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RADIATION DAMAGE MEASUREMENTS ON
ROCK SALT AND OTHER MINERALS FOR WASTE DISPOSAL APPLICATIONS

QUARTERLY REPORT

FOR THE PERIOD

January 1, 1980 to March 31, 1980

by

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K. J. SWYLER, J. M. LOMAN, L. J. TEUTONICO, G. E. ELGORT AND P. W. LEVY

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BROOKHAVEN NATIONAL LABORATORY

UPTON, NEW YORK 11973

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I. INTRODUCTION

This report covers the activities of a small group at Brookhaven National Laboratory (BNL) studying radiation damage in natural rock salt, synthetic NaCl crystals, granite, basalt and other minerals of interest for radioactive waste disposal applications. The people involved are:

Physics Department:

Paul W. Levy (Principal Investigator)

James M. Loman

(Vacant Position)

Louis J. Teutonico (Temporary Consultant)

Gary Elgort (Graduate Student)

Richard Schwartzman (Graduate Student)

Department of Energy and Environment:

Karl K. Swyler (Transferred to NRC Project, January 30, 1980)

II. PROGRAM OBJECTIVE

Different aspects of radiation damage in both synthetic NaCl crystals and various natural rock salt samples as well as granite, basalt and other minerals which will be important for radioactive waste disposal applications are being investigated. The information obtained is needed for repository site selection, design and engineering, the evaluation of interactions between waste and host rock, risk assessment and other considerations. Most measurements are being made with unique equipment at Brookhaven National Laboratory for making optical and other measurements on samples before, during and after irradiation with 0.5 to 3.0 MeV electrons. The ability to make measurements during irradiation is particularly important. Rock salt and other minerals surrounding emplaced waste will be continuously irradiated and it is essential to determine if important radiation damage processes occur during irradiation which might not be detected in samples studied only after irradiation. Up to the present, in these studies, the principal means of measuring radiation damage is the determination of F-center, i.e. negative ion defect, concentrations, and the concentration and size of sodium metal colloid particles. The formation of these and other defects during irradiation and the annealing of defects and characterization of other processes occurring after irradiation are being studied as a function of dose rate, total dose, sample temperature during irradiation, strain applied prior to and during irradiation, etc. Measurements are being made on synthetic NaCl and natural rock salt samples from different geological locations, including some potential repository sites. The research goal is well defined. Namely it will provide sufficient information, as analytical expressions, graphs, tables, etc., to determine the radiation damage developed in rock salt,

granite, basalt and other minerals surrounding emplaced canisters as a function of radiation levels, time after emplacement, mineral temperature and other parameters. To extrapolate from the laboratory studies to the dose rates, exposure times and other parameters applying to repositories it will be necessary to make a reasonably complete determination of the radiation damage formation and annealing kinetics in the pertinent minerals. Also, it will be necessary to determine if radiation damage in the minerals from different localities is similar. If non-negligible differences are observed a detailed study must be made for each locality under consideration. Almost all current studies are being made on rock salt but other minerals particularly granite and basalt are being phased into the program.

It is now clearly established that radiation damage formation in both natural and synthetic rock salt is strongly dependent on strain. For example, the dose required to induce rapid colloid growth, i.e. the induction period dose, decreases as strain applied prior to irradiation is increased. The strain related effects strongly indicate that the damage formation processes and in particular the colloid nucleation processes are related to the strain induced dislocations. Consequently a temporary theoretical effort has been started to determine which dislocation related effects are important for radiation damage processes and, most importantly, what dislocation interaction are most likely to create nucleation sites for colloid particles. If these preliminary studies indicate that additional theoretical studies will be useful an effort will be made to have them extended.

III. PROGRESS REPORTED PREVIOUSLY

1. Previous Reports

Published Papers:

Swyler, K. J., Klaffky, R. W., and Levy, P. W. Radiation Damage Studies on Synthetic NaCl Crystals and Natural Rock Salt for Waste Disposal Applications. Scientific Basis of Nuclear Waste Management, Vol. 1, G. J. McCarthy, editor (Plenum, New York), p. 349. (This paper is included in ONWI Report No. ONWI/SUB/78/E511-01000-2).

Klaffky, R. W., Swyler, K. J., and Levy, P. W. Radiation Damage Studies on Synthetic NaCl Crystals and Natural Rock Salt for Waste Disposal Applications, II: Ceramics in Nuclear Waste Management, Conf. 790420, Proc. of an Int. Symp., Cincinnati, Ohio, April 30-May 2, 1979, T. D. Chikalla and J. E. Mendel, editors (Tech. Info. Center, U.S. Dept. of Energy, Washington, 1979), pp. 310-314.

Papers Submitted for Publication:

Levy, P. W., Swyler, K. J., Klaffky, R. W. Radiation Induced Color Center and Colloid Formation in Synthetic NaCl and Natural Rock Salt. To be published in Proc. 3rd Europhysics Topical Conf. "Lattice Defects in Ionic Crystals" (J. de Physique, Supplement), Canterbury, England, September 1979.

Swyler, K. J., Klaffky, R. W., and Levy, P. W. Recent Studies on Radiation Induced Color Centers and Colloid Formation in Synthetic NaCl and Natural Rock Salt for Waste Disposal Applications. Proc. Materials Res. Soc. 2nd Symp. on the Scientific Basis of Nuclear Waste Management, Cambridge, Mass., November 1979.

Quarterly Reports Submitted to ONWI:

<u>ONWI Report No.</u>	<u>BNL Report No.</u>	<u>Period</u>
ONWI/SUB/78/E511-01000-2	25833	Oct.-Dec. 1978
ONWI/SUB/78/E511-01000-5	26028	Jan.-Mar. 1979
ONWI/SUB/78/E511-01000-8	26455	Apr.-June 1979
ONWI/SUB/78/E511-01000-11	26931	July-Sept. 1979
ONWI/SUB/78/E511-01000-14	27382	Oct.-Dec. 1979

2. Previous Results

As mentioned above, radiation damage in natural and synthetic rock salt is being studied by measuring optical absorption bands associated with lattice defects and colloid particles. Most of the measurements are being made with unique equipment at Brookhaven National Laboratory for making optical absorption, radioluminescence, and other measurements on samples before, during and after irradiation with 0.5 to 3.0 MeV electrons. The samples are irradiated in a temperature controlled chamber. Individual absorption spectra are recorded as often as every 40 sec and, to date, have been confined to the 3.1 to 1.55 eV (400 to 800 nm) region. Most of the completed studies have been made on the F-center and colloid absorption bands. The F-band absorption is a measure of the Cl^- negative ion defects present in the lattice. Colloid band measurements provide information on the concentration and size of the radiation induced metallic sodium particles. Curves of F-center and colloid band absorption vs. irradiation time--and time after irradiation--are prepared using a variety of techniques for resolving the recorded spectra into individual bands. These curves, usually called growth curves, have been obtained for a variety of different experimental conditions. The parameters varied include temperature, dose, dose rate, and strain applied prior to irradiation. A large fraction of the completed studies have been made on synthetic melt-grown NaCl crystals. An increasing number of measurements are being made on natural rock salt samples from different geological localities, including some localities under consideration as repository sites.

The completed measurements indicate that abundant colloid formation occurs in both natural and synthetic crystals. Unless the crystals are heavily strained, the F-center and colloid absorption vs. irradiation time curves, i.e. the growth curves, follow a consistent pattern. The F-band increases monotonically to a well-defined plateau while the colloid absorption is zero or negligible and, in the dose region where the F-center plateau is reached, the colloid formation rate increases from a low to a high value. In other words, the colloid formation described by a classical nucleation and growth curve containing an obvious induction period. The induction period is reduced by straining the samples prior to irradiation or by cycles of irradiation, annealing and reirradiation. A 10 percent strain applied prior to

irradiation reduces the induction period to zero. The F-center growth rate and saturation level and the colloid growth are functions of irradiation temperature, dose, dose rate and other parameters.

Both the F-center formation and colloid particle formation are strong functions of temperature. As the temperature increases the F-center saturation level decreases and is near zero at 300C. In contrast, the colloid particle formation rate is low or zero near 100C increases to a maximum in the 150 or 175C range and has decreased to a negligible level at 300C.

Results obtained to date on synthetic NaCl are in qualitative agreement with previous publications. They indicate that rock salt adjacent to emplaced canisters will develop copious radiation damage in the form of colloid metal sodium particles.¹⁻⁴ The presence of the colloid metal sodium will be particularly important in waste-host rock interactions, under both "wet" and "dry" conditions. Also, the mechanical properties will be altered by radiation.³ As the radiation increases the rock salt yield point increases and the rock ultimately becomes quite brittle.

Most importantly, it is becoming increasingly apparent that radiation damage formation in natural rock salt differs from that in synthetic NaCl in several respects that are important for waste disposal applications. For example, at room temperature F-center, i.e. negative ion vacancy, formation followed a square root of irradiation time behavior in natural rock salt which is not observed with synthetic crystals. In addition, the colloid formation processes in synthetic NaCl are different from those in natural crystals. At doses where both types are exhibiting rapid colloid formation, the colloids in synthetic NaCl appear to be nucleating at a small diameter and increasing in size as the dose increases while, in contrast, in natural rock salt the total number of colloid particles is increasing but all of the observed particles appear to have approximately the same diameter. Synthetic NaCl crystals strained roughly 10 percent before irradiation exhibit colloid growth characteristics similar to those observed with natural samples. Strain applied prior to irradiation has a profound effect on colloid formation in natural rock salt from AEC-8. The growth curve changes in a way which is usually interpreted as indicating that the induction period growth rate has increased greatly or that the induction period has been eliminated. The extrapolation of this data to repository doses indicates that rock salt strained 11 percent will contain 2 to 3 times the colloid concentration induced in unstrained rock salt.

Measurement on unirradiated synthetic and natural rock salt indicate that there are differences in the absorption spectra near 200 nm. Synthetic NaCl is moderately strongly absorbing at this wavelength whereas natural crystals are not. This suggests that synthetic crystals contain a defect or impurity, e.g. OH⁻, not present in natural samples which strongly effects the damage formation process. Further evidence to conclude that the 200 nm absorption is due to OH⁻ is obtained by heating natural crystals in air. AEC-8 natural crystals, which originally did not contain a 200 nm band, developed a strong band when heated for 60 hours at 600C.

In addition to the experimental studies described above, theoretical studies are under way to determine if dislocations are an important factor in the colloid nucleation process. Anisotropic elasticity theory is being employed to calculate dislocation stress fields and related properties, e.g. the rate of flow of defects to dislocations, to determine if any of the more common dislocation configurations are likely sites for colloid nucleation.

IV. RESULTS FOR THE PERIOD JANUARY 1, 1980 TO MARCH 31, 1980.

1. Introduction.

This period included studies in two areas which probably will not produce reportable results for three or six months. These are: first, thermoluminescence studies on Climax Stock granite and, second, the preparation of samples to determine if Small Angle Neutron Scattering (SANS) techniques can be used to study the radiation induced colloids in rock salt. It is hoped that the SANS measurements can be used to study samples with colloids that are either too small or large or in concentrations too high to study with the currently used optical techniques. Reported this quarter are results in three areas. First, F-center annealing studies in both natural and synthetic rock salt. Second, the generation of colloids in samples that are subjected to a cycle of irradiation near room temperature and heating at temperatures which generate colloids. Third, additional theoretical studies to determine if and how dislocations are nucleation sites for colloids.

2. F-Center Annealing in Natural and Synthetic Rock Salt.

The thermal annealing of radiation induced F-centers, i.e. vacancies, in rock salt can be studied in a convenient manner with the equipment for making optical measurements on samples during and after irradiation. This can be done in two ways. A sample can be irradiated at a low temperature and the thermally induced decay of the F-centers determined at various temperatures as the sample is heated. The samples temperature is raised to a new value, allowed to come to equilibrium, and the F-center concentration measured as a function of time. Alternatively, the sample may be irradiated at a fixed temperature and the decay monitored after the irradiation is terminated. Sometimes, especially if the beam current is high and is contributing appreciable heat to the irradiation chamber, reliable data is obtained only after the sample has reached a fixed temperature. When these procedures are used it is possible that short-lived components may not be observed. In both cases, at a given temperature, a semi-log plot of F, or F-center concentration vs. time will contain one linear component for each observed decay process. Usually, the decay occurring immediately after irradiation at a fixed temperature, in both natural and synthetic rock salt, is described by a single decaying exponential component. Also the decay rate λ , as it occurs in the usual equation $dF/dt = -\lambda F$, is the slope of the curve on the semi-log plot.

Fig. 1:-An Arrhenius plot showing the rate of decay of radiation induced F-centers in natural and synthetic rock salt at various temperatures. The plot indicates that the activation energy for thermal decay, approximately 1.0 eV, at least for the most rapidly decaying component, is the same for both materials but the pre-exponential factor for synthetic salt is larger than that for the AEC-8 natural salt.

An Arrhenius plot showing measured F-center decay rates at different temperatures for both synthetic rock salt and natural rock salt from Drill Hole AEC-8 in southeast NM is shown in Fig. 1. This plot shows a number of results. In the temperature range covered, the natural rock salt decay appears to be described by a single component. However, in this range the synthetic salt F-center decay could contain two components. The second component would apply only to a small fraction of centers present and will be assumed negligible in the discussion that follows.

If only the clearly observable decay components are considered, Fig. 1 indicates that both processes depend on the same activation energy, namely $E \approx 1.0$ eV, but that the decay rate for synthetic salt is approximately 1.5 times that of natural salt. If it is assumed that the process responsible for F-center decay is F-center diffusion, the data in Fig. 1 indicates that the activation energy for diffusion is the same for natural and synthetic rock salt. However, the preexponential factors are different. This could be attributed to a difference in hopping frequencies, attempt-to-escape frequency or even differences in recombination cross sections at the site of the F-center annihilation site.

The data tends to rule-out the possibility that there are different mobile species contributing to F-center decay in natural and synthetic salt. If different species were involved it is unlikely that closely similar activation energies would be obtained.

3. Colloid Formation Induced by Heating Previously Irradiated Natural Rock Salt.

Prior reports⁵ have included cursory descriptions of colloid formation in rock salt samples that were irradiated at room temperature and subsequently heated to temperatures in the 150 to 200C range. There are numerous reasons to study this process in detail. For example, in a radioactive waste repository it is likely that salt will be subjected to one or more cycles of irradiation followed by heating. Such a cycle could occur soon after canister emplacement or at a much longer time. Also, colloid formation during irradiation can be regarded as the superposition of two processes; those associated with the presence of radiation and one or more thermally activated processes operating on irradiated salt. In any case, the study of heat induced colloid formation in previously irradiated rock salt should provide appreciable fundamental information on the basic colloid formation process.

To provide the information needed for the planning of detailed measurements, samples of natural rock salt from Drill Hole AEC-9 were irradiated with CO-60 gamma ray to a total dose of 5×10^7 rad at approximately 9C. One sample had been uniaxially strained by a 2.2 percent compression prior to irradiation, one was similarly strained 6.7 percent after irradiation and a third was not strained. All samples almost certainly contained some "natural" strain. The optical absorption spectra of these samples, measured after irradiation and after different heating sequences, are shown in Fig. 2. All three samples exhibit prominent F-center bands near 2.7 eV, pronounced M-center bands near 1.7 eV and obvious V-region absorption--particularly in the 5 to 6.5 eV region.

Fig. 2a:-Radiation induced absorption spectra of natural rock salt samples from Drill Hole AEC-9 after a CO-60 irradiation of 5×10^7 rad at 9C. All absorption measurements were made at 22C.

Fig. 2b:-The absorption spectra of the samples used for Fig. 2a, after heating for 2 hours at 150C and 1 hour at 175C.

Fig. 2c:-The absorption spectra of the same samples, after heating for a total of 2 hours at 150C, 2.5 hours at 175C and 1 hour at 200C.

Fig. 2d:-The absorption spectra after heating for a total of 2 hours at 150C, 2.5 hours at 175C and 2 hours at 200C.

(The V-region absorption lies to the high energy side of the F-band and is generally attributed to electronic hole centers.) The sample strained prior to irradiation contains the most intense absorption, as would be expected from previous studies. It is likely that the 6.7 percent strain applied after irradiation reduced the absorption (in this sample) but in this series of measurements the strain induced changes were not monitored. The spectra in Fig. 2a do not contain evidence for colloid absorption near 2 eV.

The absorption spectra measured after the samples had been heated in air for 2 hours at 150C and then for an additional 1 hour at 175C is shown in Fig. 2b. Heating has produced a large reduction in F-centers and the V-region absorption in the 5 to 6.5 eV region. Absorption near 2.2 eV attributable to colloid bands is clearly observed in the unstrained sample and in the sample strained prior to irradiation. Only slight evidence for colloid absorption is present in the third spectrum. Also, the M bands appear to have increased but this may be due to the superposition of M and colloid absorption at the M-band peak. However, spectra measured after heat treatment of 1 and 2 hours at 150C showed that the M band had increased.

The spectra obtained after the samples had been heated for 2 hours at 150C, 2.5 hours at 175C and 1 hour at 200C are shown in Fig. 2c. The previously observed trends have continued, except that M-band absorption is now absent. However, the sample strained after irradiation exhibits appreciably less colloid absorption than the other. In other words, straining after irradiation appears to have reduced the thermally induced colloid formation.

The absorption present after heating 2 hours at 150C, 2.5 hours at 175C and 2 hours at 200C are shown in Fig. 2d. Very little F-center absorption is present; there is no evidence for M-center absorption, but there is moderate V-region and strong colloid band absorption. However, the sample strained after irradiation contains roughly one-half the colloid absorption of the other samples.

From the data included in Fig. 2 one can conclude that, in this series of measurements, straining after irradiation has reduced the tendency to form colloids. However, such a conclusion may be premature. For example, these samples were not subjected to a radiation other than the initial one. Considering that samples strained prior to irradiation exhibit increased tendencies to form colloids during irradiation it is possible that additional radiation will undo or reverse the tendencies described in Fig. 2. Also, it is entirely possible that the samples strained after irradiation contains a larger number of nucleation sites than the others. Thus, an appreciable number of colloid particles may have been formed which are too small to be observed by optical absorption techniques. (Previous measurements on both synthetic and natural salt indicate that colloid particles are not observed until they reach a critical size.)

These measurements lead to several conclusions. First, as indicated by earlier measurements, heating of previously irradiated salt leads to colloid formation. Second, the strain present strongly influences the colloid formation. Third, radiation induced and thermally activated changes in the F-center and colloid absorption are accompanied by V-region absorption changes. Changes in the V-region hole center absorption are known--and are

expected--from previous studies on color centers, to accompany changes in the F-center and colloid electron center absorption. It may be possible to utilize the V-region absorption changes to determine the fraction of F-centers that undergo electron-hole recombination without contributing to colloid formation.

Additional studies of the type described above are now under way. However, at least for the present, the emphasis will be on studies to determine the mechanisms and kinetic parameters, such as the activation energy, or energies, for thermally induced colloid formation in previously irradiated natural and synthetic rock salt.

4. Theoretical Studies on the Role of Dislocations in the Nucleation of Colloidal Sodium Particles in Irradiated Rock Salt.

As explained in previous reports, theoretical calculations are being made to determine which dislocations in NaCl are the most likely sites for nucleating the sodium metal colloid particles formed during irradiation. Although detailed in previous reports, the prior calculations will be sketched here.

The calculation of the kinetics of point defect migration to dislocations proceeds in three steps: First, a determination of the form and magnitude of each dislocation-point defect interaction. Second, a calculation of the lines of flow along which the defects move to dislocations. Third, the computation of the number of defects that arrive at dislocations as a function of time. In this manner one determines which dislocation-point defect interactions are the most probable for point defect accumulation and, in turn, for the nucleation of colloidal sodium particles.

As it is expected that the dislocation-point defect interactions will be greatest for edge dislocations, the initial calculations are being carried out for the two most likely edge dislocations. First is the edge dislocation in the primary NaCl slip system. It lies along the $\langle 001 \rangle$ direction, glides on the $\{110\}$ plane, and produces slip in the $\langle 1\bar{1}0 \rangle$ direction. The second to be studied is the edge dislocation resulting from interaction of dislocations on obliquely intersecting $\{110\}$ slip planes. This dislocation lies along the $\langle 111 \rangle$ direction, has a slip vector parallel to $\langle 1\bar{1}0 \rangle$ and could glide only on the $\{11\bar{2}\}$ plane. Since glide on $\{11\bar{2}\}$ is not commonly observed in NaCl, this dislocation is thought to be immobile.

The first two steps in the computations for the $\langle 001 \rangle$ dislocations are completed and have been described previously. The third step in the calculations is described in this report. Also included are some results for defect migration to $\langle 001 \rangle$ edge dislocations.

As described in previous reports, there is an interaction potential between a dislocation and a point defect.⁶ Because of this interaction the point defect experiences a force, proportional to the gradient of the potential, which causes the defect to flow to, or be repulsed from, the dislocation. This flow, called drift flow,⁷ is superimposed on any defect migration which occurs by diffusion processes alone. When both processes are operating the defect migration kinetics depend upon the precise nature of the interaction potential and any concentration gradients which are present. The equation which describes the migration when both drift flow

and diffusion occur is

$$\frac{1}{D_f} \frac{\partial c}{\partial t} = \nabla^2 c + \frac{\nabla(c \nabla U)}{kT} \quad (1)$$

where c is the defect concentration, D_f the diffusion coefficient at temperature, T , k Boltzmann's constant, and U the interaction energy between the dislocation and the point defect. If drift flow predominates the $\nabla^2 c$ term can be put equal to zero. This is the case that is being considered in these calculations. Physically, this corresponds to cases where the temperature is so low the thermally activated processes do not contribute to the defect migration. In other words, only the dislocation-defect interaction is contributing to defect motion. In this case the center of the dislocation becomes an ideal sink. Mathematically, this corresponds to putting $c = 0$ at this point. After the dislocation-defect interaction has been operating for some time a well-defined area surrounding the dislocation will have been depleted of defects. This area is called the depletion zone. The principal object of these calculations is to determine the number of defects which have migrated to the dislocation as a function of time. This is equivalent to determining the depletion zone area as a function of time.

Specifically, as described previously, for the $\langle 001 \rangle$ edge dislocation under consideration, the interaction energy is given by

$$U = \frac{A}{R} \frac{\sin \phi (1 + \alpha \cos^2 \phi)}{(1 - \alpha \cos^2 2\phi)} \quad (2)$$

The quantities in this equation were given in the previous reports. When this potential is inserted in eq. (1) and the normal diffusion term $\nabla^2 c$ is put equal to zero, one obtains the equation

$$\lambda \frac{\partial c}{\partial \rho} + \mu \frac{\partial c}{\partial \phi} + \tau \frac{\partial c}{\partial t} = \nu c \quad (3)$$

where

$$\rho = R/b \quad (4)$$

and τ is a temperature dependent time parameter, given by

$$\tau = kTb^3/D_f A. \quad (5)$$

The coefficients λ , μ , ν are all functions of the polar coordinates ρ and ϕ and the parameter α , where, as described in previous reports, $\alpha = H/(4+H)$ and H is the previously described anisotropy parameter. The coefficients in (3) are given by

$$\begin{aligned} \lambda(\rho, \phi, \alpha) &= \frac{\sin \phi (1 + \alpha x)}{\rho^2 (1 - \alpha x^2)} \\ \mu(\rho, \phi, \alpha) &= \frac{-\cos \phi f(x, \alpha)}{\rho^3 (1 - \alpha x^2)^2} \\ \nu(\rho, \phi, \alpha) &= \frac{4\alpha \sin \phi g(x, \alpha)}{\rho^3 (1 - \alpha x^2)^3} \end{aligned} \quad (6)$$

where

$$x \equiv \cos 2\phi$$

$$f(x, \alpha) \equiv (1-2\alpha) - \alpha x + \alpha x^2(3-2\alpha) + \alpha^2 x^3 \quad (7)$$

$$g(x, \alpha) \equiv 1-2x(2-3\alpha) - 6x^2(1-\alpha) - \alpha x^3(2+x)(2-\alpha).$$

The solution of eq. (3) is obtained, in a straightforward manner, using the standard techniques for solving first-order partial differential equations.⁸ The curves represented by the solutions of the subsidiary equations

$$\frac{d\rho}{\lambda} = \frac{d\phi}{\mu} = \frac{d\tau}{\tau} = \frac{dc}{vc} \quad (8)$$

are called characteristic curves of the partial differential equation (3). If these are denoted by $u_i(\rho, \phi, t) = k_i(\text{const.})$, $i = 1, 2, 3$, then $f(u_1, u_2, u_3) = 0$ where f is an arbitrary function represents the general solution of eq. (3). A particular solution is obtained by applying appropriate initial and boundary conditions to this general solution. In the specific problem being considered, the initial concentration of defects is taken to be uniform, i.e. $c = c_0(\text{const.})$ at $t = 0$, except at the dislocation which is located at $\rho = 0$, where $c = 0$ at all times. This last condition represents the sink for defects described above. At time t the concentration of defects is zero within an area bounded by the curve

$$\rho = \left(\frac{t}{\tau} \right)^{1/3} \frac{\gamma_1(\phi, \alpha)}{[\gamma_3(\phi, \alpha)]^{1/3}} \quad (9)$$

In this expression the functions γ_1, γ_3 are given by

$$\gamma_1(\phi, \alpha) = \exp \left\{ \int \frac{(1-\alpha x^2)(1+\alpha x)}{2(x+1)f(x, \alpha)} dx \right\} \quad (10)$$

$$\gamma_3(\phi, \alpha) = - \int \frac{[\gamma_1(\phi, \alpha)]^3 (1-\alpha x^2)^2}{\cos \phi f(x, \alpha)} d\phi.$$

The curve given by eq. (9) represents the boundary of the depletion zone. It expands with time. Outside the depletion zone the concentration of defects is given by

$$c = c_0 \exp\{\gamma_2(\phi, \alpha)\} \quad (11)$$

where

$$\gamma_2(\phi, \alpha) = 2\alpha \int \frac{g(x, \alpha) dx}{(x+1)(1-\alpha x^2)f(x, \alpha)} \quad (12)$$

Thus the solution of eq. (3) is not continuous across the depletion zone boundary.

The three functions $\gamma_1, \gamma_2, \gamma_3$ all depend on the anisotropy parameter H and the parameter α , where $\alpha = H/(4+H)$. Hence the form of the depletion zone boundary (9), and the concentration of defects outside the depletion zone, i.e. eq. (11), depend on the value of H . A typical depletion zone boundary is shown in Fig. 3 for an H value of -0.85 . More specifically Fig. 1 applies to unirradiated NaCl at room temperature.

It should be pointed out that the curves given by $\rho = \text{constant} \times \gamma_1(\phi, \alpha)$ are the flow lines along which the defects move to dislocations. These curves, which were previously obtained independently as the orthogonal trajectories of the interaction potential (2), also represent a separate family of characteristics of the differential equation (3).

Once the depletion zone boundary is known, one can calculate the number of defects which arrive at the dislocation as a function of time. The defect flow $N(t)$ is given by the area enclosed by this boundary weighted by the angular distribution function $c_0 \exp[\gamma_2(\phi, \alpha)]$, or

$$N(t) = c_0 b^2 \int_{-\pi/2}^{\pi/2} e^{\gamma_2} \rho^2 d\phi \quad (13)$$

where ρ is given by eq. (9). The flow of defects to dislocations $N(t)$ can be written as

$$N(t) = c_0 b^2 (t/\tau)^{2/3} \psi(H) \quad (14)$$

where the dependence on the anisotropy parameter H is contained in the function

$$\psi(H) = \int_{-\pi/2}^{\pi/2} \frac{\exp[\gamma_2(\phi, \alpha)] [\gamma_1(\phi, \alpha)]^2}{[\gamma_3(\phi, \alpha)]^{2/3}} d\phi. \quad (15)$$

In unirradiated NaCl and in the temperature range 24°C – 700°C , the reported values of the elastic constants⁹ lead to values of H between -0.85 and $+0.85$. It is important to note that the effects of irradiation on the values of H have not been determined. If irradiation has the same effect on elastic constants as going to very low temperatures, then H values less than -1 could be obtained with irradiated NaCl. Inasmuch as the character of the flow lines changes radically at $H = -1$, this could lead to significant changes in the dislocation related properties, e.g. the depletion zone boundaries, as the irradiation progresses.

To determine the depletion zone boundary and the number of defects that accumulate at the dislocation it is necessary to evaluate the integrals $\gamma_1, \gamma_2, \gamma_3$ as functions of H . The function γ_1 has already been evaluated analytically in obtaining the flow lines. As described in the previous report, the flow lines are closed curves passing through the center of the dislocation for H values in the range $-1 \leq H < H_0 \approx 3.58$. For values of H outside this interval there are certain flow lines which diverge to infinity, in addition to closed loops passing through the center of the dislocation (see Figs. 5, 6 of previous report).

Fig. 3:-The area, or depletion zone, normal to a $\langle 001 \rangle$ edge dislocation in NaCl from which defects have been removed, by migrating to the dislocation, by the interaction between the strain field of the dislocation and the defect. These curves show the boundaries for various values of the anisotropy constant H . The value $H = -0.85$ corresponds to unirradiated NaCl at room temperature. The axes lie along $\langle 110 \rangle$ directions.

To determine the depletion zone boundary, eq. (9), one must evaluate both of the functions γ_1 and γ_3 . At the present time this has been done only for values of H in the interval $-1 < H < H_0$. For H values outside of this interval infinities in the flow line solutions create computational difficulties which have not been overcome. However, the variation in depletion zone boundary which occurs in the range where computations have been completed is illustrated in Fig. 3. This shows the depletion zone boundaries for $H = -0.85, 0$ and 3.50 .

The parameter τ , defined by (5), describes the depletion zone boundary at different times. It depends on both the strength of the dislocation-defect interaction, A , and the defect diffusion coefficient, D_f . As is well known, D_f varies exponentially with $1/T$. Thus, over the entire range of interest τ will be a decreasing function of temperature.

Both the depletion zone boundary and the number of defects arriving at the dislocation are functions of time, or, more precisely, the reduced time t/τ . Since τ decreases with temperature the time required for the depletion zone to expand to a given dimension, and analogously for a specified number of defects to reach the dislocation, will decrease with increasing temperature. A computation of the movement of vacancies to the NaCl $\langle 001 \rangle$ edge dislocation, as indicated by the outward movement of the depletion zone, is illustrated in Figs. 4, 5 for crystals at 24°C and 150°C . Using the elastic constants of Hart,⁹ a misfit parameter $\epsilon = 0.02$, and the diffusion coefficient used by Jain and Lidiard⁴

$$D_f = 0.1 \exp\{-0.8 \text{ eV}/kT\} \text{ cm}^2/\text{sec}, \quad (20)$$

one finds that the τ values at 24°C and 150°C are 16.88 seconds and 1.820×10^{-3} seconds, respectively. The H values at these temperatures are -0.85 and -0.62 . At both temperatures, the depletion zone boundaries are shown for reduced times, t/τ , of $1/3 \times 10^4$, $1/3 \times 10^5$, $1/3 \times 10^6$. Obviously, the depletion process is strongly temperature dependent. For the depletion zone to reach a dimension of 100 Burgers vectors (~ 71 lattice constants) at room temperature requires more than two months. At 150°C the corresponding time is approximately 10 minutes.

It is interesting that these times correspond well to observed values. When a sample is irradiated at room temperature and then stored in the dark, colloids are observed to form in 2 to 6 months. In contrast, samples irradiated at 150°C usually form colloids during irradiation.

Planned for the next quarter are the completion of the drift flow solution for the $\langle 001 \rangle$ edge dislocation in rock salt and a start will be made on the corresponding analysis for the immobile $\langle 111 \rangle$ edge dislocation. Once the two analyses are completed one can compare, first, the relative strengths of the dislocation-defect interactions and, second, the relative times required for a specific number of defects to accumulate at each type of dislocation.

The results reported here indicate that the $\langle 001 \rangle$ edge dislocation is a viable site for colloid particle nucleation in NaCl. The planned work should indicate if the $\langle 111 \rangle$ edge dislocation is an equally good, more likely, or less likely site for colloid nucleation.

SYNTHETIC ROCK SALT 24°C
 DEPLETION ZONE AS FUNCTION OF TIME

Fig.4:-Depletion zone boundaries, normal to a $\langle 001 \rangle$ edge dislocation in NaCl at room temperature, as a function of time after defects have been introduced into the lattice. The axes lie along $\langle 110 \rangle$ directions. The units are burgers vectors; $100 \text{ b} = \sqrt{2}/2 \times 100 \text{ lattice constants}$.

SYNTHETIC ROCK SALT 150°C
DEPLETION ZONE AS FUNCTION OF TIME

Fig.5:-Same as Fig. 4 except that the crystal is at 150C instead of 24C. It is interesting that the computed times agree well with the times associated with the appearance of colloids in salt irradiated at roughly 24 and 150C.

V. PROBLEMS

The only major problem continues to be personnel. The staff member who left in February of 1979 was replaced by Dr. J. M. Loman in February 1980. An offer has been made to a possible replacement for Dr. K. J. Swyler who transferred in February to a permanent Brookhaven National Laboratory group that will provide technical assistance to the Nuclear Regulatory Commission. Efforts are being made to recruit additional staff.

VI. ACTIVITIES PLANNED FOR THE NEXT QUARTER

1. Measurements on natural rock salt samples from different geological localities, and from different horizons at certain localities, will continue to be the major activity. This will include additional efforts to determine how strain applied prior to irradiation influences the radiation induced defect formation.

2. Studies on radiation effects in granite, basalt, and related minerals will be continued. Emphasis will be on thermoluminescence studies of granite from the Climax Stock at the Nevada Test Site.

3. Theoretical studies on the role of dislocations in the nucleation of radiation induced colloid particles will continue, subject to the availability of funds.

4. Analysis of previously acquired data and the preparation of papers will continue.

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