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Possibility of elastico-mechanoluminescence dosimetry using alkali halides and other crystals

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HIGHLIGHTS

- Elastico-mechanoluminescence (EML) can be used in radiation dosimetry.
- As EML occurs in the elastic region, the same sample can be used number of times.
- The EML can be used for X-ray, γ-ray and ultraviolet dosimetry.
- Mathematical approach verifies the possibility of EML dosimetry.

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ABSTRACT

The elastico-mechanoluminescence (EML) intensity of X or γ-irradiated alkali halide crystals can be used in radiation dosimetry. The EML intensity of X or γ-irradiated alkali halide crystals increases linearly with the strain of the crystals, and when the crosshead of the testing machine deforming an X or γ-irradiated crystal is stopped, then the EML intensity decreases with time. The semilog plot of the EML intensity versus $(t - t_c)$ (where t_c is the time where the crosshead of the testing machine is stopped) indicates that, in the post-deformation region, the EML intensity initially decreases exponentially at a fast rate and later on it decreases exponentially at a slow rate. The EML intensity increases linearly with the density of the F-centres in the crystals. This fact indicates that elastico-ML can suitably be used for the radiation dosimetry. The EML spectra of X or γ-irradiated alkali halide crystals are similar to their thermoluminescence spectra. Based on the detrapping of electrons during the mechanical interaction between the dislocation segments and F-centres, an expression is derived, which indicates that the EML intensity should increase linearly with the density of F-centres in the crystals. The expression derived for the decay of EML indicates that the decay time for the fast decrease of EML should gives the pinning time of dislocation segments (lifetime of interacting F-centres), and the decay time for the slow decrease of EML intensity should gives the lifetime of electrons in the shallow traps. As the elastic deformation is non-destructive phenomenon and the EML intensity depends on the radiation dosage given to the alkali halide crystals, similar to the thermoluminescence and photo-stimulated luminescence, the EML of alkali halide crystals and other crystals may be used for the radiation dosimetry. In EML dosimetry, the same crystal can be used number of times because the elastic deformation does not cause permanent deformation in the crystals, and moreover, comparatively the devices needed for the EML measurements are of low cost and very simple. In recent years, a large number of elastico mechanoluminescent materials have been investigated, and the study of their suitability for the radiation dosimetry may be interesting.

1. Introduction

When certain materials with a crystalline structure are exposed to ionizing radiation, luminescence can arise from either thermal

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stimulation, optical stimulation, and also by some other processes. The overall luminescence emission can take place in three steps as follows: (i) ionization due to exposure of the crystal to radiation, (ii) storage of radiation energy, and (iii) stimulation. The absorption of energy, e.g., from ionizing radiation, in a crystalline structure causes the creation of electron–hole pairs. If sufficient energy is provided to an electron to break from the valence band and overcome the band gap, this electron is moved to the conduction band, where it can move freely about the crystal. The hole in the valence band can also move freely about the crystal. The electron moving in the conduction band quickly loses its excitation energy and can either fall back to the valence band immediately and recombine with a hole or get trapped at defects within the crystalline structure. Some of the holes produced in the valence band may also get trapped at defects within the crystalline structure. A trapped electron remains so until it is provided with enough stimulation energy to overcome the trap and eventually recombine with a hole at a recombination centre. These recombinations can give rise to the emission of light, i.e. luminescence. In case of an emission during stimulation, this phenomenon is referred to as ‘optically stimulated luminescence’ or ‘thermoluminescence’ depending on whether the stimulation source is light or heat.

When certain irradiated solids are subjected to elastic deformation, then the detrapping of trapped electrons takes place either due to the tunnelling of trapped electrons to the dislocation band during the mechanical interaction between the filled traps and dislocations (Chandra, 1998) or due to the tunnelling of trapped electrons to the conduction band caused by the piezoelectric field produced due to the applied pressure (Chandra, 2011), Chandra and Chandra, 2012, Chandra et al., 2010a,b, 2013a,b). The luminescence induced by the elastic deformation of solids is called elastico-mechanoluminescence. In general, the light emission induced by any mechanical action on solids is known as mechanoluminescence (ML) and the light emissions induced by elastic deformation, plastic deformation and fracture of solids are called elastico ML (EML), plastico ML (PML), and fracto ML (FML), respectively (Chandra and Ramrakhiyani, 1992, 2011). Whereas nearly 50% of all inorganic salts and organic molecular solids show ML during their fracture, only a few solids exhibit ML during their elastic and plastic deformation. The examples of elastico mechanoluminescent materials are: X or γ-irradiated alkali halide crystals, ZnS:Mn, $SrAl_2O_4$:Eu, $SrAl_2O_4$:Ce, $SrAl_2O_4$:Ce,Ho, $SrAl_2O_4$:Er, $SrAl_2O_4$:Eu,Er, $SrMgAl_6O_{11}$:Eu, $SrCaMgSi_2O_7$:Eu, $SrBaMgSi_2O_7$:Eu, $Sr_2MgSi_2O_7$:Eu, $Ca_2MgSi_2O_7$:Eu,Dy, $CaYAl_3O_7$:Eu, (Ba,Ca)TiO_3:Pr^{3+}, $BaAl_2Si_2O_8$:rare earth element, $BaSi_2O_2N_2$:Eu^{2+}, $Ca_2Al_2SiO_7$:Ce (Gehlenite, one of the brightest elastico mechanoluminescent materials), ZrO_2:Ti, and ZnMnTe. The rare earth dopant can be Eu. A few polymers and rubbers have also been reported to be elastico mechanoluminescent (Chandra, 2011). Significant studies related to the characteristics of elastico ML have been done by Xu et al. (2002, 2004), Kim et al. (2009), Someya et al. (2013, 2014) and Rahimi et al. (2013).

Whereas detailed studies have been made on the plastico ML (Metz et al., 1957; Kruglov et al., 1966; Butler, 1966; Alzetta et al., 1970; Ossipyan and Shmurak, 1981; Al-Hashimi et al., 1983; Eid et al., 1986; Hagihara et al., 1989; Hayashiuchi et al., 1990; Molotskii, 1989; Ohgaku et al., 2002; Ohgaku and Inabe, 2006; Nakamura et al., 2006; Chandra and Elyas, 1978; Chandra et al., 1983, 2000a,b, 2009; 2010, Chandra, 1984; Chandra and Ramrakhiyani, 1992; Chandra, 1996; Chandra et al., 1996; Atari, 1982) and fracto ML of X- or γ-irradiated alkali halide crystals (Chandra, 1998, 2011; Chandra and Ramrakhiani, 1992; Rajput et al., 2004; Chandra et al., 1993, 2013a,b), least studies have been made on the elastico ML of X- or γ-irradiated alkali halide crystals (Alzetta et al., 1970; Chandra et al., 2010a,b; Chandra and Bisen, 1992; Chandra, 2008). In the past the fracto ML and plastico ML could not be used for the radiation dosimetry because of the destructive nature of materials during plastic deformation and fracture. The present paper shows that similar to the optically stimulated luminescence and thermoluminescence, the elastico ML can also be used in radiation dosimetry. In this case, the same crystal can be used number of times because the elastic deformation does not cause permanent deformation in the crystals, and moreover, comparatively the devices needed for the EML measurements are of low cost and very simple. In the present paper, an attempt has been made to establish the concept of elastico-mechanoluminescence dosimetry.

2. Experimental

The single crystals of KBr, KCl, NaCl and LiF used in the present investigation were supplied by Harsaw Chemical Company. For the EML measurements the crystals were made to the required size by cleaving and polishing. The specimens were annealed at 650 °C for 2 h. The colour centres in the crystals were produced by X-ray irradiation or γ-irradiation. The X-ray irradiations were carried out at room temperature with an X-ray tube having copper target and operating at 40 kV with a tube current of 6 mA. The specimens were irradiated from both sides (for about half the time from each side) for uniform colouration. The irradiated specimens, wrapped in aluminium foil, were kept in dark for about an hour to allow the afterglow to decay to a value well below that expected in the EML measurements. As we do not have the facility for measuring the radiation dose emitted from the X-ray Unit, we determined the dose level in X-irradiated crystals exposed to X-rays for 1 h dose from the ML intensity versus γ-dose plot. For the crystals of same size and same value of applied pressure, the ML intensity of X-irradiated crystals exposed to X-rays for 1 h is nearly equal to that of the γ-irradiated crystals exposed to 430 Gy. Thus, this experiment indicated that the dose level in the crystals exposed to X-rays for 1 h should be nearly 430 Gy.

The γ -irradiations were carried out using a ^{60}Co source (1.25 MeV), in which the dose rate was 690 Gy per hour. Care was taken to avoid the exposure of irradiated crystals to stray light during the X-ray and γ-irradiations and the subsequent measurements during deformation. The optical density of coloured crystal is measured by using a Shimadzu double-beamUV-240 spectrophotometer. The density of F-centres in γ-irradiated crystals was determined using Smakula's formula. In the measurement of the density of F-centers by optical absorption using Smakula's formula, uncertainties involved are as follows: (i) Smakulas formula assumes a Lorentzian form of the curve which does not fit with the experimental one and the same is true for the alternative assumption of Gaussion form which yields a lower value of density of the colour centres (Datta, 1961), (ii) in the derivation of Smakula's formula, it is assumed that all of the absorbing atoms are in the ground state, which may not be the case for many materials, (iii) the free electrons, polarons and other centres absorb the incident light, and therefore, these absorptions may give somewhat higher value of the density of F-centres. Despite these uncertainties Smakula's formula is still being used by most researchers for the determination of the density of F-centers by optical absorption.

The ML-strain curves of the samples were determined by slowly compressing the crystal of a given size at a fixed strain rate using a table model Instron testing machine, where the strain was measured using a differential linear variable transducer (LVDT) and the EML intensity was measured using a photomultiplier tube (Chandra and Zink, 1980). The crystals were deformed along (100) direction. In elastico-mechanoluminescence the crystals are deformed below the limit of elasticity, whereby the crystals return to their original state when the applied pressure is removed. When the applied pressure was released from the crystals deformed in the