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Thermoluminescence dosimetry features of DY and Cu doped SrF_2 nanoparticles under gamma irradiation

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HIGHLIGHTS

- The SrF_2:Dy and SrF_2:Cu nanoparticles were synthesized for the first time.
- The co-precipitation method as an easy and low cost method was used.
- A linear dose response in a broad range of the absorbed dose was achieved.
- The TL response remains almost unchanged after several cycles of irradiation, annealing and readout.

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ABSTRACT

Dy and Cu-doped SrF_2 nanoparticles (NPs) were synthesized by using co-precipitation method and their possible application to solid state dosimetry were studied and compared to that of pure SrF_2 NPs. X-ray diffraction (XRD), scanning electron microscopy (SEM) and energy dispersive spectrometer (EDS) were used for sample characterization. The highest thermoluminescence (TL) response of SrF_2:Dy and SrF_2:Cu NPs were found respectively at 0.5 and 0.7 mol% of Dy and Cu impurities. Seven overlapping glow peaks at 384, 406, 421, 449, 569, 495, 508 K and three component glow peaks at 381, 421 and 467 K were identified respectively for SrF_2:Dy and SrF_2:Cu NPs employing T_m–T_{stop} and computerized glow curve deconvolution (CGCD) methods. The TL sensitivity of SrF_2:Dy is approximately the same as that of LiF:Mg, Ti (TLD-100) cheeps. Linear dose response were observed for the SrF_2:Dy and SrF_2:Cu NPs up to the absorbed doses of 1 kGy and 10 kGy correspondingly. Regarding other dosimetry characteristics of the produced NPs such as fading, reproducibility and thermal treatment, Dy and Cu doped SrF_2 NPs recommend for high dose TL dosimetry applications.

1. Introduction

The micro-scale alkaline earth fluorides have been amongst the most popular TL dosimeters (McKeever, 1985; Chen, 1984, Vol. 1, Chapter 4). The luminescence of fluorides is connected mainly to the presence of rare earth ions in them (Radzhabov, 2002). Strontium fluoride (SrF2) is one of the most widely used alkaline earth materials because of its interesting luminescent, optical, and physical properties. It has a wide band gap, low phonon energy, low refraction index, high radiation resistance, and good mechanical strength (Ivanovskikh et al., 2005; Van der Ende et al., 2009). Optical properties and irradiation effects in micro-scale Nd^{3+}, Pr^{3+}, Tb^{3+} and Tm^{3+} doped SrF_2 crystals and their possible application to solid-state dosimetry have been studied and compared to those induced in pure SrF_2 crystals (Kristianpoller et al., 2004). However, the TL characteristics of SrF_2 under gamma irradiation have not adequately been studied, especially compared to other alkaline earth fluorides.

In recent years, nanomaterials because of their exceptional TL properties compared with their bulk equivalents, have attracted much attention (Zahedifar et al., 2011; Salah, 2011; Vij et al., 2010). Special optical properties of nano-scaled materials can be explained based on quantum size effect and large surface to volume ratio. Studies on different luminescent nanomaterials have revealed their excellent features such as simple TL glow curve structure and linear TL dose response over wider range of absorbed dose compared with those of corresponding microcrystalline samples. These behaviors have been explained using the track interaction model (Horowitz et al., 2001). Therefore, investigating

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the TL features of nanostructured counterparts of rare earth doped alkaline earth fluorides would be of importance for their potential application to estimate high doses of ionizing radiations, where the conventional microcrystalline phosphors have a limitation.

In this work, the synthesis procedure, TL kinetic parameters and dosimetry features of SrF_2:Dy and SrF_2:Cu NPs produced by using co-precipitation method are presented and discussed.

2. Experimental procedure

The SrF_2:M (M=Dy, Cu) NPs were prepared from strontium nitrate ($Sr(NO3)_2$, 99.99% purity), ammonium fluoride (NH_4F, 99.99% purity), Brij 35 (99.9% purity), dysprosium nitrate ($Dy(NO_3)_3$ of 99.99% purity) and copper nitrate ($Cu(No_3)_2$ of 99.99% purity), distilled de-ionized water and acetone (Merck Chemical). Firstly, 20 cc solution A and 20 cc solution B were prepared respectively by dissolving 0.500 gr strontium nitrate and 0.500 gr Brij in a mixture of aceton and deionized water. The 20 cc ammonium fluoride solution was prepared by dissolving 0.175 gr NH_4F in a mixture of acetone/water (solution C). Equal volumes of aceton and deionized water were used in the above solutions. The Solution B was added slowly to solution A while was placed on a stirrer and different amounts of dysprosium nitrate (copper nitrate) was added to it and finally solution C was added slowly while stirred. Lastly, the precipitate was centrifuged, washed with the mixture of acetone/water for several times. The final SrF_2:Dy (SrF2:Cu) NPs were obtained following drying the product in an oven at 90 °C and atmosphere pressure for 3 h.

3. Characterization

Powder X-ray diffraction patterns of the samples were recorded using an X-ray diffractometer (Bruker D8 Advance) with Cu K_α radiation (λ=1.54 Å) under the conditions of 40 kV and 30 mA, at a step size of 2θ=0.02°. SEM images were obtained using a scanning electron microscope model Philips XL-30 ESEM equipped with energy dispersive spectrometer (EDS).

The powder samples were placed in a Teflon container and the electron equilibrium condition was fulfilled in irradiation process. Dose of ionizing radiation were administrated to the powder samples with a ^{60}Co gamma source at the secondary standard dosimetry laboratory (SSDL) facilities, Karaj, Iran. The TL readouts were taken in a Harshaw model 4500 computer-based TL reader using a contact heating where the temperature of the heater strip (planchet) is recorded as indicator of the temperature of the sample with a precision of 1 °C. All TL readouts (apart from readouts in studying the fading characteristic), were carried out 3 hours after irradiation to allow tunneling and transitions between trapping states. The heating rate for readout was 1 °C/s (with preheat of 50 °C) to a maximum temperature of 350 °C. The samples were annealed at 400 °C for 15 min using a programmable oven with temperature precision of 1 °C and then were cooled rapidly to room temperature (75 °C/min). All the experiments were carried out using the samples produced in a single synthesis procedure. The TL responses of the samples of different batches under the same irradiation conditions were the same through an uncertainty of 5%. Since the TL response depends on the mass of the sample, it was kept constant at 0.005 gr using Sartorius Research. R 160 P weigher (with a mass accuracy of ±0.01 mg).

4. Results and discussion

Fig. 1(a–c) shows the X-ray diffraction patterns of the synthesized SrF_2:Dy (a), SrF_2:Cu (b) and pure SrF_2 (c) NPs. Except for very low differences in the peak intensities due to different impurities, the patterns are the same which confirm that the sample is SrF_2 with cubic structure and corresponds to JCPDS card no. 86-2418. From XRD results, the average particle size was calculated by Scherrer's formula from which the crystalline size of approximately 21 nm obtained using the dominant (111) peak. This result is in agreement with the SEM image of Fig. 2. Fig. 2(a and b) shows respectively the SEM images of SrF_2:Dy and SrF_2:Cu NPs. As is evident in this figure, the NPs have approximately the same size and spherical shape. EDS spectrum is observed in Fig. 3(a and b) which reveal that the products are SrF_2: Dy (a) and SrF_2: Cu (b) without contamination. The elemental concentrations are shown in the insets of Fig.3.

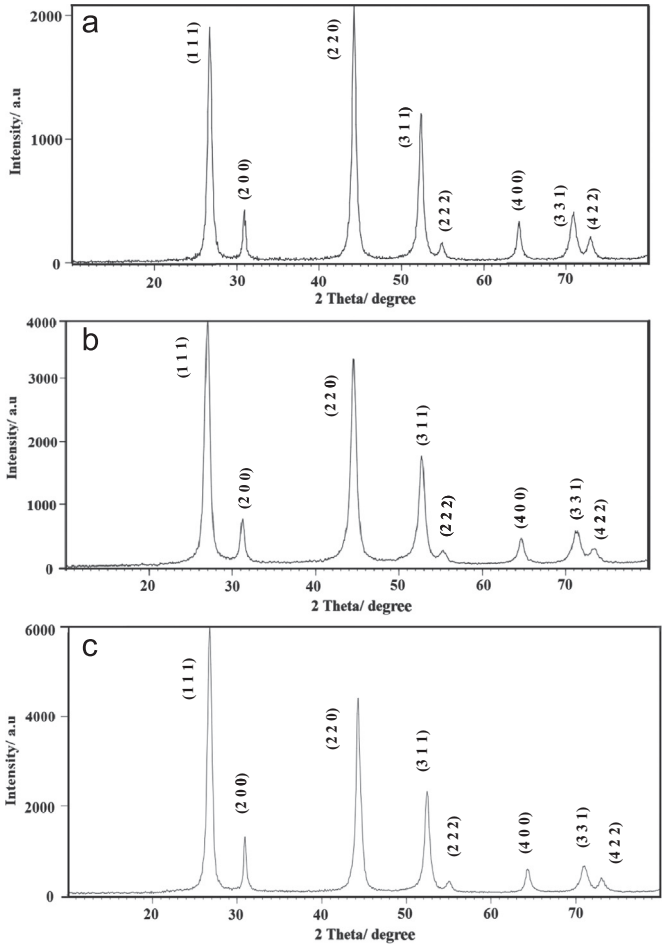

Fig. 1. (a–c). X-ray diffraction pattern of the synthesized SrF_2:Dy (a), SrF_2:Cu (b) and pure SrF_2 (c) NPs. Except for very low differences in peak intensities, the XRD patterns are the same, indicating that the host crystal is SrF_2.

The $T_m - T_{stop}$ method was employed to identify the number of component glow peaks contained in the complex glow curve of the synthesized NPs. Using this method, firstly a pre-readout anneal up to T_{stop}, in the temperature range of 90–220 °C was applied to bleach the low temperature portion of the glow curve, followed by cooling the sample to room temperature and readout for recording the TL glow curve (Zahedifar et al., 2013). Fig. 4(a and b) shows respectively the variation of T_m versus T_{stop} for the glow curves of SrF_2:Dy and SrF_2:Cu NPs subsequent to administrated dose of 100 Gy by ^{60}Co gamma source. All the samples were first annealed to a specific temperature, T_{stop} with a constant heating rate of 5 °C/s, then cooled rapidly to room temperature in nitrogen atmosphere followed by readout with a heating rate of 2 °C/s for