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He^+ induced changes in the surface structure and optical properties of RF-sputtered amorphous alumina thin films

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ABSTRACT

High quality 50 nm thick stoichiometric amorphous aluminium oxide films were reactively sputtered on microscope glass slide substrates. The films were exposed to energetic (2.20 MeV) He^+ at different ion fluences of 6×10^{12} ion/cm^2; 1×10^{13} ion/cm^2; 2×10^{13} ion/cm^2; 3×10^{13} ion/cm^2; and 4×10^{13} ion/cm^2. The effect of the ion irradiation on the optical, structural phase and surface properties of the alumina films was investigated via UV–VIS–NIR spectroscopy, X-ray diffraction analysis and the atomic force microscopy respectively. The transmission and absorption spectra of the irradiated films showed variation that depended on ion fluence. The refractive index, extinction coefficient, optical conductivity, dielectric constant and energy loss functions of the films were also affected by He^+ irradiation. Optical band gap and films' structural phase were however not altered by the ion irradiation. The variation in optical constants induced by radiation was attributed to electronic excitation and increase in surface roughness of the films.

1. Introduction

The exposure of solid materials to particulate and electromagnetic ionising radiation has been established [1–3] to have tremendous micro-structural effects on the solid. For materials in thin film form, these effects are strongly dependent on the film's chemical nature, thickness, structural phase, and radiation parameters such as: energy, linear energy transfer (LET), fluence, mass, charge, and dose [1–9]. In many cases, the changes in the micro-structure manifest as optical and electrical properties variations.

Photon and ion irradiation of thin films and their devices could be deliberate due to some inherent benefits. The use of radiation for material characterisation is one of such beneficial intentional exposure. The radiations that are commonly used for material characterisation include: photons in X-ray diffraction (XRD); electrons in scanning electron microscopy (SEM), transmission electron microscopy (TEM) and electron beam lithography; ions such as protons and helium in PIXE and RBS respectively. Although these techniques provide useful information about material structure and constituents, they can however cause serious temporary/permanent unplanned modification to the properties of thin film materials. The understanding of interaction and effect of irradiation of materials during these analytical procedures is thus highly essential. On the other hand high energy photon and ion irradiation has become an effective tool for introducing [9–16] planned modification in the structural, mechanical, electrical and optical properties of thin film based materials and devices. In such instances, the tailored properties are chosen based on the requirement of the desired application. However, for such thin film's property engineering, prior knowledge of the effect of different radiation parameters is required. This possibility of modifications of thin film properties by irradiation requires that attention should be paid to this effect when developing thin film devices for use in radiation environments. Therefore, the effects of irradiation on such thin film materials/devices, if not known before, must be established.

Aluminium oxide in thin films is a material that has been used in several technological applications due to their excellent optical properties [17–21]. One of the commonest optical applications of alumina thin films is in protective optical coatings due to its high: transparency [17], dielectric constant [19,20], refractive index [18], chemical and thermal stability [21,22]. In nuclear science and technology, alumina thin films are used in fusion reactors [23] and have a potential of being used as RF windows in International Thermonuclear Experimental Reactor [24]. A crucial problem that may occur, as earlier pointed out in such radiation environment is radiation induced modification of its properties. Design of optical coatings such as antireflective coatings or filters requires that such properties remained unaffected by harsh environmental conditions such as ionising radiation exposure. Consequently, the possible use of alumina film based materials for optical functions in radiation environment requires prior knowledge of the variation of its properties with radiation quality factor and dose. Furthermore, aluminium oxide thin film has a potential to be used as thin film layer for protective coating for luminescent phosphors used for radiation dosimeters (luminescence phosphor). For such applications, the high transparency

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of the layer for the emission wavelength of the luminescence material is essential to be constant with respect to radiation dose so as to avoid radiation induced absorption losses in the layer. In view of the practical importance of alumina thin film in radiation environment, the properties of the film under various radiation condition is worthy of investigation. There have been previous studies on the effect of neutron irradiation on the electrical [25] and optical [26] properties of bulk crystalline alumina and also the effect of photons on the electrical property of thin film alumina [27]. The effect of ion irradiation on the optical properties of the thin film has however not been investigated.

In this study, the $^4He^+$ induced changes in the optical properties of stoichiometric alumina films deposited by Olarinoye and Ogundare [28] using RF sputtering technique was investigated. The radiation resilience of alumina films will be established. The relationship between $^4He^+$ dose and optical constants and surface roughness of alumina films will also be determined.

2. Experimental details

Aluminium oxide films were deposited on cleaned 1.20 mm thick 25 mm × 76 mm glass (microscope slide) substrates in a RF (13.56 MHz) sputtering unit (Edwards Auto306). Al (99.99%) of 10 cm diameter was used as the target while argon (99.99%) and oxygen (99.99%) gases as sputtering and reactive gas respectively. Stoichiometric, dense and high refractive index alumina film was obtained at 11% oxygen flow ratio, RF power of 300 W, annealing temperature of 573 K and Argon flow rate of 1.0 sccm [28]. Further details on the deposition are contained in the work of Olarinoye and Ogundare [28].

The deposited films were irradiated with different doses of 2.2 MeV $^4He^+$ at low and constant ion currents to avoid thermal degradation of the films and substrates. The irradiation was done using 2.2 MeV $^4He^+$ beam from ion beam analysis facility at the Centre for Energy Research and Development, (O.A.U.) Ile-Ife, Nigeria. This facility is centred on a NEC 5SDH 1.7 MV Pelletron Accelerator, equipped with a RF charge exchange ion source. The ion source is equipped to provide proton and helium ions. The irradiated area of the film was 1 cm^2 and samples were mounted perpendicular to the beam. The irradiation was carried out at 6×10^{12} ion/cm^2; 1×10^{13} ion/cm^2; 2×10^{13} ion/cm^2; 3×10^{13} ion/cm^2; and 4×10^{13} ion/cm^2 designated as R1, R2, R3, R4 and R5 respectively.

The films' structure and phase were analysed before and after irradiation using an X-ray diffraction (XRD) unit (PAnalytical X'pert Pro MPD) with Cu–K-α photons. To avoid the contribution of the substrate, XRD at grazing angle was used. Optical characterisation of the film before and after irradiation was studied by measuring and recording the optical transmission and reflectance spectra for photon wavelength between 200 nm and 900 nm at room temperature using a UV–VIS spectrophotometer (Thermo Scientific Helios Omega). In order to correct for the absorption of the substrate and normalise transmittance measurements, absorption and transmittance measurement of blank pristine and irradiated substrate were carried out. Consequently, the effect of bare and irradiated substrate was eliminated in the optical measurements. Effects of ion irradiation on the surface morphology of the films were investigated with an atomic force microscope (AFM).

3. Calculation

3.1. Optical constants

Refractive index (n) and extinction coefficient (k) are two parameters which characterise how a material interacts with electromagnetic radiation. The refractive index is a measure of electronic polarisation of ions and local fields inside a material. The evaluation of n is thus of high importance for many applications especially in optical devices. On the other hand, k represents the imaginary part of a complex refractive index; it is a measure of the (exponential) decay of photon as it moves through a medium. In this study, the refractive index, and the extinction coefficient, of the films were calculated from measured reflectance (R) and transmittance (T) data according to the equations [29,30]:

$$n(\lambda) = \frac{1+\sqrt{R(\lambda)}}{1-\sqrt{R(\lambda)}} \tag{1}$$

$$k = \frac{\alpha(\lambda)\lambda}{4\pi} \tag{2}$$

where α is the absorption coefficient defined by the relation [31]:

$$\alpha(\lambda) = \frac{2.303}{d}\log_{10}\left(\frac{1-R}{T}\right). \tag{3}$$

3.2. Optical band gap energy

Determination of the band gap energy E_g requires an analysis of the energy dependent absorption coefficient (α). According to Tauc [32], the absorption coefficient in the high absorption region is dependent on the photon energy according to the relation:

$$\alpha(\upsilon)h\upsilon = B\left(h\upsilon - E_g\right)^{m/2} \tag{4}$$

where B is an energy dependent constant, m is a number which characterises the process of absorption, equal to 1 and 4 for direct allowed transition and indirect transition respectively. The determination of E_g was made by extrapolating the linear portion of the curves until they intercept the photon energy axis.

3.3. Packing density of thin film

To evaluate the film porosity and hence the relative density of the pristine and irradiated film with respect to corundum, the effective medium theory [33] was used according to the equation:

$$(1-p) = \frac{{n_f}^2 - 1}{n^2 - 1} \tag{5}$$

where p is the porosity of the film, n and n_f are the refractive indices of bulk alumina and that of the deposited film respectively. The packing density (1-p) can be used to estimate the density of the film by multiplying it with the density of the bulk material.

4. Results and discussion

4.1. Structural phase

The XRD spectrum of the pristine film presented in Fig. 1 suggested that the deposited alumina film was predominantly amorphous. As shown in Fig. 1, the spectra of the irradiated films do not differ from that of the pristine film. The structural phase of the amorphous alumina is thus not affected by He^+ radiation within the fluence range considered here.

4.2. Ion energy loss and range

The irradiation of energetic ion on solid materials leads to interaction between the ion and the solid resulting in energy loss by the ion and consequently the penetration of the ion in the material up to a certain depth (range) before coming to rest. The energy loss could arise due to inelastic electronic excitations (electronic stopping) as well as by elastic collisions with atoms (nuclear stopping). The projected range, electronic and nuclear stopping of the 2.2 MeV helium ion in the film calculated using the SRIM (Stopping and Range of Ions in Matter) code