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# Exploring stable thermoluminescence signal in natural Barite ($BaSO_4$) for retrospective dosimetry

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## HIGHLIGHTS

- Glow peak at 608 K in Mn doped Barite.
- Minimum detectable dose of 1.45 ± 0.12 mGy with 35% fading in 30 days storage time.
- Reproducible TL signal within ± 5%.
- Suitable for retrospective dosimetry and geochronology of young deposits extending to 20 ka.

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## ABSTRACT

We explore the possible use of Barite ($BaSO_4$) for radiation dosimetry and geochronology using thermoluminescence technique. Natural Barite with Mn as an impurity has a glow peak at 608 K with a minimum detectable dose of 1.45 ± 0.12 mGy. This peak shows ~35% fading on 30 days of storage time and is photo-bleachable with excellent reproducibility on repeated read-out. The sensitivity changes with dose and evidence of athermal fading is also seen. We infer that the signal can be used for both retrospective dosimetry and geochronology of young deposits extending to 20 ka.

## 1. Introduction

Use of natural Barite as a radiation dosimeter was examined in detail in view of its multifaceted use in X-ray storage properties, medical diagnostics, drilling activities (Shen et al., 2007; Lee et al., 2003) and due to its occurrence in geological strata that provides a new possibility for geochronology (Iwata et al., 1993; Shinsho et al., 2011; Ramaswamy et al., 2010; Bernstein and Byrne, 2004). Barite crystallizes in diverse depositional environments ranging from hydrothermal, evaporatic or biogenic.

## 2. Barite luminescence: present status

Considerable studies have been carried out on Barite doped with either transition-metal or rare-earth ions. These include, measurement of spatial distribution of radiation dose around a target using $BaSO_4$: Eu sheet (Ramaswamy et al., 2010) and environmental dosimetry using samples from shielding blocks of Barite from CERN (Kiyak et al., 2010; Kitis et al., 2010). Other applications are the use of $BaSO_4/Y_2O_3$: $Eu^{3+}$ core–shell microspheres for lighting and plasma display (Zhang et al., 2009) and the use of $BaSO_4$: Eu nanoparticles (Salah et al., 2009), $BaSO_4$: Cu (Manam and Das, 2009) and $BaSO_4$: Mn (Manam and Das, 2010) for radiation dosimetry. Sato et al. (2011) explored Barite for luminescence–ESR dating for detection of cosmic ray exposure and estimated the thermal stabilities and decay kinetics of $SO_3^-$ center.

Manam and Das (2010) examined the trapping dynamics in Mn

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[1] Prof. P.S. Rao died during the course of the present work and we dedicate this work to him.

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doped synthetic $BaSO_4$ samples and reported glow peaks at 420 K and 498 K with trap depths of 0.59 ± 0.006 eV and 0.63 ± 0.005 eV, respectively. In the present work, we revisit these results due to our concerns on, (a) the activation energy values of 0.63 eV for 498 K peak being too low, given that the materials with 0.6–0.8 eV trap depth should exhibit long lasting phosphorescence (LLP) (Bessière et al., 2014), which is not the case; (b) Mn doped sample had a long decay tail above 573 K and was completely ignored (see Fig. 4 in Manam and Das, 2010); (c) trap distribution with glow curve temperature was not explored; and (d) reproducibility after repeated measurements and impact of high doses on luminescence sensitivity remain unsolved.

We establish a stable TL signal in $BaSO_4$:Mn (white colored) with glow curve maximum at 608 K possessing lifetime of the order of $10^{11}$ s as compared to $10^7$ s for 498 K glow peak as reported in Manam and Das (2010). We explore trap distribution and stability, variation of TL sensitivity under high doses, dose response; minimum detectable dose and athermal fading of this new 608 K TL signal.

## 3. Experimental and instrumentation

Two natural-Barite samples were used in their as-received state except that these were broken down; washed with alcohol and sieved to 90–150 μm size fractions. No thermal treatment/annealing were done on these samples. X-ray diffraction studies using a Philips PANalytical X'pert Pro-diffractometer employing Cu-Kα radiation ($\lambda = 1.54060$ Å) between Bragg-angles ($2\theta = 5$–$70°$) and EPR measurements in X-band using JEOL JES-TE100 ESR spectrometer with 100 kHz field modulation were carried out.

Thermoluminescence (TL) studies were carried out at a linear heating rate of 2 °C/s in an inert $N_2$ atmosphere using either of a Risoe TL-DA-15 or a Daybreak-2200 TL/OSL reader equipped with a $^{90}Sr$–$^{90}Y$ beta source providing dose rates 0.0533 Gy/s and 0.0167 Gy/s, respectively. The TL measurements were performed in temperature region 313–723 K. The detection channel comprised EMI 9235 QA (bi-alkali type) photomultiplier tube (PMT) with filter assemblies, Schott BG-39 + Hoya U-340 (340–372 nm UV transmission) or Schott BG-39 + Cs 7–59 (384–445 nm blue transmission).

## 4. Results and discussion

X-ray diffraction studies and the electron paramagnetic resonance (EPR) studies showed that the samples were phase pure. A significant difference between two Barite samples was their Mn content. Mn was undetectable in Barite 1 and had a concentration of $0.7 \times 10^{18}$ spin/cm$^3$ in this Barite 2, as measured by electron paramagnetic resonance (EPR) technique. Mn was incorporated into the lattice with +2 valence states. We designated these samples as Barite 1 (without Mn) and Barite 2 (with Mn). Presence of quartz or any other major phases of natural minerals could not be detected in XRD scans.

### 4.1. Glow curves

The glow curves of the Mn containing samples as received (natural TL or NTL) and for NTL + beta doses are presented in Fig. 1. Glow curves possess maxima at 371 K, 508 K and 608 K. Natural sample without laboratory dose exhibit maximum at 608 K and this peak was explored further.

Fig. 2 shows the glow curve in two different emission windows for Barite 1 and Barite 2 samples. The results enabled following observations.

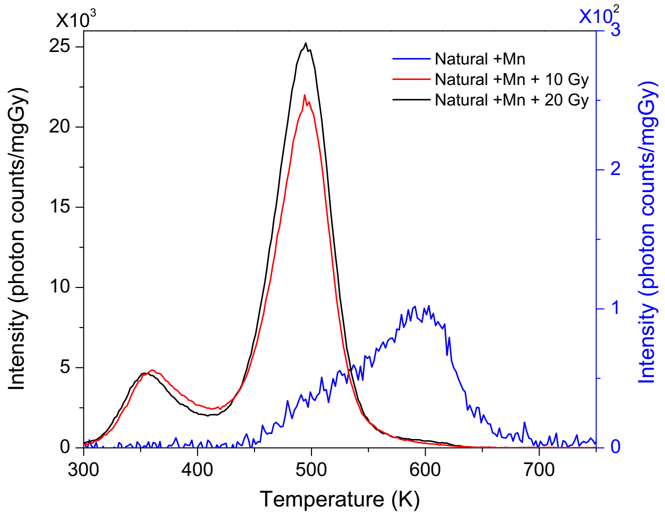

**Fig. 1.** TL glow curve for natural and natural + irradiated sample.

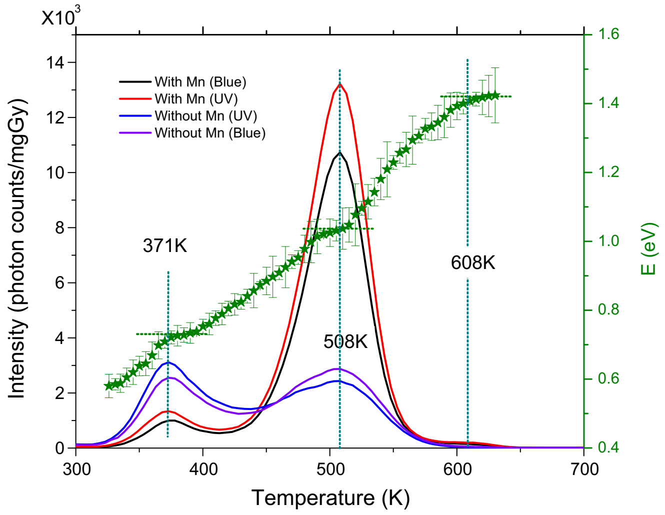

**Fig. 2.** TSL glow curves of barite (without and with Mn) under 5 Gy β-dose using different filter combinations. $E$–$T_{stop}$ is plotted on right scale (in green) showing three different regions and trap distribution. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

a. Barite 1 sample gave glow peaks at 371 K, 475 K and 508 K and Barite 2 had glow peaks at 371 k, 508 K and 608 K.
b. TL intensity in Barite 1 was highest at 371 K (in UV-emission window) and at 508 K (in blue-emission window). For Barite 2, the TL intensity was always highest at 508 K (both UV and blue-emission windows).
c. For 371 K glow peak, TL intensity was ~1.5 times higher in Barite 2 and ~1.3 times higher for Barite 1 in UV-emission window when compared with blue-emission window.
d. For 508 K glow peak, the TL intensity was ~1.27 times higher in the UV-emission window when compared with blue-emission window in Barite 2, whereas, TL intensity decreased by ~16% in UV/blue-emission windows.
e. On comparison of results among Barite 1 and Barite 2 samples for 508 K glow peak, the TL intensity was ~7 times higher for Barite 2 in UV-emission window and ~3.6 times higher for Barite 1 in blue-emission window. This suggests that the presence of Mn enhances charge trapping and recombination pathways and the formation of deep traps are favored.