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Blue and infra-red stimulated luminescence in Cu^+ doped fused quartz for application in radiation dosimetry

Rujuta Barve [a], R.R. Patil [a,*], N.S. Rawat [b], N.P. Gaikwad [b], Ratna Pradeep [b], B.C. Bhatt [b], S.V. Moharil [c], M.S. Kulkarni [b]

[a] Institute of Science, R.T. Road, Civil Lines, Nagpur, India
[b] Radiation Safety Systems Division, Bhabha Atomic Research Centre, Mumbai, India
[c] R.T.M. Nagpur University, Nagpur, India

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ABSTRACT

Cu^+ was doped in fused quartz using simple technique and the BSL/IRSL and TL studies were carried out. The Cu^+ doped quartz shows enhanced TL as well as OSL sensitivity compared to the undoped material. The OSL sensitivity with blue stimulation, in particular, is relatively high, about 13% of the commercially used OSL phosphor (Al_2O_3:C-Landauer Inc.) on the basis of initial OSL intensity measurements (i.e. luminescence averaged over first 3 s). Earlier works show that OSL/TL signals in natural quartz is stable and thus can be used in archeological dating and other dosimetric applications. The aim of the present study is to investigate TL/OSL characteristics of Cu^+ doped fused quartz.

1. Introduction

Optically stimulated luminescence (OSL) is a relatively new technique in dosimetry of ionizing radiations. In OSL the defects are stimulated by the light in the visible/IR region and as a result, release of either the electron or hole and subsequent capture at the recombination centre leads to emission of radiation which is generally at a shorter wavelength compared to the wavelength of the stimulating radiation. The general requirement for material to be a good OSL phosphor is that the emission should be between 350 and 425 nm and the defects should have strong photo-ionization cross-section in blue green-region (450–550 nm) or IR region (650–800 nm). This limit on wavelength is due to availability of suitable filters, stimulation sources as well as sensitive PM tubes in this range and most importantly the requirement of separation of stimulating wavelength from the emission wavelength which ensures better signal to noise ratio. OSL was first used in archeological dating [1] and later proposed for personnel monitoring and environmental assessment of radiation with the development of Al_2O_3:C [2]. More recently, OSL of cerium, europium, samarium and copper ions doped into borate and silicate glasses has been reported [3–7]. Attempts are also made to develop materials like BeO [8], $NaMgF_3$ [9], MgO:Tb [10], $LiMgPO_4$:Tb,B [11] but, except for BeO and $LiMgPO_4$:Tb,B they still remain in development stage as far as their potential use in routine radiation dosimetry is concerned. OSL has been observed previously in natural and synthetic quartz materials [12,13] and has been used as a technique for archeological dating [14] with natural quartz and feldspars as the basic materials. Attempts were also made to diffuse impurities like Cu, Eu in quartz and feldspars in order to study their role as recombination centers in natural and synthetic quartz. It was reported that the OSL stimulation spectra of Cu-doped synthetic and natural quartz are similar and differ from those for alkali feldspars [15]. In the reported works the impurities are doped in quartz based materials via thermo diffusion at 1100 °C for Cu^+, and at 1700 °C for Eu^{2+}. The fused quartz developed by Justus et al. [16], shows prominent emission of Cu^+ around 550 nm with shoulder around 400 nm. Therefore, the sample is only suitable for infrared stimulated luminescence (IRSL) and not for blue stimulated luminescence (BSL). We have succeeded in preparing Cu^+ doped fused quartz at relatively low temperature ~800 °C which shows Cu^+ emission around 395 nm. Thus it is suitable for BSL as well as IRSL. Synthesis and TL/OSL properties of this phosphor are described in this paper.

2. Experimental

Fused quartz plates of 5–7 mm^2 were cleaned with hydrochloric acid and subsequently washed several times with distilled water. These plates were then immersed in a suspension of CuF_2 in water. The amount of CuF_2 in suspension was taken as

* Corresponding author. Tel.: +91 09890359291; fax: +91 07122565581.
E-mail address: rvapatil@yahoo.com (R.R. Patil).

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50 mol% with respect to quartz. Such high amount is necessary because only a fraction of Cu diffuses in the plates. The water was slowly evaporated so as to get all the CuF_2 particles deposited on the quartz plates. These plates were heated in tubular furnace at 800 °C for three hours and then cooled to room temperature by switching off the power to the furnace. The undoped plates were also given identical heat treatment. This is done to differentiate between the sensitization due to thermal treatment and due to doping. These plates were washed again with hydrochloric acid and then with water to remove the unreacted CuF_2 on the surface of the plates. The washed plates were dried and ground using pestle and mortar and then sieved to the 90–210 μm particle size. To see the effect of diffusion temperature on the Cu^+ emission some of the samples were also prepared at 900 °C.

The TL readouts were taken on the BARC developed TL reader system. The BSL measurements were carried out using blue/green LED based measurement setup described elsewhere [17]. The IRSL measurements were carried out using a Riso reader having an array of IR LEDs emitting at 880 nm and a band-pass green filter. All TL and OSL measurements were carried out 10 min after the irradiation of the samples. All the samples were irradiated with $^{90}Sr/^{90}Y$ beta source with dose of 0.5 Gy. Sensitivity comparison between Cu^+ doped fused quartz and Al_2O_3:C has been done using OSL technique by normalizing their signals for the same weight and dose. For this, the unirradiated samples were bleached with blue (470 nm) light at 50 mW/cm^2 for 5 min in order to completely erase residual signal, if any.

The photoluminescence measurements were carried out on Hitachi F-4000 spectrofluorometer.

3. Results and discussion

Fig. 1 shows the photoluminescence spectra of Cu^+ doped samples. The emission band is observed at 395 nm (Fig. 1a) with the excitation at 247 nm (Fig. 1d). The undoped quartz shows very weak and unstructured PL emission hence it can be concluded that the emission band is due to intraconfigurational transition of Cu^+. Normally the emission from copper (Cu^+) comes from $3d^94s^1 \rightarrow 3d^{10}$ transition. The single emission band indicates that the lattice is crystalline quartz and not glassy as multiple Cu bands are observed in SiO_2 based glasses due to multiple sites of varying symmetry [18]. It has been observed earlier that natural quartz doped with Cu using electro diffusion technique shows emission around 360 nm with a shoulder around 550 nm whereas the synthetic quartz doped with thermo diffusion at 1700 °C shows prominent emission around 515 nm with shoulder around 395 nm. The reason cited for this change is the conversion from crystalline to glassy phase [15]. The photoluminescence of Cu^+ in fused quartz

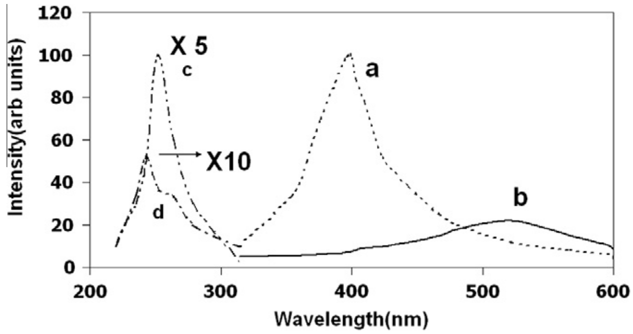

Fig. 1. Photoluminescence of Cu^+ doped quartz: (a) emission of Cu^+ doped quartz prepared at 800 °C; (b) emission of Cu^+ doped quartz prepared at 900 °C; (c) excitation for Cu^+ emission prepared at 900 °C and (d) excitation for Cu^+ emission prepared at 800 °C.

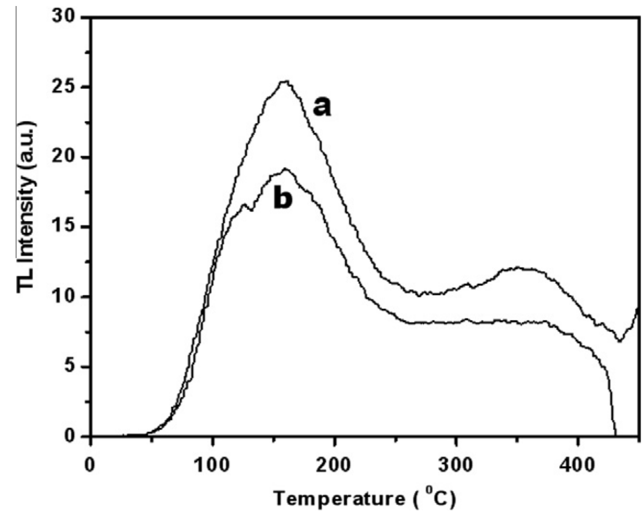

Fig. 2. Thermoluminescence pattern of (a) Cu^+ doped quartz and (b) undoped quartz.

is reported earlier at 550 nm. However the diffusion was carried out at 1100 °C [16]. As it is well known that quartz goes structural changes with temperature and therefore temperature of diffusion is very important factor. When the diffusion of Cu^+ was carried at 900 °C, we observed the changes in emission band. The emission is observed at 520 nm (Fig. 1b) with the excitation shifting to 254 nm (Fig. 1c).

The TL/OSL intensities of doped samples were found to be enhanced. Fig. 2 shows the TL patterns for doped and undoped quartz samples. For the doped and undoped sample two TL peaks, one around 160 °C and other broad peak around 350 °C are observed (Fig. 2a and b). However the doped sample shows the enhancement in intensity compared to the TL intensity of the undoped sample. The prominent peak of doped sample is about 1.5 times more intense than that of the undoped sample. As both the doped and undoped samples were subjected to similar heat treatment the increased sensitivity due to thermal sensitization will be same for both samples and hence the observed increase in sensitivity in the doped sample is due to presence of Cu^+ ions. The TL peak pattern of sample in which Cu^+ is diffused at 900 °C is similar to the sample

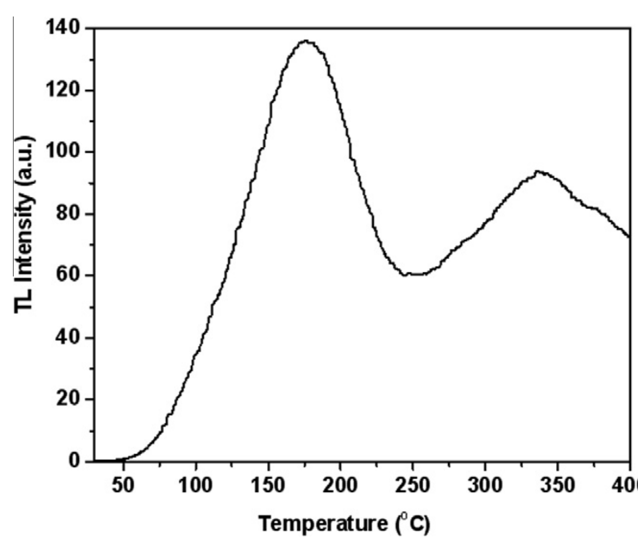

Fig. 3. Thermoluminescence pattern of Cu^+ doped quartz treated at 900 °C.