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Optically stimulated luminescence in doped NaF

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HIGHLIGHTS

- NaF:Mg,Cu,P shows good OSL properties.
- The OSL sensitivity is 37% to that of commercially available Al_2O_3:C.
- The sample possesses good linearity in the dose range 10 mGy to 1 Gy.
- Negligible fading is observed during first 10 days.

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ABSTRACT

OSL in doped NaF is studied. Study shows that NaF:Mg,Cu,P phosphor possess good OSL properties having sensitivity comparable to that of commercially available Al_2O_3:C (Landauer Inc.). For the luminescence averaged over 3 s the obtained OSL is 37% of that commercial available Al_2O_3:C. Of the several phosphors investigated, phosphor with impurities concentration Mg(0.01 mol%), Cu(0.2 mol%), P(1 mol%) shows good OSL sensitivity good linearity in the 10 mGy to 1 Gy dose range and negligible fading. This sample shows a intense single TL peak around 350 °C which gets depleted by 14% after the OSL readout. This imply that maximum OSL is coming from deep traps giving stability to the signal. The ease of preparation along with other good OSL properties will make this phosphor suitable for radiation dosimetry applications using OSL.

1. Introduction

With the development of Al_2O_3:C as a highly sensitive TL and OSL phosphor (McKeever et al., 1996; Akselrod et al., 1990) great interest has been developed in the dosimetry using OSL (optically stimulated luminescence). This development has also led to wide acceptance of OSL technique in radiation dosimetry. In OSL the electrons/holes, created due to ionizing radiation and trapped in the defects, are stimulated by the light in the visible/IR region. The release of either the electron or hole, and subsequent capture at the recombination centre leads to emission of light. The emitted light is generally at a shorter wavelength compared to the wavelength of the stimulating radiation (Akselrod et al., 2007; Bøtter-Jensen et al., 2003). The general requirement for material to be a good OSL phosphor is that the emission should be in between 350 and 425 nm and the defects should have high photo-ionization cross-section in blue-green region (450–550 nm) or IR region (650–800 nm). This limit on these ranges of wavelength is due to the 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 (Bøtter-Jensen et al., 2003). The basic process in OSL and TL is similar except the mode of stimulation. In OSL the charge carriers are evicted from the traps using light as a mode of stimulation which otherwise are evicted by thermal energy in the TL readout. OSL based dosimeters are being used increasingly in radiation dosimetry due to various advantages it has over TL based dosimeters such as faster and multiple readouts, absence (no role) of thermal quenching, high sensitivity, possible use of phosphor in plastic binders. Though Al_2O_3:C is best known OSL phosphor because of its excellent OSL properties (McKeever et al., 1996; Akselrod and McKeever, 1999) several efforts have been made to prepare other OSL materials, like KBr:Eu (Douguchi et al., 1999), $(NH_4)_2SiF_6$:Tl (Masson et al., 2004), MgO:Tb (Bos et al., 2006),

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$NaMgF_3$: Eu^{2+} (Dotzler et al., 2007), $LiAlO_2$:Tb/Ce (Dhabekar et al., 2008; Mittani et al., 2008), $ZnAl_2O_4$:Tb (Menon et al., 2008), $Y_3Al_5O_{12}$:C (Kulkarni et al., 2008), $MgAl_2O_4$:Tb (Raja et al., 2009), BeO (Sommer et al., 2007), $LiMgPO_4$: Tb,B (Dhabekar et al., 2011). Recently Cu, P doped sodium fluorosilicate (Barve et al., 2014) and Europium doped $CaSO_4$ (Kulkarni et al., 2014) have been reported.

Optical sensitivity in alkali halides is well known and is reported and studied extensively. The optical sensitivity in alkali halides arises due to absorption band of F or M centres in the visible region. In the coloured NaF the absorption bands with peak positions at 341 and 505 nm have been identified due to F and M centres, respectively (Gupta and Rao, 1980). The absorption band for M centre is broad and around 505 nm and can be easily bleached when exposed to light in blue/green region. Since the reported Cu^+ emission in NaF is below 420 nm and absorption wavelength of M centres lies in blue green region it satisfies both general requirements for material to be a good OSL phosphor. Thus it is expected that NaF doped with Cu will be a good candidate as a OSL phosphor. Additional dopants Mg, P were added along with Cu to increase the trapping efficiency in NaF as this combination of impurities imparts very high trapping efficiency increasing the sensitivity in LiF:Mg,Cu,P phosphor.

In this work Photoluminescence (PL), Thermoluminescence (TL), and Optically stimulated luminescence in doped NaF is presented.

2. Experimental

All the NaF based phosphors were synthesized from freshly obtained NaF prepared by reacting A.R grade Sodium Carbonate with Hydro-Fluoric acid(HF) in slightly excess amount. The NaF slurry thus formed in the reaction was dried at room temperature for several days to get the dried NaF powder.

For preparing Cu doped NaF, desired amount of NaF powder was sprinkled with $CuCl_2$ solution in desired amount. The wet powder was then dried for several hours under the drying lamp. This dried powder was heated for one hour in reactive atmosphere of NH_4Cl at 500 °C. This treated powder was then melted in graphite crucible and quenched quickly by transferring the melt into a flat graphite crucible. The solidified lump was then crushed to obtain the Cu doped powder of NaF. Several Cu doped samples with varying concentration of Cu (0.02–0.2 mol%) were prepared. Magnesium co-doped samples with varying concentration (0–0.2 mol%) were also prepared in similar way. For doping Mg, Magnesium Chloride solution in desired amount was sprinkled on the NaF powder along with $CuCl_2$ solution. Some phosphorous co-doped samples were also prepared. Doping of phosphorus was carried out in different way. For this, desired amount of phosphorous salt $NH_4H_2PO_4$ was added to a graphite crucible. The Cu doped or Cu,Mg doped NaF was then added to this graphite crucible. The mixture was then melted at 1000 °C to obtain Cu,P or Mg,Cu,P doped NaF. Several samples with varying concentration of P (0–2 mol%) were prepared. The various samples prepared with varying concentration of impurities are summarized in Table 1.

Photoluminescence studies were carried on Hitachi F-4000 Spectrofluorometer with the excitation slit 1.5 nm and the emission slit 5 nm.

For studying the TL and OSL response, all the samples were irradiated using $^{90}Sr/^{90}Y$ beta source with the dose rate of 20 mGy per min. The samples were given a test dose of 100 mGy. The CW-OSL response and thermoluminescence of the samples is recorded on the assembly described elsewhere (Kulkarni et al., 2007). The assembly consist of array of blue/green LEDs as a stimulation source with adjustable power from 11 mW/cm^2 to 48 mW/cm^2. Two optical filters viz. UG-1 (across PMT), to prevent stimulation signal from reaching PMT (9111B, 25 mm diameter end window PMT) and GG-435 (across LEDs), to cut-off the stimulation wavelengths below 435 nm, were used in the assembly. All the operations in the assembly are controlled by the suitable software.

During the OSL measurements the LED power was kept at 11 mW/cm^2 and signal was recorded for 90 s with the acquisition time 0.1 s. All the thermoluminescence measurements were taken at the 4 °C/s. heating rate.

3. Results and discussion

3.1. Photoluminescence

Fig. 1 shows photoluminescence spectra of various Cu doped NaF samples. In Cu (0.02 mol%) doped sample double humped emission is observed around 360 nm and 375 nm (Fig. 1c). The excitation to this band is observed around 254 nm. (Fig. 1e). This is similar to what is reported in our earlier work (Patil and Moharil, 1995). Mg(0.2 mol%) co-doped sample shows similar emission with increase in emission intensity (Fig. 1d). No change in excitation is observed and is similar to only Cu doped sample (Fig. 1e). When sample is co doped with P(2 mol%) along with Mg, peak structure remain same but decrease in intensity is observed (Fig. 1a). The excitation wavelength is observed at 261 nm (Fig. 1b). This emission is attributed to transitions arising from $3d^9 4s^1 \rightarrow 3d^{10}$ transitions of Cu^+ ions (Payne et al., 1983).

3.2. TL-OSL response

Fig. 2 shows TL glow curves of various doped NaF samples. In general in all the samples the TL glow curve comprises of two TL peaks, one around 85 °C and other around 135 °C. One high temperature peak around 350 °C is also observed. This is in agreement what is observed earlier in pure NaF (Moharil and Deshmmukh, 1976) For only Cu(0.02 mol%) doped sample (Fig. 2a) the peak around 85 °C is prominent compared to 135 °C peak. Doping Mg (0.2 mol%) along with Cu (0.02 mol%) increases the TL intensity by 15% compared to only Cu doped sample (Fig. 2b). A less intense new high temperature peak around 350 °C is also observed. When P (2 mol%) is doped and keeping other two impurity concentration same similar peak structure with 30% increase in the intensity of the low temperature peak compared to only Cu doped sample is observed (Fig. 2c).

Fig. 3 shows BSL(blue stimulated luminescence) from various doped NaF samples. Intense BSL(blue stimulated luminescence) from various NaF samples doped with Cu; Cu,Mg; Mg,Cu,P impurities. Inset shows CW-OSL normalized curves along with normalized curve of Al_2O_3:C (Landauer Inc.). In all the samples the observed decay is fast compared to Al_2O_3:C and whole signal decays within 30 s. The OSL and TL data is summarized in Table 1. For only Cu doped sample the OSL sensitivity is minimum whereas maximum OSL sensitivity is observed for the sample doped with Mg (0.2 mol%), Cu (0.02%), P(2 mol%) (Fig. 3c). The OSL from this sample is compared with Al_2O_3:C Landauer and is 2.1 times more when compared by averaging counts over initial 3 s by using method suggested by Yukihara et al. (2004). It is observed that the OSL intensity is well correlated to the observed TL intensity in these samples. Thus samples which show high TL intensity also shows intense OSL.

Fig. 4 shows the TL before (Fig. 4a) and after BSL (Fig. 4b) for the NaF: Mg (0.2 mol%),Cu (0.02%),P(2 mol%) sample. It is observed that both the peaks, peak around 85 °C and 135 °C vanish completely, correlating these peaks to the observed OSL. Fig. 3(d) also shows the OSL after removing the peaks around 85 and 135 °C thermally. It is observed that the sample show 27% OSL even after