THE USE OF ELECTRON ACCELERATORS FOR SIMULATION OF RADIATION DAMAGE IN ROCK SALT CONSIDERED FOR SAFE STORAGE OF NUCLEAR WASTE

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Rock salt is one of the considered storage mediums for nuclear high level waste (HLW), and so the behavior of rock salt in the vicinity of HLW is of great practical interest. The main contribution to the irradiation from a radioactive waste comes from gamma-radiation, which requires a comparison of its action with that of the electron irradiation, widely used for simulation of radiation damage in rock salt. To solve this problem, we have used a linear electron accelerator to produce bremsstrahlung gamma-radiation spectrum similar to the real spectrum of HLW. Samples of synthetic crystals of NaCl have been γ- irradiated up to the dose ranging from 1.2 10^8 Gy to 7.8 10^8 Gy and subsequently investigated by means of differential scanning calorimetry and atomic force microscopy. Electron and gamma irradiated samples have shown a comparable damage concentration and the structure of the radiolytic products, which justifies the use of electron irradiation for the imitation of radiation damage in the vicinity of HLW.

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1. INTRODUCTION

Rock salt is an attractive storage medium for nuclear high level waste (HLW) since geological rock salt deposits stay away from water for millions of years. However, in-laboratory electron irradiation has been shown to result in formation of radiolytic products, namely, nano-sized sodium colloids and chlorine bubbles, followed by formation of relatively large voids eventually bringing the halogen gas and metal to a back reaction inside the voids resulting in explosive fracture of samples with particular dopants [1-3]. So, the behavior of rock salt in the vicinity of HLW should be investigated very carefully.

One of the problems of imitation of actual conditions of irradiation is that the main contribution to the irradiation from a radioactive waste consists of gamma-radiation, which requires a comparison of its action with that of the electron irradiation. To solve this problem, we have used a linear electron accelerator ELIAS of the NSC KIPT to produce bremsstrahlung gamma-radiation spectrum similar to the real spectrum of HLW. Samples of synthetic crystals of NaCl (pure and doped by KBF_4, KCl, and NaBr) have been γ- irradiated up to the dose ranging from 1.2 10^8 Gy to 7.8 10^8 Gy (12 Grad to 78 Grad) and subsequently investigated by means of differential scanning calorimetry and atomic force microscopy. The results have been compared with those produced by electron irradiation of NaCl samples at the University of Groningen, and analyzed in the framework of a new theoretical model [4,5].

2. EXPERIMENTAL PROCEDURE

2.1. ELECTRON IRRADIATION

The irradiation source consisted of a LINear electron ACcelerator (LINAC), driven by the high voltage produced by a cascade generator, which was enclosed by a steel tank and placed (for reasons of radiation safety) in a bunker. With this LINAC system we were able to irradiate a target area of 320 cm^2 at dose rates between 10 Mrad/hr and 1.5 Grad/hr in the temperature interval 40…150°C.

We have carried out four irradiation runs to different dose levels in the temperature range from 40 to 150°C. As a result, a total number of 1335 samples (pure and alloyed by KBF_4, K, Br, Ba, Li and F) have been irradiated up to the total absorbed dose in the range from 20 to 1000 Grad with the dose rate ranging from 240 to 1000 Mrad/hour.

2.2. GAMMA-IRRADIATION

For obtaining bremsstrahlung gamma-radiation spectrum that is similar to the one from a real radioactive waste we have used a 3 MeV electron beam produced by Van-de-Graaf electrostatic electron accelerator ELIAS of the model KS/3000. The electron beam was converted into a beam of gamma-quanta by a specially designed converter. The converter design was based on modeling results on 3 MeV electron interaction with converter construction elements with account of radiation and radiation-chemical stability of converter construction elements. Thus, the 3 MeV electron beam with current density 150 μA/cm^2 was converted into the gamma-quanta beam with intensity of about 10^{14} γ-quanta/cm^2sec.

A special thermostatic chamber for irradiation of rock-salt specimens has been designed and manufactured together with the converter. It allows irradiating four rolls of rock-salt tablet-shaped specimens with diameter of 5.5…6.0 mm by gamma-quanta simultaneously at a constant temperature. Each roll may contain up to 25 specimens of one material with thickness of 0.7 mm.

The temperature can be stabilized during irradiation in the range from 40°C to 150°C with accuracy of 1

°C. Along the specimen roll, the intensity of gamma-quanta decreases monotonously, which allows obtaining specimens with different absorbed dose during one irradiation run.

In this study, 100 samples of synthetic crystals of NaCl (pure and alloyed by KBF_4, K, and Br, 25 samples of each kind) have been irradiated by γ-quanta at 100°C up to the total absorbed dose in the range from 12 to 78 Grad.

2.3. ANALYTYCAL METHODS

The irradiated samples have been examined by means of differential scanning calorimetry (DSC) and atomic force microscopy (AFM).

The AFM results were obtained in ultra high vacuum (UHV) using an UHV AFM/STM Omicron system in the non-contact mode. To avoid chemical reactions between the radiolytic Na colloids and oxygen or water, the irradiated samples were cleaved, prepared and kept under UHV conditions prior to and during the experiments. Several AFM images have been obtained from different areas for each sample. The radii of AFM tips, used in this investigation, have been estimated to be in the range between 20 and 30 nm

DSC measurements are very suitable for the determination of the volume fraction of metallic Na produced inside the sample during the irradiation process. The latent heat of melting (LHM) of the Na colloids has been measured for each sample with a Perkin-Elmer DSC-7 system. The LHM is proportional to the volume fraction of the Na colloids so that the LHM equal to 1 J/g corresponds approximately to the volume fraction of 2%. The difference in heat flow between the irradiated sample and the reference sample (unirradiated NaCl) was measured during heating from 50ºC to 150ºC with a heating rate of 10ºC/min. The appearance of the LHM peaks in the 'DSC-spectrum' in this temperature interval is evidence for the presence of sodium colloids in the sample.

3. RESULTS AND DISCUSSION

The nano-particles of metallic sodium formed after electron and gamma irradiation of NaCl crystals, doped by K are visualized on the AFM images shown in Fig.1. The precipitates shape and sizes are slightly distorted because of the convolution of the surface with the AFM tip with irregular-shaped apex. It can be seen that the sizes and the concentrations of sodium colloids formed after electron and gamma irradiation are comparable.

The dose dependence of the latent heat of melting (LHM) of the Na colloids in electron and gamma-irradiated NaCl crystals, doped by K is shown in Fig.2.

It can be seen that the LHM (and consequently the volume fraction) of Na colloids produced by gamma irradiation at the maximum absorbed dose is lower than that produced by electron irradiation by a factor of two.

This difference can be explained in the framework of the model [4,5], as shown in Fig.2, which compares the theoretical and experimental results.

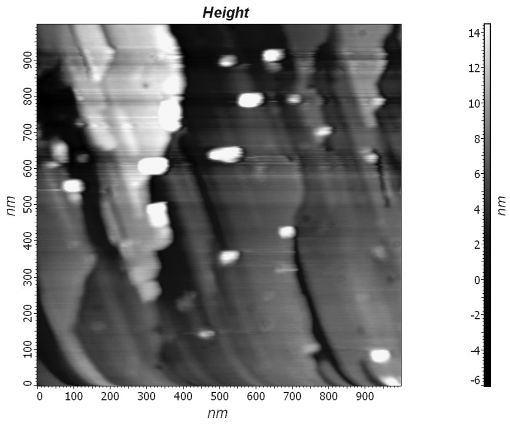

Fig.1. Typical AFM-images of NaCl crystals, doped by K and irradiated at 100 C up to 10 Grad with electrons (the upper picture) and up to 11.4 Grad with gamma-quanta (the lower picture)

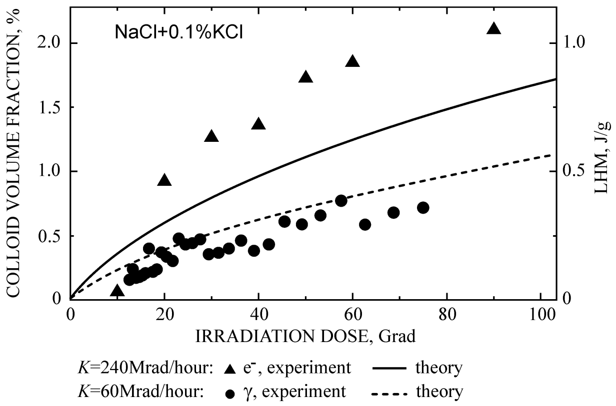

Fig.2. Dose dependence of the colloid volume fraction in NaCl+0.1%K at 100°C for different types and dose rates of irradiation: K = 60 Mrad/h (gamma) and 240 Mrad/h (electron)

The model describes the evolution of sodium colloids, chlorine bubbles and voids under given irradiation conditions that include the absorbed dose rate and temperature. The theoretical curves in Fig.2 have been calculated for different dose rates without taking into account the type of irradiation.

The comparison between theoretical and experimental results shows that the observed difference in colloid production under electron and gamma irradiation may be explained by the difference in the dose rates rather than the irradiation type.

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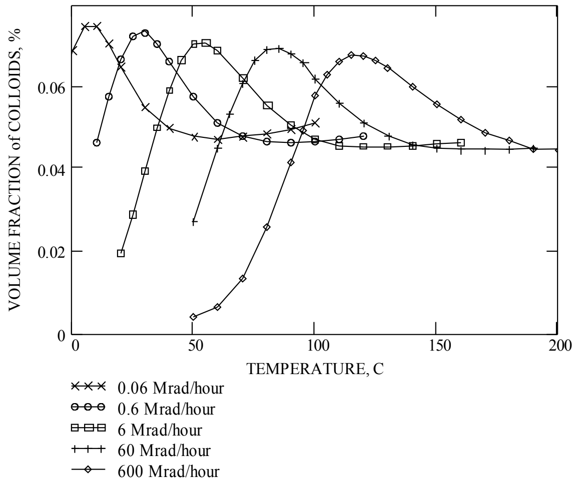

Fig.3. Temperature dependence of the colloid volume fraction in NaCl calculated at different dose rates for the total absorbed dose of 1 Grad

The temperature dependence of the colloid volume fraction in NaCl calculated at different dose rates (Fig.3) has a bell-shaped form, which shifts to higher temperatures with increasing dose rate. It means that in order to imitate irradiation conditions in HLW repository (which corresponds to about 0.06 Mrad/hour) one should increase the temperature of in-laboratory irradiation (60…600 Mrad/hour) by 100…150°C.

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ИСПОЛЬЗОВАНИЕ ЭЛЕКТРОННЫХ УСКОРИТЕЛЕЙ ДЛЯ ИМИТАЦИИ РАДИАЦИОННЫХ ПОВРЕЖДЕНИЙ В КАМЕННОЙ СОЛИ, РАССМАТРИВАЕМОЙ В КАЧЕСТВЕ СРЕДЫ ЗАХОРОНЕНИЯ РАДИОАКТИВНЫХ ОТХОДОВ

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Одна из проблем имитации действия радиоактивных отходов (РАО) на среду захоронения заключается в том, что основной составляющей радиационного фона вокруг РАО является гамма-облучение, что требует сравнения его действия с действием имитационного электронного облучения. Для решения этой проблемы было использовано тормозное гамма-излучение, получаемое на линейном электронном ускорителе, близкое по спектру к гамма-излучению РАО. Образцы NaCl были подвергнуты гамма-облучению до доз в интервале 1.2 10^8…7.8 10^8 Грей и затем исследованы методом дифференциальной сканирующей калориметрии и атомно-полевой микроскопии. Исследование показало, что концентрация и структура продуктов радиолиза в образцах, облученных гамма-квантами и электронами, хорошо согласуются.

ВИКОРИСТАННЯ ЕЛЕКТРОННИХ ПРИСКОРЮВАЧІВ ДЛЯ ІМІТАЦІЇ РАДІАЦІЙНИХ УШКОДЖЕНЬ У КАМ'ЯНІЙ СОЛІ, РОЗГЛЯНУТОЇ ЯК СЕРЕДОВИЩЕ ПОХОВАННЯ РАДІОАКТИВНИХ ВІДХОДІВ

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Одна із проблем імітації дії радіоактивних відходів (РАВ) на середовище поховання полягає в тім, що основної складової опромінення навколо РАВ є гамма-опромінення, що вимагає порівняння його дії з дією імітаційного електронного опромінення. Для вирішення цієї проблеми було використано гальмове гамма-випромінювання, одержуване на лінійному електронному прискорювачі, що близьке по спектру до гамма-випромінювання РАВ. Зразки NaCl були піддані гамма-опроміненню до доз в інтервалі 1.2 10^8 …7.8 10^8 Грей і потім досліджені методом диференціальної скануючої калориметрії і атомно-польовій мікроскопії. Дослідження показало, що концентрація й структура продуктів радіолізу в зразках, опромінених гамма-квантами і електронами, добре погоджуються.