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Dosimetric properties of Cr doped Al_2O_3 nanophosphors

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ABSTRACT

A novel series of Cr_2O_3-Al_2O_3 nanoparticles containing various molar compositions of Cr_2O_3 (0–2 wt%) were prepared by sol-gel method. An octadecylamine was used as nano-assembling template to prepare novel dosimeter nanoparticles. The crystalline features, morphology, sample composition and the nanostructure of the prepared samples were investigated by X-ray diffraction (XRD), Scanning electron microscopy (SEM), Energy dispersive X-ray (EDX) and high resolution transmission electron microscopy (HRTEM). The XRD and HRTEM results reflected the existence of nanoparticles with rod-like structure. It was found that Al_2O_3 nanoparticles with compositions of 1.5% Cr_2O_3 exhibited the highest response for gamma rays than all different compositions. This composition displayed a linear gamma dose response in the range from 3.5 mGy up to 350 Gy. The investigated samples exhibited a difference of the sensitivity for beta rays according to the applied dose ranges. The minimum detectable dose of the prepared samples was evaluated as 27.2 μGy. The results showed that no fading was exhibited for the investigated samples by measuring one day after the irradiation process.

1. Introduction

Recently, there are great opportunities in using nanoparticles in the field of luminescence, especially since they exhibit enhanced optical, electronic and structural properties. Many new physical and chemical methods of preparations have also been developed in the last two decades to prepare nanoparticles and nano rods of definite structure [1–3]. More recent investigations have displayed that luminescence, optical and other properties depend on the particle size and its shape, incorporation of impurity at different sites and also due to the presence or absence of certain defects. Thermoluminescence is the emission of light during thermal stimulation of the investigated insulator or semiconductor, following a previous absorption of energy from ionizing radiation. This thermoluminescence (TL) phenomenon has been involved in many physical applications. The main applications are in detection and measurement of absorbed radiation and detecting defects in solids. It is more popular for the dosimetry of ionizing radiations, since the TL intensity is usually proportional to the applied radiation doses. Alumina is a low cost material most widely used as a catalyst and as an active dosimeter [4–6]. The corundum or α-alumina has excellent mechanical, electrical, thermal and optical properties due to hexagonal close packing of oxygen ions [7–9].

On the other hand, transition alumina, including α-Al_2O_3, have a cubic close packing of oxygen ions resulting in high surface area, mesoporosity and surface acidity. As a result of these important properties, α-Al_2O_3 is also extensively used as an adsorbent and a membrane. Nano alumina can be a good candidate for luminance materials because of the presence of some luminescent centers such as; OH [10], non-bridging oxygen hole centers [11], oxygen vacancies [12], V-type centers [13], the electron/hole traps produced by the defects, and other impurity defects. The alumina nanoparticles also have outstanding luminescent properties not found in bulk materials. In various investigations, the challenges have been devoted to achieve the preparation of α-Al_2O_3 powders by using various chemical routes to obtain a thermally stable α-Al_2O_3 at fixed preparation conditions. The evaluation of the reactivity of α-Al_2O_3 prepared from aluminum nitrate, chloride and sulfate precursor via a stringent control of composition, surface area, porosity and surface acidity are also essential.

However, little research has been conducted to prepare alumina from organometallic precursors in order to obtain a homogeneous matrix structure in nano dimensions. Various chemical routes have been conducted; however, the sol–gel route offers a tremendous opportunity for controlling the physical, chemical and textural properties of the aluminum oxide. However, the manipulation of particle and pore structure is still an obstacle that limits the sample applications. Alumina nano rods are expected to exhibit high surface area that improves the

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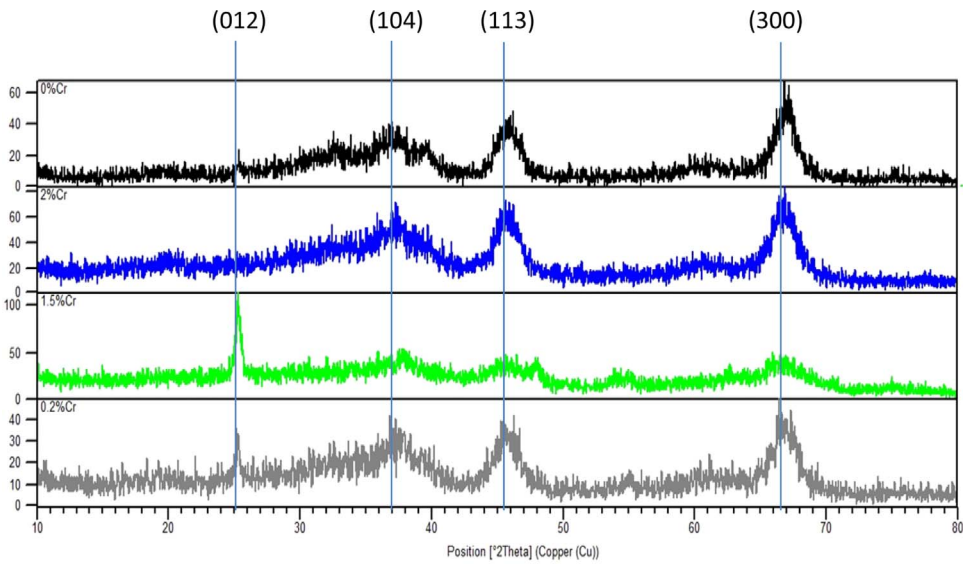

Fig. 1. XRD analysis of α-Al_2O_3 doped by nano-powder Cr with different concentration 0%, 0.2%, 1.5%, 2%.

exposure of particles towards radiation doses. Moreover, modification of alumina with transition elements as chromium oxide is expected to increase the number of defects in alumina structure and improve its absorption ability. Alumina-chrome (Al_2O_3–Cr_2O_3) is a simple binary system which exhibits a complete substitutional solid solution at high temperature without formation of any eutectic cover the entire range of composition [14–16]. Chromia (Cr_2O_3) has long been used to improve the physical properties of Al_2O_3. Chromia forms a solid solution with Al_2O_3 over the full range of compositions. The addition of Cr_2O_3 leads to increasing hardness, tensile strength, and thermal shock resistance of Al_2O_3. Therefore, it is important to synthesize α- Al_2O_3 with controllable and reproducible properties to get the stable and reactive oxide. It is also necessary to investigate the effect of various chromium oxide concentrations on the physicochemical properties of the solid, with particular attention given to the changes its irradiation features.

The present investigation focuses on elucidating the effects of varying chromium oxide concentration on the physicochemical features and radiation properties of α- Al_2O_3 prepared by sol-gel route using octadecylamine as template.

2. Materials and methods

In the present research, appropriate amounts of aluminum-propoxide and chromium sulfate dissolved in isopropanol are mixed together in certain proportions in order to obtain mixed oxides with various Cr_2O_3 composition (0.1–5 wt%). An appropriate amount of octadecylamine just above the critical micelle concentration is added to the above mixture with constant stirring for one hour. Then, drops of ammonia solution (1 N) are added to the mixed salt solution until turbid sol is formed. Then, the mixture is subjected to vigorous stirring for two hours. The sol particles are subjected to condensation for two days until sol particles condense into solid gel particles. Filtration is followed by washing with distilled water to remove foreign ions. Finally, the solid is dried at 100 °C then heating to 800 °C for 2 h with a rate of 10°/min.

The X-ray diffraction (XRD) pattern of a given material was obtained using a fully computerized X-ray diffractometer, Shimadzu XRD-6000, Fig. 3.2: b with Cu radiation of λ = 1.54056 Å. The X-ray tube was operated at 40 kV and 30 mA anode current throughout the measurements. The pattern was recorded at a scanning rate of 80/min. The above operation conditions were maintained during all the relevant measurements. The 2θ degrees scan range is from 20° to 70°. Continuous scanning with scan speed, 8(°/min) and pre-set time of 0.15 s was used for the entire measurement. The obtained diffraction pattern of the sample is compared with "Joint Council Powder Diffraction data (JCPDS)" for standards. This gives information of different crystallographic phases, the relative abundance and preferred orientations.

The chemical composition of the prepared samples was investigated by energy dispersion X-ray EDX detector linked to the scanning electron microscope (JEOL SEM5400). Full quantitative analysis results were obtained from the spectra by processing the data through Z_{af} correction program. During the quantification step of the X-ray analytical process, the measured X-ray intensities are converted into elemental concentrations. The nanostructure of the prepared samples was investigated by high resolution transmission electron microscopy (JEOL 2000FX microscope).

To investigate the dosimetric properties, the TL measurements were achieved by RISO-TL-DA-12 reader with a built-in $^{90}Sr/^{90}Y$ β-source with rate (3.33 mGy/s) at the Institute of Radiation Protection, Helmholtz research centre, Munich, Germany.

3. Results and discussion

3.1. X-ray diffraction

The XRD patterns of the system Al_2O_3-xCr_2O_3 with x = 0, 0.01, 0.05, 0.2, 1 and 2 wt% annealed at 800∘C are shown in Fig. 1 The XRD pattern of alumina powder represents the crystalline pattern of α-phase of Al_2O_3. The peaks observed at 2θ degrees = 26.6°, 39.5°, 46.7° and 68.8° are attributed to the α-phase of alumina. On careful examination of Fig. 1, one can notice, the disappearance of the peak centered at 26.6° upon increasing Cr_2O_3 contents revealing its adsorption on this crystalline site. Examining XRD figures, one can notice the remarkable peak broadening for the doped samples compared with pure Al_2O_3, indicating that Cr_2O_3 can effectively inhibit Al_2O_3 crystallites from further growing up during the process of calcination. A remarkable shift is noticed in the position of alumina crystalline peaks suggesting that part of Cr^{+3} ions are incorporated in substitution positions in the Al_2O_3 lattice replacing the Al^{3+} ions in the crystalline positions. The formation of the solid solution between alumina and chromium oxide is observed due to the similarities of ionic radii of Al^{3+} and Cr^{3+}. The remainder of Cr_2O_3 particles is diffused on the grain boundary surface of alumina. This implies that part of the Cr_2O_3 exists as a highly dispersed amorphous oxide coating on the Al_2O_3 nano crystallites and suppresses the grain growth of Al_2O_3. The average crystallite size calculated according to Scherrer equation varies between 6 and 11 nm confirming the successful synthetic method in preparing nano alumina.