RECENT RADIATION EFFECT STUDIES ON NATURAL AND
SYNTHETIC ROCK SALT FOR WASTE DISPOSAL APPLICATIONS [†]

K. J. Swyler, R. W. Klaffky* and P. W. Levy
Brookhaven National Laboratory
Upton, New York 11973

NOTICE
This report was prepared as an account of work
sponsored by the United States Government. Neither the
United States nor the United States Department of
Energy, nor any of their employees, nor any of their
contractors, subcontractors, or their employees, makes
any warranty, express or implied, or assumes any legal
liability or responsibility for the accuracy, completeness,
or usefulness of any information, apparatus, product,
or process disclosed, or represents that its use would not
infringe privately owned rights.

MASTER

Extensive radiation damage will be formed in rock salt and other minerals adjacent to high level radioactive waste canisters. If the canisters remain intact only gamma-ray induced damage will be formed. Appreciable additional damage will be created by alpha particles and beta rays if the canisters are breached and mixing occurs. If water is present, extensive mixing is expected. Radiation damage information is essential for repository design, site selection, risk assessment, etc. To extrapolate from laboratory measurements to the long time periods and dose rates in repositories, it is essential to develop a reasonably complete understanding of radiation effects in selected geological materials.

To date, the waste disposal radiation damage studies at Brookhaven National Laboratory have been concentrated on synthetic and natural rock salt. Studies are made with equipment for recording the optical absorption of samples, located in a temperature controlled chamber containing inert gas, during and after irradiation with 0.5 to 3.0 MeV electrons. Spectra are recorded as often as every 40 sec in the 3.1 to 1.55 eV (400 to 800 nm) region. This region contains the F-center band, which is a measure of the Cl^- ion vacancies, and the colloid band which is a measure of the concentration and size of the colloid particles.

As shown by previous studies on natural and synthetic salt,⁽¹⁻³⁾ during irradiation the F-band increases monotonically to a plateau while the colloid band absorption remains at a low level until the F-center plateau is reached. At this dose the colloid formation rate changes from a small to a large value.

*Now at Los Alamos Scientific Laboratory.

[†] Work supported by DOE under contract EY-76-C-02-0016.

59

Thus the colloid growth follows a nucleation and growth curve. This is shown in Fig. 1. The growth curves are functions of dose, dose rate, temperature, strain, geological origin of natural samples, and numerous other factors. For example, strain applied prior to irradiation decreases the induction period (dose). Also, the colloid formation rate is low at 125C, reaches a maximum near 175C and is negligible at 300C. In several respects damage in synthetic NaCl is different from that in natural crystals.

Recently, radiation induced colloid formation has been studied extensively. First, a computerized iterative procedure has been developed to separate the overlapping F-center and colloid bands. Second, the resulting colloid band spectra have been fitted by a small particle version of Mie theory to obtain information on the average diameter and the total amount of sodium in colloid particles. Proceeding on the basis that this procedure is reliable, one concludes that colloid formation in natural and synthetic rock salt is quite different. In natural salt the colloid concentration increases with dose but the colloid particle diameter remains nearly constant. In synthetic NaCl both the colloid concentration and particle size increase with dose. Other recent measurements show additional differences between natural and synthetic salt. Unirradiated synthetic NaCl contains an absorption band near 6 eV (210 nm) not present in unirradiated natural salt. This suggests numerous possibilities; e.g. impurities in one type of crystal may suppress or enhance radiation damage processes occurring in the other type. The results have been used to evaluate the Jain-Lidiard⁽⁴⁾ theory for radiation induced colloid formation in NaCl. In many respects, the agreement between data and theory is quite satisfactory. However, the theory does not describe all of the observed effects. For example, the theory assumes that F-center diffusion is described by a single activation energy. The data for both synthetic and

natural crystals provides one value at low temperatures and another at high temperatures. Most likely the theory can be modified to be very useful for repository calculations.

The research planned for the immediate future includes numerous studies on radiation damage in natural rock salt. Included are studies on strain applied before and during irradiation, studies on salt from different horizons in the same drill hole (probably ERDA-9) to determine if layer-to-layer variations are important, etc. Some measurements will be undertaken to improve basic understanding, e.g., the study of differences between natural and synthetic salt. Lastly, increased effort will be devoted to other rocks and minerals of interest to the waste disposal program, particularly granite, basalt and shales.

References

1. Radiation Damage Studies on Synthetic NaCl Crystals and Natural Rock Salt for Waste Disposal Applications. K. J. Swyler, R. W. Klaffky and P. W. Levy, Scientific Basis of Nuclear Waste Management, Vol. 1, G. J. McCarthy, editor (Plenum, New York), p. 349.
2. R. W. Klaffky, K. J. Swyler and P. W. Levy, "Radiation Damage Studies on Synthetic NaCl Crystals and Natural Rock Salt for Waste Disposal Applications," Proc. Int. Symp. on Ceramics in Nuclear Waste Management, American Ceramic Soc., Cincinnati, Ohio, April 30 - May 2, 1979 (in press).
3. ONWI Reports: ONWI/SUB/78/E511-01000-2
ONWI/SUB/78/E511-01000-5
ONWI/SUB/78/E511-01000-8
4. U. Jain and A. B. Lidiard, "The Growth of Colloidal Centres in Irradiated Alkali Halides," Phil. Mag. 35, 245 (1977).

Figure Caption

Radiation induced absorption in synthetic rock salt measured during irradiation with 1.5 MeV electrons. Only selected spectra are shown. The F-band appears immediately and at the dose where it reaches saturation the colloid band begins to grow rapidly.

SYNTHETIC NaCl

1.2×10^8 rad/h

163 C