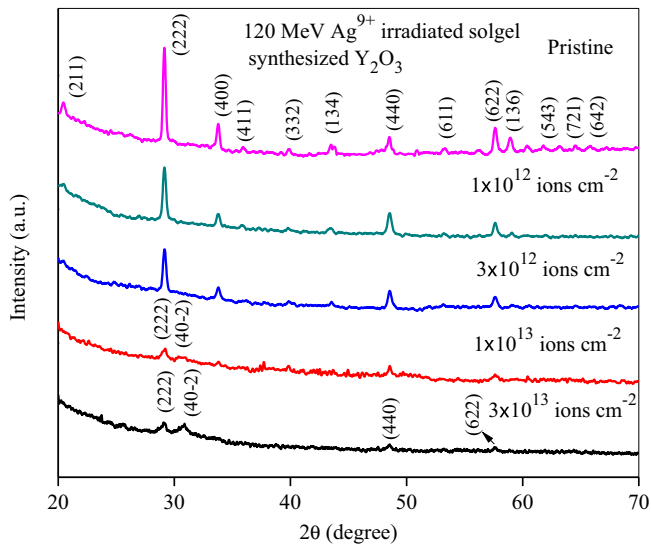


Fig. 1. X-ray diffraction patterns of pristine and 120 MeV swift Ag^{9+} ion irradiated Y_2O_3 .

4 MPa using a homemade pelletizer. The pellets are annealed at 900°C for 2 h in a muffle furnace. One of these pellets is kept as pristine and the others are irradiated with 120 MeV Ag^{9+} ions in the fluence range 1×10^{12} – 3×10^{13} ions cm^{-2} at Inter University Accelerator Center, New Delhi, India.

The pristine and irradiated samples are characterized by X-ray diffraction (XRD) method [Bruker D8, using Cu-K_α radiation of wavelength 1.5406 \AA]. The morphology and atomic structural information of the samples are studied by transmission electron microscopy (TEM) high resolution TEM (HR-TEM) and selected area electron diffraction (SAED) techniques [MIRA II LMH from TESCAN]. The powder sample of sol-gel synthesized Y_2O_3 is dispersed in ethanol for ~ 30 min and a drop was placed on the surface of a carbon-coated grid (300 mesh) to record TEM micrographs. Raman spectra are recorded in the range 200 – 1050 cm^{-1} using ID Raman micro-785 Ocean Optics microscope with an excitation wavelength of 785 nm .

3. Results and discussion

3.1. X-ray diffraction

XRD patterns of pristine and 120 MeV Ag^{9+} ion irradiated yttrium oxide are shown in Fig. 1. Pristine sample exhibits XRD peaks centered at 20.44° , 29.14° , 33.78° , 35.98° , 39.79° , 43.51° , 48.52° , 53.30° , 56.21° , 57.62° , 58.94° , 60.40° and 61.80° corresponding to (211), (222), (400), (411), (332), (134), (440), (611), (541), (622), (136), (444), (543) respectively. These planes are indexed to the cubic phase of Y_2O_3 with the space group $\text{Ia}\bar{3}$ (JCPDS No. 88-1040) [12]. At lower fluence (1×10^{12} – 3×10^{12} ions cm^{-2}) 2θ positions of XRD lines closely match the values of pristine sample. However, the relative intensities are less due to loss of crystallinity with increase of ion fluence. Further, a weak and new diffraction peak at 30.36° is observed at a fluence 1×10^{13} ions cm^{-2} . This peak corresponds to the monoclinic phase with space group C2/m [9,10,13]. This peak is assigned to (40 $\bar{2}$) plane (JCPDS 47-1274). The intensity of all the diffracted peaks of the cubic phase decreases with ion fluence where as the peak correspond to monoclinic phase (40 $\bar{2}$) increases. Thus, integral intensity of all the diffraction peaks (cubic phase) exponentially decreases with increasing ion fluence. It is due to the fact that,

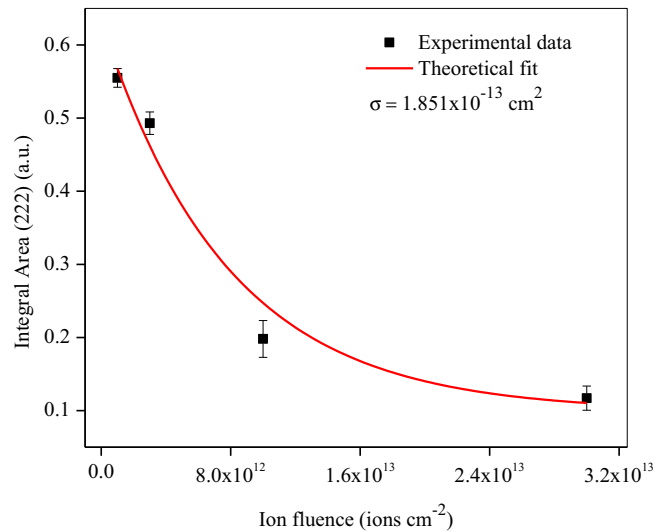


Fig. 2. Variation of integral area of (222) plane with ion fluences in Y_2O_3 .

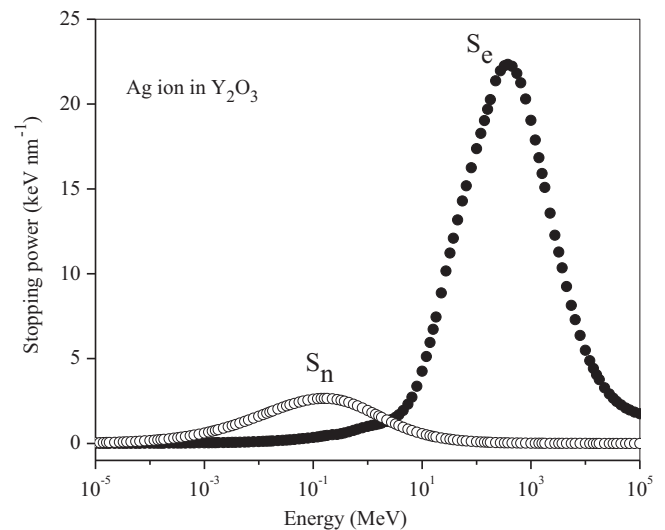


Fig. 3. Variation of electronic energy loss (S_e) and nuclear energy loss (S_n) for Ag ion in yttrium oxide target.

Table 1
Ion impact parameters.

Ions	Energy (MeV)	S_e (keV nm^{-1})	Range (μm)	Target type	References
Si^{8+}	100	4.103	21.65	Nanocrystal	Lakshminarasappa et al. [17]
Xe	0.38	18.0	–	Thin film	Gaboriaud et al. [15]
Ag^{9+}	120	18.270	11.10	Nanocrystal	Present work

ion irradiation leads to the damage of Y_2O_3 lattice and induces more number of oxygen vacancies, extended defects and internal stress in materials and these may also responsible of the phase transformation. A strong intense diffraction peak at 29.14° corresponds to (222) plane is selected for the analysis. The effect of ion fluence on integral intensity of 29.14° peak is fitted using by Poisson equation [14,15]

$$I(\varphi) = I_0 \exp(-\sigma\varphi) \quad (1)$$

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