



Peculiarities of optically stimulated luminescence in halite



M. Biernacka^{a,*}, R. Majgier^a, K. Maternicki^a, M. Liang^b, A. Mandowski^a

^a Jan Dlugosz University, Institute of Physics, Ul. Armii Krajowej 13/15, 42-200 Częstochowa, Poland

^b Université du Maine, Avenue O. Messiaen, 72087 Le Mans, Cedex 9, France

HIGHLIGHTS

- Optically stimulated luminescence (OSL) of rock salt (halite) was studied.
- The measurements were performed using the variable delay OSL method (VD-OSL).
- It was found that the relative regeneration of OSL intensity in halite could be as high as 600–700%.
- Inverse fading was observed.
- The regeneration effect (RE) remains at a constant level in the long time scale.

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ABSTRACT

Sodium chloride – NaCl is one of materials indicating strong OSL signal after exposure on ionizing radiation. Previous studies of the OSL response in pure sodium chloride showed coexistence of fading and regeneration of the signal using the newly developed *variable delay optically stimulated luminescence* technique (VD-OSL). This paper presents investigations of some peculiarities of long time scale OSL properties of rock salt (halite) including sensitization phenomena for various bleaching methods. Dose response characteristics were studied in the range from 200 mGy to 1 Gy for series of halite aliquots without signal bleaching and for single aliquot using zeroing by blue and green light.

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1. Introduction

Halite (rock salt) is a mineral occurring in nature, which has interesting ability to store information on the absorbed dose of ionizing radiation. Irradiated salt crystals exhibit pronounced thermoluminescence (TL) and optically stimulated luminescence (OSL). Hence, in principle, it can be used in retrospective dosimetry as well as a luminescence dating material (Zhang et al., 2005; Bernhardsson et al., 2009; Spooner et al., 2011; Christiansson et al., 2012; Spooner et al., 2012; Hunter and Spooner, 2012; Christiansson et al., 2014). At this time, the exact mechanism of the OSL is not well explained even for the chemically pure sodium chloride (NaCl), which exhibits surprisingly strong luminescence. It was suggested that OSL and TL can be attributed to simultaneous localized and delocalized transitions (Biernacka and Mandowski,

2013; Mandowski and Biernacka, 2014). For the rock salt, which is a natural composite with a complex defect structure, the nature of the radiation induced luminescence phenomena (OSL and TL) is much more difficult to explain.

Interaction of high-energetic radiation with ions in an alkali-halide crystal can lead to a variety of lattice defects. Radiation induced F centers are generated together with so called H centers in geminate pairs. The H center can be regarded as a halogen interstitial defect, where the F center can be considered as a halogen vacancy. Recombination of charge carriers between F and H centers is connected with emission wavelength of 425 nm (McKeever, 1985; Sunta, 2015 and references therein). TL measurement for pure NaCl (Halperin et al., 1959) confirm this data. It has been well established that divalent impurities, such as Mn²⁺, Zn²⁺, Cd²⁺, act as electron traps, while the Mg²⁺ cation vacancies complexes do not act as electron-trapping centers in NaCl crystals (Egranov and Nepomnyachikh, 1988).

Presented results relate to some peculiarities of long time scale

* Corresponding author.

E-mail address: m.biernacka@ajd.czyst.pl (M. Biernacka).

OSL properties of halite including sensitization phenomena for various bleaching (removing of residual OSL signal between readouts) methods. The measurements were performed for different wavelength of bleaching and various sample storage time as well as using newly developed *variable delay optically stimulated luminescence* technique (VD-OSL). Dose response characteristics were studied in the range from 200 mGy to 1 Gy for series of halite aliquots without signal bleaching and for single aliquot using zeroing by blue and green light.

2. Experiment and results

2.1. Samples and instrumentation

Natural colorless rock salt (halite) was used. Aliquots were prepared by cutting and polishing pieces of crystals having similar masses: 25 ± 1 mg, thickness about 1 mm and surface area 9–16 mm².

As a reference material for the analysis pure sodium chloride crystals were used. NaCl crystals were prepared from analytical quality sodium chloride material, which was dissolved in distillate water and re-crystallized (with presence of daylight). The single-grain crystals were colorless with white inclusions and grain size of 2–5 mm.

The equipment used for measurements was custom-made OSL reader 'HELIOS-1' (Mandowski et al., 2010). The reader consists of interchangeable modules for stimulation in the form of five LEDs with optical lenses and maximum power of 5 W per each diode. Detection of luminescence is made by a modified integrated photon counting module H7360 (Hamamatsu) with quartz window, counter electronics and computer interface. Additional photosensor is applied in order to control the LEDs emission. Irradiations were made using ⁹⁰Sr/⁹⁰Y β source with activity of 37 MBq.

In this series of measurements we used for stimulation green LEDs (peak at 520–532 nm) with cut-off filters GG495 and OG515. OSL UV light emission was detected using Schott UG11 filters (transmission range 300–380 nm).

2.2. Dose–response characteristic of halite mineral

Dose response characteristics were studied in the range from 200 mGy to 1 Gy for series of halite aliquots (where signal resetting was unnecessary) as well as for a single halite crystal using various method of optical bleaching with blue and green light. In the case of single sample, specific dose was applied and then *continuous wave* (CW-OSL) readout was carried out. Subsequent measurements were preceded by the removal of the residual signal using optical zeroing. The results are shown in Figs. 1, 2a and b. The dependencies show linearity in the studied range of doses. This is similar as in the case of other alkali halides (e.g. Popov and Plavina, 1995).

2.3. Investigations of the regeneration effect using VD-OSL method for blue and green light of bleaching

Long time scale luminescence phenomena may be well studied using VD-OSL method (Biernacka and Mandowski, 2013). This method consists of the following steps:

1. Sample preparation – optical (blue or green light) bleaching.
2. Excitation of the sample by ionizing radiation (defined dose).
3. Repeated CW-OSL measurements for a fixed time delay between readouts.
4. Repeating steps 1–3 for the next time delay (e.g. time delay = 10 s, 20 s, 40 s, etc.).

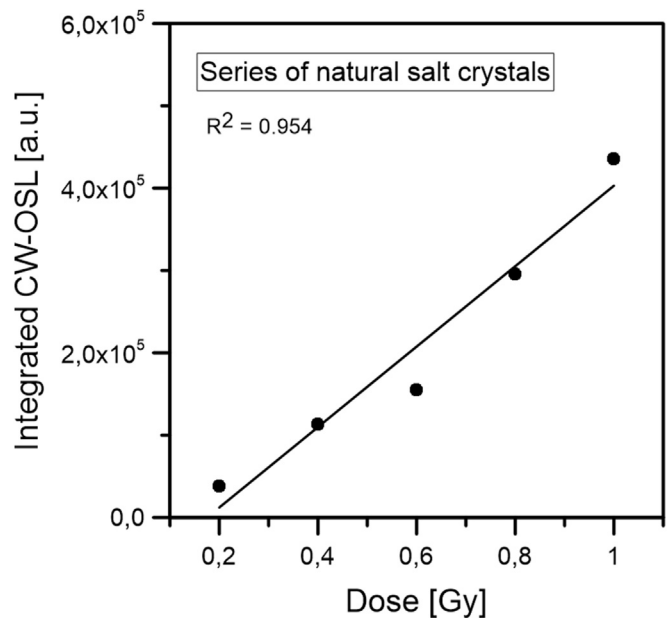


Fig. 1. Dose response characteristics for series of halite samples. Each point represents the average of 3 CW-OSL readouts for various samples with similar masses about 25 mg. CW-OSL readout parameters: counting time 60 s, storage time 120 s. CW-OSL decay integrated in the range from 0 s to 60 s.

VD-OSL results are presented in Fig. 3. The relative OSL regeneration enhancement may be defined as follows (Mandowski and Biernacka, 2014):

$$\Delta = \frac{\mathcal{I}_{\text{start}}^{(n+1)} - \mathcal{I}_{\text{end}}^{(n)}}{\mathcal{I}_{\text{end}}^{(n)}} \quad (1)$$

where $\mathcal{I}_{\text{end}}^{(n)}$ and $\mathcal{I}_{\text{start}}^{(n+1)}$ denote final and initial OSL intensities, for the n -th and $(n + 1)$ -th measurement in a series, respectively. The time delay is time interval between the first and second CW-OSL readout.

Normally, Δ parameter should always be negative or close to zero. Nevertheless, experimentally measured values for pure NaCl and halite were always positive, indicating that regeneration in this material really occurs. Maximal Δ values reached almost 735% for blue and 695% for green bleaching for time delays 20 495 s and 10 245 s, respectively. For comparison the maximum value of regeneration effect for pure NaCl was found in the similar times scale of the order of 10³ s however Δ values achieved only 190% (Mandowski and Biernacka, 2014) for identical measurement conditions. The discrepancy between regeneration characteristics for these two kinds of salts could be related to the some natural dopants present in halite samples. In general the regeneration effect does not depend on the wavelength of optical bleaching (Fig. 3).

2.4. Investigations of the sensitization for blue and green light of bleaching

The OSL measurement- and bleaching-induced sensitivity changes of halite were observed by the following procedure. After readout, the samples were irradiated with 189 mGy dose and then after short storage (120 s) OSL for 60 s was measured. After readout, the samples were bleached by green or blue light. This procedure was repeated several times (the cycle). The cycles were repeated after one, two and three weeks. The integrated OSL in the range from 0 s to 60 s and normalized to the first data point was plotted as

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