

RADIATION DOSE DEPOSITION AND ENERGY ACCUMULATION IN A ROCK SALT WASTE REPOSITORY

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2. IRRADIATION EFFECTS IN ALKALI HALIDES

Mechanisms of defect creation by the irradiation of alkali halides and of their annealing afterwards have been subject of many theoretical and experimental studies [12,13] during the last decade. Such studies became of technological interest when geological rock salt formations were considered for use as nuclear waste repository medium. It is in view of such applications that the presently available knowledge and information will be briefly reviewed. Based on this information calculated values for irradiation dose deposition will be used to estimate such effects as build-up of stored energy and formation of colloidal alkali metal. It seems to be evident that a relation exists between these two effects although this may not be a simple one.

Most experimental investigations have been carried out by means of high energy electron beams from an accelerator facility or by means of gamma radiation or X-rays. It is important to understand that the effects in the alkali halide structure of gamma-ray produced Compton and

photo-electric recoil electrons are very similar to those of high-energy electrons. Of course, the transmission properties through the material are very much different for the various types of radiation. In a study of stored energy effects Delgado and Alvarez Rivas [6] pointed out that there may occur appreciable quantitative differences in defect creation by gamma radiation and high energy electrons and for the latter there are very noticeable differences if the electron energy has different values.

Considering processes of irradiation damage, different stages of effects should be distinguished. The primary effect of electron or gamma radiation penetrating into the alkali halide lattice is ionization, i.e. the formation of a free electron and an electron hole. In many cases they will quickly afterwards recombine leaving an undamaged lattice in which the energy will be released as heat.

In other cases the ionization products will be trapped in the lattice. Due to this ionization the chlorine anions in sodium chloride may become positively charged and are then easily expelled from their lattice positions.

This may give rise to two effects of irradiations.

(1) The anion vacancy will catch an electron thus forming an F-centre and

(2) the chlorine ion will occupy an interstitial position. Associating with another chlorine ion located at a lattice site it can form a pair of chlorine ions now located around the lattice position and aligning its molecular axis either along the direction $[110]$ or $[111]$. Defects in the form of such molecular centres are generally characterized as H-centres [14].

Depending on temperature, impurities and dislocation density both the F-centres and the H-centres may become mobile and then give rise either to the formation of conglomerates or to annealing and restoring of the original lattice.

(1) Conglomeration of F-centres will lead to local regions with excess of sodium ions and if such regions become sufficiently large it is probable that due to gradually changed atom potentials the electron energy bands will be rearranged according to a scheme for sodium metal. In that case one may speak of particles of colloidal sodium.

(2) Similarly a mechanism of coagulation of chlorine molecular centres may be expected, giving rise to minor chlorine bubbles.

Although the processes for the primary defect creation are now reasonably well understood, the complexity of secondary processes is still such that no unified description has been accomplished.

With the purpose of making estimates of the build-up of stored energy due to radiation damage and the possible hazards of this phenomenon when using salt mines as waste repositories, an experimental programme was carried out at Oak Ridge National Laboratory by Jenks and Bopp [3,4,5]. They measured the stored energy build-up in rock salt specimens from a variety of sources and irradiated by gamma-radiation to several dose values and they studied the kinetics of the defect creation of the simultaneously occurring back-reaction and a following annealing process. It is important to note that they carried out their investigations in the range of dose levels that will be encountered in rock salt directly surrounding waste containers in a repository. Their observations and conclusions can be summarized as follows.

The rate of formation of stored radiation energy can be expressed by

$$dE/dt = k_1 I \quad (2.1)$$

Here E is the amount of stored energy, I is the dose rate and k_1 is the formation constant.

Taking into account a term for radiation induced annealing the expression becomes

$$dE/dt = k_1 I - k_2 I E \quad (2.2)$$

where k_2 is an annealing constant.

The formation term $k_1 I$ can be understood on the basis of defect-formation as has been described above. The form of the back-reaction term can be explained if it is assumed that a fraction of the freshly formed primary radiation defects react with existing defects to form undamaged NaCl.

That fraction is proportional to the number of defects already present in the crystal. The reaction constant k_2 varies with temperature since the back reaction term in expression (2.2) depends on the state of aggregation of these defects. At lower temperatures the defects will be more in the form of F-centres whereas at higher temperatures they will have coagulated into colloidal sodium. It is evident that mobilities associated with the two states of defects will be different.

Expression (2.2) leads to the integral

$$E = k_1/k_2 (1 - \exp(-k_2 D)) + E_0 \exp(-k_2 D) \quad (2.3)$$

Here D is the collected radiation dose $D = \int I dt$ and E_0 is the stored energy at zero time, which usually can be taken equal to zero. Expression (2.3) shows that this phenomenology leads to a saturation for the amount of stored energy.

At temperatures above 150 C the annealing apparently occurs according to an additional term in equation (2.2), which turned out to be of zero-kinetic order:

$$dE/dt = -K.$$

This means that the rate of annealing is independent of the amount of stored energy. This is a rather rarely occurring process, which can be explained by assuming a rapidly adjusting equilibrium between coagulated and single defects, e.g. colloidal sodium and F-centres. The rate determining step is some kind of an "evaporation" process due to which the concentration of evaporated single defects does not depend on the amount of conglomerated defects.

This concentration will be maintained at a constant value until e.g. the colloidal sodium has disappeared. The evaporation process is thermally activated and consequently the single defect concentration is temperature dependent. For temperatures significantly higher than 150 C the effect of this annealing process is such that build-up of stored energy can be neglected. With this additional process taken into account expression (2.2) becomes

$$dE/dt = k_1 I - k_2 I E - K \quad (2.4)$$

and by integration

$$E = (k_1 - (K/I)) / k_2 (1 - \exp(-k_2 D)) \quad (2.5)$$

Here the zero-time stored energy has been neglected. The saturation level associated with expression (2.5) depends strongly on the temperature regime. Between 100 and 150 C it amounts to about 75 J/g and below 100 C it rises to 245 J/g.

Assuming a stored energy of 4.25 eV per defect pair $|I_2|$ these saturation values correspond to a defect concentration of 1.1% and 3.5% mole fraction.

Lidiard $|12|$ pointed out that such large concentrations can only be retained by colloids since F-centre concentrations saturate at much lower values.

However, he argues that according to the Jain-Lidiard theory $|7|$ the rate of energy release is not compatible with the evaporation of F-centres from colloids being the rate-determining step, but that it is rather the recombination of F-centres with molecular centres. This can only be a valid interpretation if the molecular centres are clustered together, because of the zero-order character of the reaction. The F-centres apparently maintain an equilibrium by relatively rapid evaporation from colloids.

In agreement with the above-mentioned theory it was found by Jenks

and Bopp [4]:

i) that the formation of stored energy in the dose regime when saturation may be reached is insensitive for dose rate and

ii) that beyond a maximum temperature and a minimum dose rate the build-up of stored energy is negligible.

The measurements of the energy release were confirmed by chemical analysis determining the amounts of hydrogen and Cl⁻, that were formed by solution of specimens in water.

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3.5. CALCULATION OF ENERGY STORAGE

Based on the calculations of radiation dose deposition in rock salt around radioactive waste containers it has been attempted to make estimates of the build-up of energy storage. It has been pointed out in chapter 2 that this energy build-up is strongly dependent on the temperature of the rock salt. For the configurations that were considered and that have been specified in table 3.1. the development of temperature with time has been calculated at several distances from the waste containers.

These calculations show that in only a few configurations the material directly surrounding a container will reach temperatures above 150 °C and then only during a limited period of time.

(*) Therefore, it has been assumed that the zero-order annealing process according to the third term in expression (2.4) may be neglected. In the few cases in which this process may be active it will have the effect to decrease both the build-up of stored energy and the formation of colloidal sodium.

(*) The calculations have been carried out according to expressions (2.2) and (2.3) with the assumption that the zero-time stored energy E_0 is equal to zero.

(*) From the investigations by Jenks and Bopp [4,5] it follows that the energy build-up is strongly temperature dependent. From their results the following data have been used in the calculations.

For $T > 100^\circ\text{C}$:

$$k_1 = 6.95 \cdot 10^{-9} \text{ J/(g rad)}$$

$$k_2 = 9.3 \cdot 10^{-11} \text{ rad}^{-1}$$

For $T < 100^\circ\text{C}$:

$$k_1 = 20.9 \cdot 10^{-9} \text{ J/(g rad)}$$

$$k_2 = 8.5 \cdot 10^{-11} \text{ rad}^{-1}$$

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