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ON THE SATURATION OF RADIATION DAMAGE IN IRRADIATED NATURAL ROCK SALT

H. Donker and A. Garcia Celma

ABSTRACT

Natural rock salt samples of the 800 m. level from the Asse mine, Remlingen, Germany were gamma–irradiated at 100 °C with spent fuel elements from the High Flux Reactor at Petten, The Netherlands. Dose rates in the experiments varied between 200 and 20 kGy/h in monthly cycles. After irradiation the radiation induced stored energy was studied as a function of total dose. Total doses of up to 1200 MGy were reached. Initially there is an approximately linear increase of stored energy with increasing total dose, which levels of at higher doses until it reaches a saturation value of about 140 J/g. The results of the stored energy measurements were compared with those obtained by other scientists for irradiated pure and K–doped NaCl single crystals.

1. INTRODUCTION

One of the considered options for the disposal of radioactive waste consists of depositing the waste in deep rock salt formations. Rock salt however, is known to be very susceptible to radiation damage when exposed to ionizing radiation. Therefore, much research on radiation damage in rock salt and its safety aspects for repository concepts has been performed during the last decades.

The primary defects formed upon irradiation of alkali halides are F– and H–centres, which result from the radiation–less decay of excitons [Itoh, 1982]. Upon prolonged irradiation, depending on the temperature, the F–centres can aggregate forming colloidal metallic particles [Hughes, 1983; Hughes and Jain, 1979]. Also prismatic dislocation loops have been observed to develop during irradiation of alkali halides. According to Hobbs et al. [1973] these dislocation

loops are formed due to the displacement of neighbouring Na^+ and Cl^- ions from their normal lattice sites to the extra half–plane of the dislocation loop. The Na^+ and Cl^- ion are displaced by two H–centres which on their turn form molecular halide centres which occupy the created divacancies. These molecular halide centres surrounding the dislocation loops can possibly collapse into halide gas bubbles [Hobbs, 1974]. A detailed discussion on the basic principles of the damage formation process in NaCl can be found in [Soppe et al., 1994]

One of the considered risks for a repository is the possibility of a sudden back reaction of the Na colloids and the molecular Cl_2 produced under irradiation. Such a back reaction could occur if the concentration of these defects exceeds a certain percolation threshold. It has been reported that such a back reaction might have an explosive character [den Hartog et al., 1993]. Therefore, one of the questions that has to be answered for a repository is whether the created damage will only reach a saturation level well below the percolation threshold or whether this treshold will be reached, making a back reaction possible.

The experimental results obtained for pure single crystals of NaCl by Jenks and Bopp [1974; 1977] and Groote and Weerkamp [1990] show a saturation of damage at a few mol%. These experimental results can be satisfactorily described by the modified Jain–Lidiard model [Jain and Lidiard, 1977; Lidiard, 1979; van Opbroek and den Hartog, 1985]. Calculations with this model simulating repository conditions showed that if a proper disposal strategy is chosen the damage in a repository would also be limited to a few mol% [Bergsma et al, 1985; de Haas and Helmholdt, 1989]. However, results obtained by Den Hartog and coworkers for some doped single crystalline samples showed that for these samples the damage levels were much higher than for pure NaCl, reaching up to 10 mol% damage [den Hartog et al., 1993; Groote and Weerkamp, 1990]. Furthermore Den Hartog claims that natural rock salt samples show a similar behaviour as these doped samples [den Hartog et al., 1993]. The modified Jain–Lidiard model cannot explain the enhancement of damage formation by impurities. Since the natural rock salts in a repository are usually highly impure, the results of Den Hartog et al. put doubts on the predictions obtained with the modified Jain–Lidiard model. The model recently developed by Soppe [1993] however, reasonably reproduces the damage enhancement found for K–doped samples. Simulations of repository conditions with this model showed that also for doped NaCl the damage in a repository would not exceed a few mol% [Soppe and Prij, 1994]. The model of Soppe was shown to be capable of reproducing a broad spectrum of experimental results.

The natural rock salt in a repository is, however, very different from the homogeneous single crystals studied in most experiments. Natural rock salts are polycrystalline and very heterogeneous. Although some irradiation experiments on natural rock salts have been performed [Swyler et al., 1979; 1980; Levy, 1983; Levy et al., 1980; 1983; 1984; Levy and Kierstead, 1982; Loman et al., 1982], very little is known about the effects of polycristallinity and heterogeneity on the formation of radiation damage. It has been known that after irradiation Fluid Assisted Recrystallization occurs in polycrystalline salt samples [Garcia Celma et al., 1988]. Recently, Garcia Celma and Donker [1994a] have shown that in natural samples irradiated at relatively low dose rates various recrystallization and recovery processes occur during irradiation.

In this paper the results of stored energy measurements on natural rock salt samples (Asse speisesalz of the 800 meter level) irradiated at relatively high dose rates (see below) are presented. These irradiation experiments have been performed in order to see whether the damage in these samples would reach a saturation and if so at which level. And also in order to check whether these samples would behave like K–doped single crystals as claimed by den Hartog et al.[1993] or not. The results will therefore, be compared with results obtained by other scientists for pure and doped NaCl.

2. EXPERIMENTAL

2.1. Samples

For the presently described experiment we have used natural salt samples, i.e. Speisesalz from the 800 meter level of the Asse mine, Remlingen, Germany (Sp–800). These salt samples consist of a relatively high purity (> 99.0 %) polycrystalline halite rock, with a grain size of 3 – 10 mm. The main impurity phase in these samples is polyhalite ($K_2MgCa_2(SO_4)_4.2H_2O$ – ~1%), while occasionally anhydrite ($CaSO_4$) is also present. The material has besides the structurally bound water (polyhalite) a water content of about 0.05 wt % which is mainly present in fluid inclusions at grain boundaries. Dislocation densities within individual grains are rather low while slip and kink bands are almost entirely absent [Spiers et al., 1986; Gies, 1995].

Samples in the shape of cylindrical tablets with a diameter of about 24 mm and a height of 10 mm were prepared as described in [Garcia Celma et al., 1991] and placed in golden sample holders. Five of these golden sample holders were placed on top of each other in a stainless steel holder where they were held in position by a spring. Care was taken that the fitting between the samples and the holders and between the golden and steel holders was as tight as possible to ensure a good thermal contact between the samples and the irradiation facility. Eight of the stainless steel holders were placed in one of the Gamma Irradiation Facilities (GIF) in the pool of the reactor at Petten (the Netherlands) so that 40 samples were irradiated at the same time [Garcia Celma et al., 1995].

2.2. Irradiation set–up

The Gamma Irradiation Facility (GIF) of the ECN is extensively described in [Garcia Celma and Donker, 1994b]. Essentially each GIF experiment is performed in a long container equipped with heating and cooling devices in which eight sample holders can be placed. The container is placed between the spent fuel elements in the cooling pool of the High Flux Reactor (HFR). The temperature for the experiments was kept at 100°C. For the GIF A experiment, fresh spent fuel elements from the HFR were used in order to obtain a dose rate as high as possible. Each month four of the older spent fuel elements were replaced by fresh elements. As a consequence, the dose rate in our experiment varied between 200 and 20 kGy/h in monthly cycles. Each month one of the samples was retrieved from the facility and replaced by a new one.

Dose rates were measured at regular intervals using red perspex dosimeters. The measurements are performed by temporarily removing the sample container from its position and lowering a measuring tube containing the perspex between the spent fuel elements at the position of the sample container. The measuring tube is designed in such a way that its absorbing power for gamma rays is equal to that of the sample container. To calculate the total dose received by the samples in each cycle the measured dose rates are fitted to the equation:

$$D = A_0 + A_1 e^{(-A_2 t)} + A_3 t \qquad (1)$$

in which D is the dose rate, t is time and A_0, A_1, A_2 and A_3 are fitting parameters. This equation is then integrated to calculate the total dose. As an example the measured dose rates and obtained fits for one of the samples are shown in Fig. 1.

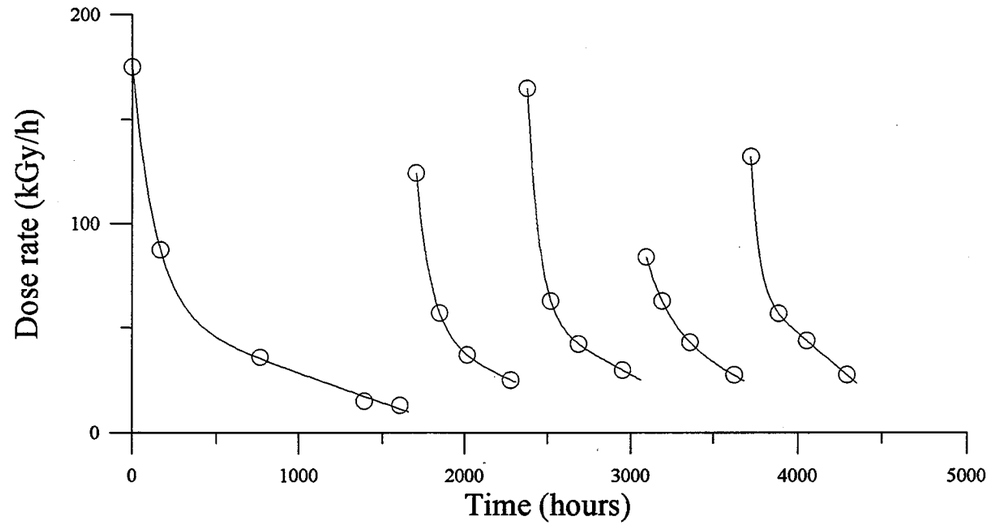

Figure 1: *Dose rate history of one of the irradiated Sp–800 samples.*

2.3. Stored energy measurements

The stored energy measurements were carried out by means of differential thermal analysis on a SETARAM DSC–111. Calibrations were performed by melting Indium metal. Anneals take place in "closed" Pt capsules with a small capillary hole to allow gases to escape in order to avoid explosion of the capsule when pressure builds up. The heating rate used for these measurements was, if not indicated otherwise, 10 K/min. In a first run the sample is measured against an empty reference sample holder up to a temperature of 750 K. Then the sample and reference are allowed to cool down and a background signal is measured in a second run. The result of the second run is subtracted from that of the first. The area closed by the resulting curve and the base line is then integrated to obtain the released stored energy.

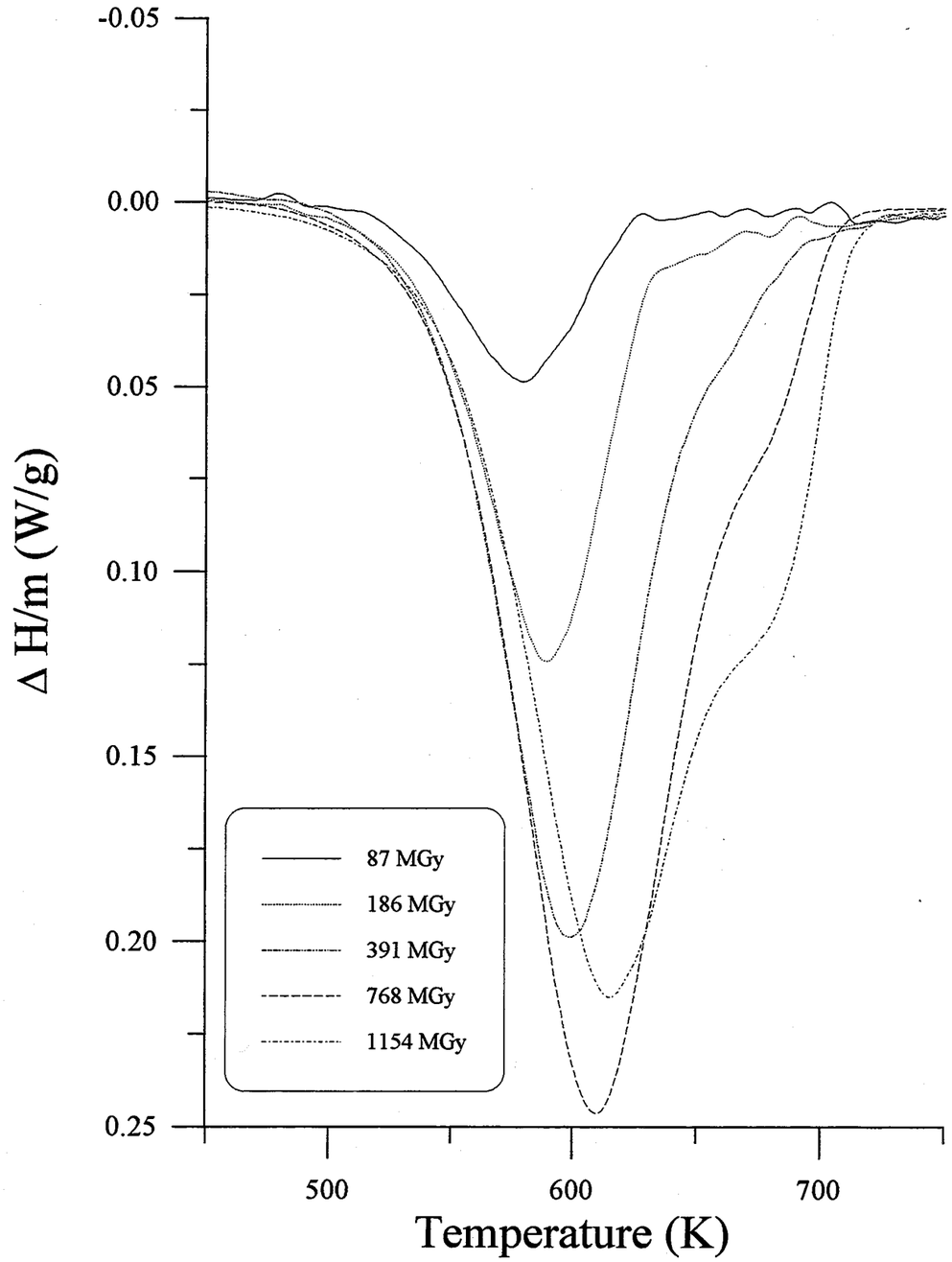

Figure 2: *DTA curves of irradiated Sp–800 samples. Heating rate 10 K/min.*

For the stored energy measurements, parts of the irradiated samples were cut in small pieces. The darkest of those pieces (i.e. free from secondary minerals and recrystallized material) were selected with the naked eye. Small portions of secondary minerals and recrystallized material may however, still have been present in the analyzed material.

According to the measurements of Jockwer [1981], polyhalite loses its crystal water between 235 and 350 °C. Our analysis showed that the dehydration of polyhalite takes place in the same temperature interval as the release of stored energy. This results in the presence of an endothermal peak superposed on the exothermal release of stored energy. When necessary our results have therefore, been corrected for the dehydration of polyhalite.

3. RESULTS AND DISCUSSION

A few representative results of the DTA measurements on the irradiated Sp–800 samples are shown in Fig. 2 where the heat flow (ΔH) divided by the sample weigth (m) is plotted versus temperature (T). At low total dose only one single stored energy peak is observed which maximum lies at about 600 K. At higher total doses, however, a shoulder develops on the high temperature side of this peak. The results of the DTA measurements performed on Sp–800 samples irradiated up to high total doses are similar to the results of the DSC measurements on Ba, K or F doped NaCl reported by Grootte and Weerkamp [1990]. We have considered that the shoulder might be caused by a possible endothermal signal from the dehydration of polyhalite. However, we have to reject this possibility for two reasons:

a) In Sp–800 samples in which large amounts of polyhalite are present the temperature of the maximum of the endothermal peak corresponding to the dehydration is situated at 625 K. If the shoulder in the presently described measurements would be due to the dehydration of polyhalite it ought to appear at a lower temperature.

b) The dehydration of polyhalite is normally accompanied by a considerable mass loss due to the evaporation of water. For most of the Sp–800 samples showing the shoulder in their DTA curve, the mass loss during these measurements was negligible.

Therefore, we have to conclude that the DTA curves consist of a double stored energy peak.

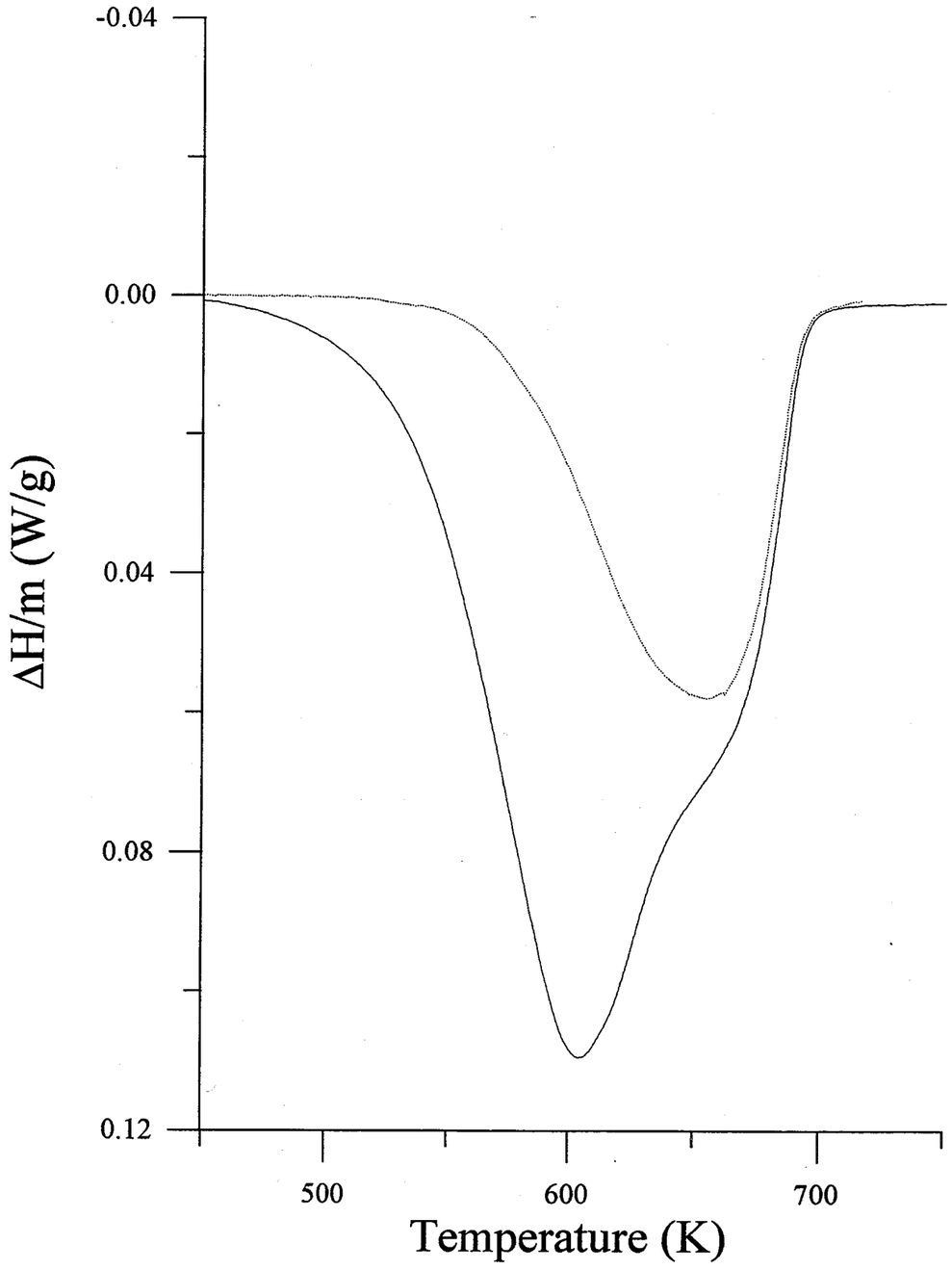

Figure 3: *DTA curves of irradiated Sp–800 sample (Total dose 1223 Mgy). Drawn line: non–annealed sample, dashed line: sample annealed for 30 minutes at 575 K. Heating rate 5 K/min.*

To check this conclusion we annealed one of the irradiated samples showing this double peak during 30 minutes at about 575 K. Then the sample was allowed to cool down to about 400 K. After that we performed a stored energy measurement as usual. The thus obtained DTA curve (Fig. 3) shows a single exothermal peak with a maximum at 655 K. This confirms our conclusion since if the shoulder would have been caused by an endothermal signal we ought to have observed it also after the anneal, what is clearly not the case. The area under the peak observed at 655 K corresponds to a stored energy of 56 J/g. After the anneal at 575 K the sample was still dark blue coloured, almost black. We therefore have to conclude that both stored energy peaks are caused by sodium colloids in the samples and that obviously there are two different kinds of colloids. A closer look at the DTA curves of all samples irradiated above 800 MGy shows that, with increasing total dose, the peak at 655 K seems to grow at the expense of the peak at 600 K, while the total stored energy remains approximately constant (see below). This indicates that one kind of colloid is converted into the other. A possible explanation for this conversion might be a phase transition which occurs when the colloids grow bigger. For very small colloids the sodium atoms are expected to retain the positions of the FCC Na sublattice of NaCl. The crystal structure of bulk sodium however, is BCC. A phase transition from FCC to BCC sodium colloids is therefore expected. Another explanation for the two kinds of colloids might be that certain sizes of colloids are energetically favourable due to the mismatch between the lattice parameters of the Na–lattice and the NaCl–lattice. Further research on this subject is however necessary.

The results of the stored energy measurements on the irradiated Sp–800 samples are shown in Fig. 4 as a function of total dose. The results show that, at low total doses, there is an approximately linear increase of stored energy with increasing total dose. At high doses (> 400 MGy) this increase levels off and the stored energy reaches a saturation value of approximately 140 J/g. In Fig. 4, the results obtained by Jenks and Bopp [1977] for pure NaCl Harshaw crystals, irradiated at 95 °C and a constant dose rate of 100 kGy/h and the results obtained by Den Hartog and coworkers [den Hartog et al., 1993; Groote and Weerkamp, 1990] for pure and 1 mol% K doped NaCl samples irradiated at 100 °C and a constant dose rate of 120 kGy/h are shown for comparison. From Fig. 4 it can be seen that the dependence of the stored energy on total dose observed for the Sp–800 samples is similar to that observed by Jenks and Bopp and Den Hartog and coworkers for pure NaCl single crystals although the results for the Sp–800 samples always are somewhat lower than the results obtained for pure NaCl by the other authors. We estimate the error for our data at about 20 % (both in the measured stored energy as in the calculated total

dose) while probably the same applies for the other experiments. In view of these large errors the correspondence between the experiments is quit good

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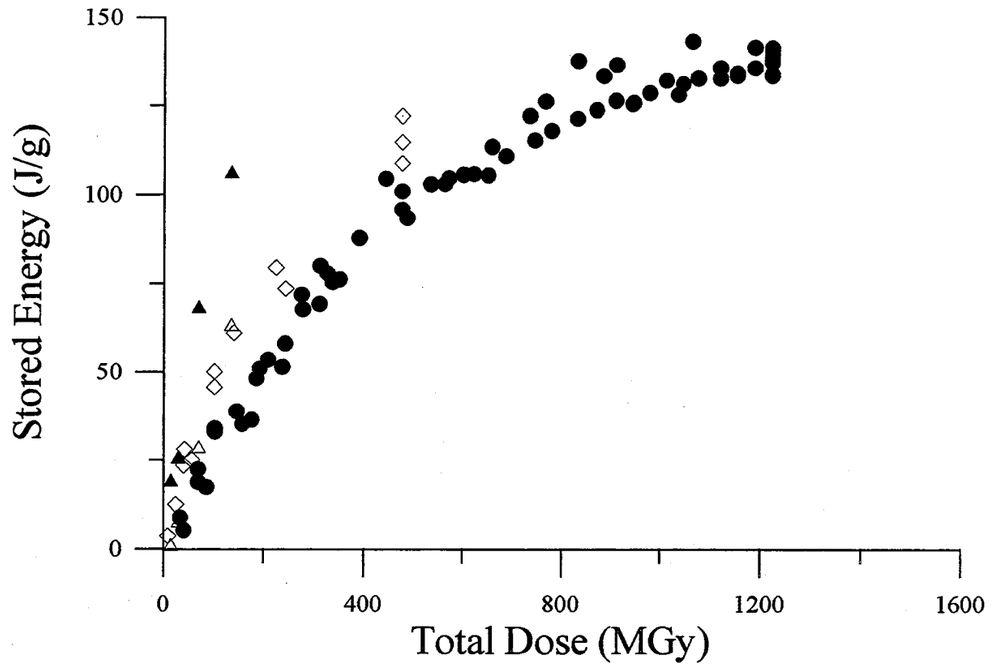

Figure 4: *Stored energy as a function of total dose. Full circles: Sp–800 samples, open squares: pure NaCl irradiated at 95 °C and 100 kGy/h by Jenks and Bopp, open triangles: pure NaCl and closed triangles: K–doped NaCl both irradiated at 100 °C and 120 kGy/h by den Hartog et al.*

The differences with the K–doped samples irradiated by Den Hartog et al. are, however, larger. Together with the Sp–800 samples shown in Fig. 4, we have also irradiated a Harshaw NaCl single crystal. The stored energy in this single crystal is approximately equal to that of the Sp–800 sample with the same irradiation history (total dose 279 MGy, stored energy 63 and 67 J/g respectively). Also from many other experiments in which we have irradiated Harshaw crystals and Sp–800 samples simultaneously we have observed that although their DTA curves are very different, the amount of stored energy developed in these samples is not very different

[Garcia Celma and Donker, 1994b]. We therefore have to conclude that the Sp–800 samples in this aspect do not show a similar behaviour as K–doped samples, contrary to the natural samples studied by Den Hartog et al. Moreover, as can be seen from Fig. 4, we have clearly observed a saturation behaviour for the stored energy developed in our Sp–800 samples. Den Hartog et al have reported that their K–doped samples (although irradiated at a much higher dose rate) do not show a saturation behaviour.

For Sp–800 samples irradiated at low dose rate (15 kGy/h) to relatively low total dose (0.02 to 44.6 MGy) we have shown that various recrystallization and recovery mechanisms are operative, competing with the damage development process [Garcia Celma and Donker, 1994a]. One of these processes, Fluid Assisted Recrystallization (FAR) can take place provided that intergranular brine is present in the salt [Garcia Celma et al., 1988; 1993]. Sp–800 samples on the average contain 0.05 wt% of water, mainly in fluid inclusions at their grain boundaries [Spiers et al., 1986]. It is therefore very unlikely that FAR will not have operated in our samples. However, if colloids are present in the salt the water needed for FAR will be decomposed during its operation. Since we have observed, from the dark blue colour of the samples, that colloids are present in all presently reported samples, FAR can only have played a role in the low dose region of the described experiment. In the high dose region all the water in the sample will have been decomposed and the operation of FAR will have been stopped. To what extent other recrystallization and recovery processes have played a role we do not know since the irradiated samples are so deeply coloured that it was impossible to produce mechanically the thin sections in which the microstructures of the samples could be observed. Recently, we produced the desired thin sections by etching with water, but the thus produced thin sections are of course not too reliable.

4. CONCLUSION

Sp–800 samples irradiated in the GIF A facility, to total doses up to 1200 MGy, show a saturation of damage at a level of about 140 J/g stored energy. The amounts of stored energy observed for these samples are not higher than those observed by other scientists for pure NaCl single crystals irradiated under comparable conditions.

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ON THE RELATIONSHIP BETWEEN STORED ENERGY AND COLLOIDAL SODIUM PRODUCTION IN IRRADIATED ROCK SALT

H. Donker, W.J. Soppe and A. García Celma

ABSTRACT

A few parameters of the theoretical models used to describe the formation of radiation damage in rock salt were critically reviewed. It is discussed that the back reaction used in the models should be described as $\gamma = 10^{16} \exp(-0.4/kT)$ and that for the conversion factor between defect concentrations and stored energy a value of 5 eV/F–H pair should be used. With these modified parameters the models were compared with the experimental results of Jenks and Bopp and the experimental results obtained from the GIF A irradiation experiments.

1. INTRODUCTION

The first model describing the formation of radiation damage in alkali halides was developed by Jain and Lidiard [1977]. This model was later modified by Van Opbroek and den Hartog [1985], according to a proposal of Lidiard [1979]. This modification, the inclusion of a back reaction, was introduced in order to be able to explain the experimental results of Jenks and Bopp [1974;1977] and Jenks et al. [1975].

A disadvantage of this model is that it does not describe the nucleation stage of the colloids and dislocation loops. Moreover, the effects of impurities, strain and grain boundaries are not taken into account. Recently two new, slightly different models based on the old Jain–Lidiard model, but with extensions describing the nucleation stage have been developed [Soppe, 1993; Seinen et al.,1992].

In a recent paper [Soppe et al., 1994] the experimental results on radiation damage