NEW KINETIC MODEL CALCULATIONS
OF COLLOID GROWTH IN NaCl

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ABSTRACT

The formation of radiation damage in NaCl has generated interest because of the relevance of this damage to the disposal of radioactive waste in rock salt formations. In order to estimate the long-term behaviour of rock salt under irradiation, an accurate theory describing the major processes of radiation damage in NaCl is required. In 1977, Jain and Lidiard presented a rather simple model which, until now, has served as a means for long-term predictions of near field radiation damage processes in rock salt repositories. Although the model of Jain and Lidiard describes many experimental results on colloid growth in NaCl rather well, it neglects the colloid nucleation stage and the effect of impurities on colloid growth. The model presented in this paper is an extended version of the Jain-Lidiard model; its extensions comprise the effect of impurities and the colloid nucleation stage on the formation of radiation damage. The extended model has been tested against various experimental data obtained from the literature and accounts for several well known aspects of radiation damage in alkali halides which were not covered by the original Jain-Lidiard model. Using this extended model, we have calculated the amounts of NaCl that can be converted into metallic Na and molecular Cl_2 for various Dutch concepts for reprocessed high-level radioactive waste (HLW) repositories. The theory predicts that the concentration of these defect aggregates in the rock salt, even very close to the HLW containers will be limited to a few mole %.

1. INTRODUCTION

Alkali halides are known to be very susceptible to radiation damage in the halogen sublattice when exposed to ionizing radiation. The primary defects are F and H centers which can be considered as halogen vacancies filled with an electron and interstitial neutral halogens, respectively. Due to recombination, these primary defects are rather unstable in the temperature regime of a repository (well above room temperature) and their concentrations will always be rather low (typical steady state concentrations are 10^{-3} mole % for the F centers and 10^{-10} mole % for the H centers). However, by a process of self trapping and trapping by dislocations and impurities, F and H centers can build up stable aggregates consisting of colloidal Na and molecular Cl_2, respectively. Laboratory experiments have revealed that the concentration of these forms of radiation damage can reach values of several mol %[Soppe et al, 1994, Jenks and Bopp, 1974 and 1977, Jenks et al., 1975, Groote and Weerkamp, 1990].

One of the risks that has to be investigated very thoroughly in respect with the disposal of nuclear waste in a rock salt repository is the possibility of a sudden back reaction[Den Hartog et al., 1992] between the Na colloids and the molecular Cl_2. Such a back reaction would take place if the concentration of these defect aggregates exceeds a certain percolation threshold. Since all significant radiation damage is built-up in the Cl (FCC) sublattice, this percolation limit is assumed to be 12%[Stauffer, 1985]. By its potentially explosive character[Den Hartog et al., 1992], this back reaction could lead to a serious deterioration of the near field integrity of the repository. Laboratory experiments, unfortunately, can provide only limited information on the radiation damage in a real repository. In order to achieve the same total radiation dose as in a repository, laboratory experiments have to be performed at dose rates which are several orders of magnitude larger than in reality. This gap between experiment and reality has urged the development of theoretical models for the build-up of radiation damage in salt.

In 1985 Bergsma, Helmholdt and Heijboer[Bergsma et al., 1985] presented the first calculations of colloid formation in a rock salt nuclear waste repository by using a kinetic rate reaction model. This model was based on a theory developed by Jain and Lidiard[Jain and Lidiard, 1977] describing the kinetics of radiation induced colloid growth in NaCl. The theory of Jain and Lidiard is very useful for understanding the high dose colloid formation in pure NaCl but it can not explain properly the effects of enlarged concentrations of impurities and

dislocations on colloid growth. Further, the theory over-simplifies the initial stage of colloid growth. In order to overcome these deficiencies, several improvements of the theory have been suggested recently. Groote and Weerkamp, 1990, presented a model, based on the Jain-Lidiard theory but with extensions with respect to trapping of H centers by impurities and nucleation of colloids. According to the model of Groote and Weerkamp, which was recently presented in a slightly revised form by Seinen.[Seinen et al., 1992], the effect of impurities on the colloid growth is very modest. However, some experiments reported by the same authors, show a much more pronounced effect of certain impurities. In order to be sure that for repository calculations the effect of impurities, which are inherent to natural salt, is not underestimated we have developed a new model based on the Jain-Lidiard theory. This model, which is described in detail elsewhere[Soppe, 1993], is rather similar to the Groote-Weerkamp model but differs in some important details. As a result of these modifications, the model provides a good explanation of most of the experimentally observed impurity-effects on the growth of colloids. In the next sections we will apply this model in order to calculate the formation of radiation damage in NaCl in general, and in rock salt for some typical HLW storage strategies.

2. MODEL FOR BUILD-UP OF RADIATION DAMAGE IN NACL

The primary defects which are created in NaCl under irradiation are F and H centers. These complementary defects result from radiation-less decay of excitons[N. Itoh, 1982]. An F center can be considered as an electron trapped at a Cl vacancy position. The H center is basically a molecular ion (Cl_2^-) located on a Cl lattice site, interacting with the two neighbouring Cl^- ions along a <110> crystal direction[Townsend and Kelly, 1973]. After their formation, the F and H centers are assumed to participate in the following reaction mechanisms:

i) Trapping of F centers and colloid growth. By a mechanism of self-trapping, F centers may precipitate into F_2 (=M), F_3 (=R) and F_4 (=N) centers. The N centers are considered as seeds for colloid growth by homogeneous nucleation. In addition, a certain amount of seeds for heterogeneous colloid growth will be assumed. The aggregation of F centers into colloids is assumed to occur via random walk bulk lattice diffusion.

ii) Trapping of H centers by impurities. This process leads to the formation of S_1, S_2 and S_3 complexes, which are combinations of an impurity with respectively one, two and three H centers. An S_3 center is considered to be the beginning of a new dislocation loop.

iii) Dislocation loop growth and formation of molecular Cl_2 centers. According to Hobbs et al., 1973, two H centers near a dislocation will form a molecular Cl_2 center by 'digging out' a NaCl molecule from its regular position. This NaCl molecule moves to the extra plane of the dislocation, giving rise to growth of perfect dislocation loops. Dislocation lines act as unsaturable sinks for both F centers and H centers but the elastic interaction with vacancy defects is smaller than with interstitial defects. As a matter of fact, this difference in elastic interaction is the driving force for colloid growth. Due to this difference, namely, the drain of F centers to dislocations is smaller than that of H centers, which allows for the formation of stable Cl_2 centers near dislocations. The resulting excess of F centers elsewhere in the crystal forms the basis for colloids. After the colloid nucleation stage, the concentration of molecular Cl_2 is practically equal to the colloid concentration.

iv) Recombination. Free F and H centers, within a certain radius, will recombine. In addition, it is assumed that F centers may recombine with molecular Cl_2 centers and that H centers can recombine with F centers which are thermally emitted from colloids. The first recombination reaction introduces a strong dose rate dependency of the colloid growth; the latter two reactions determine that at higher temperatures no colloid formation will take place.

These mechanisms lead to the following rate equations:

$$
\begin{aligned}
\frac{dc_F}{dt} = {} & K + 2K_M^{(e)}c_M - 2K_F c_F c_F - K_F c_F c_M - K_F c_F c_R - K_F c_F c_N \\
& + K_H c_H c_M - 4\pi r_c C_c D_F\left(c_F - c_F^{(e)}\right) \\
& - K_2 c_F c_H - z_F \rho_d D_F c_F - \gamma D_F c_F c_{Cl_2}
\end{aligned}
\tag{1}
$$

The first term on the right hand describes the production rate of F and H centers due to

irradiation. See also Tables I and II. The next five terms describe the disintegration of an M center and the trapping of F centers by F-, M-, R- and N centers respectively. The seventh term accounts for the annihilation reaction of an H center at an M center. It is assumed that the loss of F centers to colloids and the evaporation of F centers from colloids are processes controlled by bulk diffusion. This leads to the term $4\pi r_c D_F C_c (c_F - c_F^{(e)})$. $K_2 c_F c_H$ is the back reaction between F centers and H centers. $z_F \rho_d D_F c_F$ is the loss of F centers to dislocation loops and the last term describes the back reaction between F centers and Cl_2 molecules.

Having described all the terms in the kinetic rate reaction of the F center concentration, the other rate reactions are more or less self explaining and are given with only a few comments.

$$\frac{dc_M}{dt} = K_F c_F c_F - K_H c_H c_M - K_M^{(e)} c_M - K_R c_F c_M \qquad (2)$$

At low temperatures (*i.e.* below room temperature) M center growth is very slow. Since the growth rate of R centers ($K_R c_F c_M$) is even smaller and the recombination with H centers is in general a second order process, we may assume that in this temperature regime: $K_F c_F c_F \approx K_M^{(e)} c_M$ or:

$$c_M = \frac{K_F}{K_M^{(e)}} c_F^2 \quad . \qquad (3)$$

$$\frac{dc_N}{dt} = K_R c_F c_R - K_F c_F c_N \qquad (4)$$

$$\frac{dc_A}{dt} = K_F c_F c_N + 4\pi r_c C_c (D_F c_F - D_H c_H - D_F c_F^{(e)}) \qquad (5)$$

$$\frac{dc_H}{dt} = K - K_H c_{S_0} c_H - K_H c_{S_1} c_H - K_H c_{S_2} c_H$$
$$+ K_H c_{S_1} c_{H,S_1}^{(e)} + K_H c_{S_2} c_{H,S_2}^{(e)} - z_H \rho_d D_H c_H$$
$$- K_2 c_F c_H - 4\pi r_c C_c D_H c_H - K_H c_H c_M \qquad (6)$$

$$\frac{dc_{S_1}}{dt} = K_H c_{S_0} c_H - K_H c_{S_1} c_{H,S_1}^{(e)} - K_H c_{S_1} c_H + K_H c_{S_2} c_{H,S_2}^{(e)} \qquad (7)$$

$$\frac{dc_{S_2}}{dt} = K_H c_{S_1} c_H - K_H c_{S_2} c_{H,S_2}^{(e)} - K_H c_{S_2} c_H \qquad (8)$$

$$\frac{dc_{S_3}}{dt} = K_H c_{S_2} c_H \qquad (9)$$

$$\frac{dc_{Cl_2}}{dt} = \rho_d (z_H D_H c_H - z_F D_F c_F) - \gamma D_F c_F c_{Cl_2} \qquad (10)$$

The rate reaction for the concentration of empty traps of H centers is:

$$\frac{dc_{S_0}}{dt} = K_H c_{S_1} c_{H,S_1}^{(e)} - K_H c_{S_0} c_H \qquad (11)$$

where at $t = 0$ the concentration of impurities is $c_{S_0}^0$

The number of colloids increases during irradiation because each N center that traps an F center is considered to be a new nucleus for a colloid. This gives rise to the following rate reaction:

$$\frac{dC_c}{dt} = N K_F c_F c_N \qquad (12)$$

At $t = 0$, there already is a number of unspecified colloid nuclei present in the salt; its number concentration is C_c^0.

According to the mechanism of Hobbs *et al.*, a c_{S3} center is considered to be the beginning of a new dislocation loop. The number of dislocation loops therefore is:

$$C_l = C_l^0 + N c_{S_3} \qquad (13)$$

Table I. List of symbols

Parameter	Definition
c_F	fraction of F centers
c_M	fraction of M centers
c_R	fraction of R centers
c_N	fraction of N centers
c_A	fraction of Na in colloids
c_H	fraction of H centers
c_{S_0}	fraction of impurities
c_{S_1}	fraction of single H centers trapped at impurities
c_{S_2}	fraction of H dimers trapped at impurities
c_{S_3}	fraction of H trimers trapped at impurities
c_{Cl_2}	fraction H centers converted into molecular Cl_2 centers
$c_F^{(e)}$	fraction F centers in equilibrium with colloids
$c_{H,S_1}^{(e)}$	fraction H centers in equilibrium with S_1 centers
$c_{H,S_2}^{(e)}$	fraction H centers in equilibrium with S_2 centers
K	production rate of uncorrelated F-H pairs
K_F	rate constant for the formation of M centers and colloids
K_R	rate constant for the formation of R and N centers
K_2	rate constant for the recombination of F and H centers
K_H	rate constant for the formation of S_n clusters
$K_M^{(e)}$	dissociation rate of M centers
D_F	diffusion coefficient of F centers
D_H	diffusion coefficient of H centers
r_c	mean radius of Na colloids
C_c	number of colloids per cm^3
z_F	dislocation bias coefficient for F centers
z_H	dislocation bias coefficient for H centers
ρ_d	dislocation line density
γ	rate coefficient for back reaction between F centers and molecular Cl_2
N	number of Na^+ ions in NaCl per cm^3
C_l	number of dislocation loops per cm^3
C_l^0	number of nuclei for dislocation loops per cm^3 at $t = 0$
ρ_0	dislocation line density at $t = 0$
b	Burgers vector of dislocation loops in NaCl
ν	ratio of molecular volume of NaCl and atomic volume of metallic Na

Jain and Lidiard have derived the following cross relations between the density of dislocation lines ρ_d and the concentration of molecular Cl_2 centers and between the mean colloid radius r_c and the colloid fraction c_A respectively:

$$\rho_d = \sqrt{\left(\frac{2\pi C_l}{b} c_{Cl_2} + \rho_0^2 \right)} \qquad \text{and} \qquad (14)$$

$$r_c = \sqrt[3]{\left(\frac{3 c_A}{4\pi \nu C_c} \right)} \qquad (15)$$

Note that equations (19) and (20) have been derived for fixed values for C_l and C_c respectively and that their usage for varying values of C_l and C_c, as in this paper, is only approximately valid. Further, the dislocation loops cannot grow infinitely but will join up into a network. If r_l is the mean loop length, a rough estimate for this to happen is[Jain and Lidiard, 1977]:

$$\frac{4\pi}{3} r_l^3 = \frac{1}{C_l} \qquad (16)$$

With $\rho = 2\pi r_l C_l$, we obtain in approximation for the maximum dislocation line density:

$$\rho_m \approx C_l^{2/3} . \qquad (17)$$

With respect to the original Jain-Lidiard model, the new features in the present model are the self-trapping of F centers into M, R and N centers and the trapping of H centers by impurities leading to S_1, S_2 and S_3 centers. The reaction mechanisms give rise to different rate equations for every defect type. These rate equations, which are described elsewhere in more detail[Soppe, 1993], contain some parameters for which an accurate value has not been determined yet. For the values of these parameters we have deduced an empirical approximation by comparing the results of numerical integration of the rate equations with various experimental data. The resulting model accounts well for a broad range of experimental results on radiation damage in NaCl published in the literature until sofar.

Table II.   Parameter values used in this work

Parameter	Value
K (dpa/s)	$1.667 \cdot 10^{-7} \cdot K$ (K in kGy/hr)
D_H (cm^2/s)	$1.0 \cdot 10^{-2} \exp(-0.10/kT)$
D_F (cm^2/s)	$1.5 \cdot 10^{-2} \exp(-0.80/kT)$
K_F (s^{-1})	$10^{16} D_F$
K_2 (s^{-1})	$10^{14} \exp(-0.10/kT)$
K_R (s^{-1})	$K_F \cdot \exp(-0.20/kT)$
K_H (s^{-1})	$10^{16} D_H$
$K_M^{(e)}$	$2 \cdot 10^{-3} \cdot K_F$
$c_F^{(e)}$	$2 \cdot 10^{-4} \exp(-0.30/kT)$
$c_{H,S_1}^{(e)}$	$5 \cdot 10^{-10} \exp(-0.25/kT)$
$c_{H,S_2}^{(e)}$	$5 \cdot 10^{-10} \exp(-0.10/kT)$
C_c^0 (cm^{-3})	10^{15}
z_F	1
z_H	1.1
ρ_0 (cm^{-2})	10^5
γ (cm^{-2})	$1 \cdot 10^{18} \exp(-0.55/kT)$
N (cm^{-3})	$2.22 \cdot 10^{22}$
C_i^0 (cm^{-3})	10^{15}
b (cm)	$3.98 \cdot 10^{-8}$
ν	1.13

3. RESULTS

According to the model, the colloid growth process is very sensitive to both dose rate and temperature. At very low temperatures, the mobility of the F centers is too low to allow the formation of clusters of significant size. Increasing the temperature enhances the F center mobility and therefore enhances the formation of colloids. At higher temperatures, however, the colloids start to dissociate by thermal emission of F centers which leads to a decrease of the colloid fraction. The result of these processes is that the colloid fraction as a function of the temperature, for a given dose rate and a given total dose, has a bell-like shape as shown in Figure 1. The colloid fractions in this figure result from calculations for pure NaCl.

The behaviour of the colloid growth as a function of the dose rate is likewise. For very high dose rates, the mean distance between F centers and H centers immediately

after their birth is so small that their survival probability is nil. Reducing the dose rate allows for colloid growth until the production of F centers is smaller than the emission of F centers from colloids and the net colloid growth is zero. Thus, for a given temperature and a given total dose, the colloid fraction as a function of the dose rate shows a maximum. This maximum shifts to higher dose rates if the temperature is increased. These features are not specific for the present model. Van Opbroek and Den Hartog, 1985, have shown that the original Jain-Lidiard model displays the same characteristics.
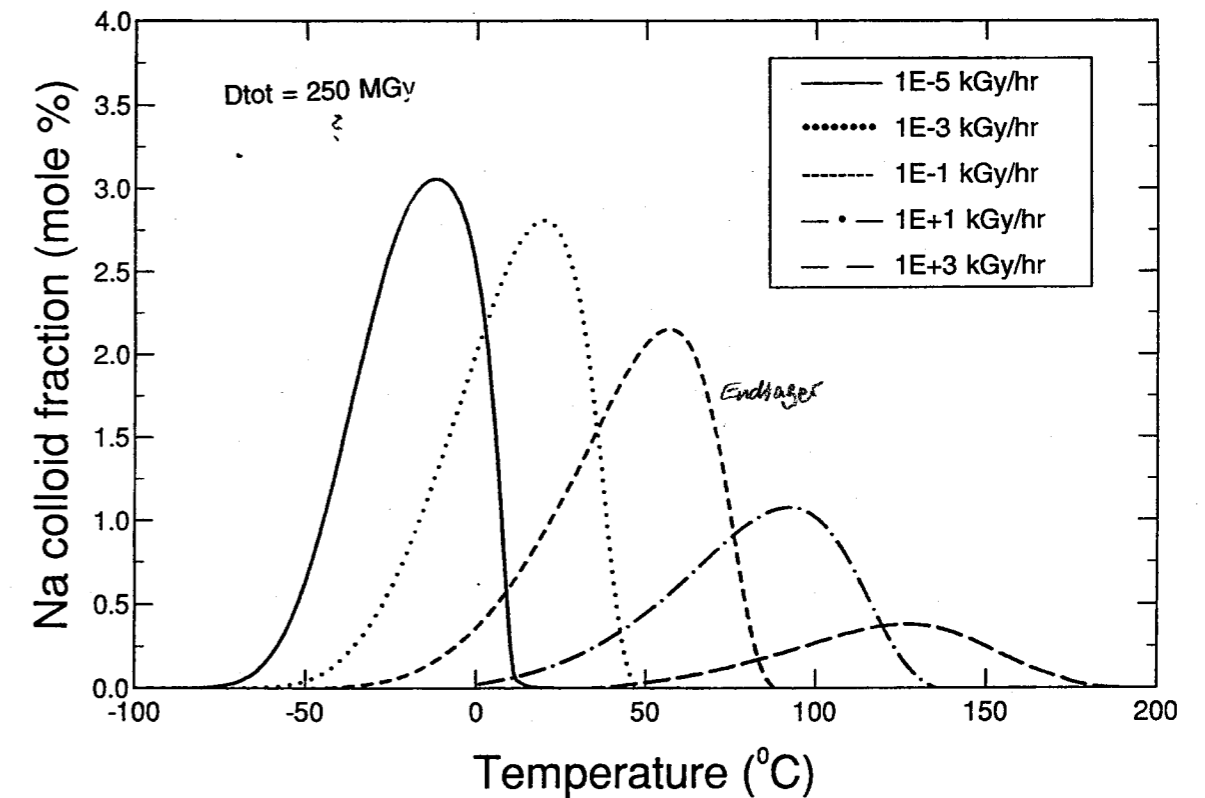

Figure 1:   *Colloid fraction in pure NaCl as a function of the temperature for various dose rates and at total dose of 2.5 GGy*

In the original model, the dose dependency of colloid growth can be devided in two regimes. In the first regime (at small doses) the colloid fraction increases quadratically with the dose. In the second regime (at larger doses) colloid growth slows down untill finallly a saturation of the colloid fraction is reached. The new model shows two extra regimes. First, at very low doses, there is an incubation period for colloid growth; colloids grow only after a certain threshold dose has been passed. This is an experimentally well determined

effect[Levy et al., 1984] and it is due to the colloid nucleation stage. Finally, at very large doses the new model predicts a second new regime in which the colloid fraction **decreases** again. This interesting (hypothetical) phenomenon is due to the circumstance that the number of colloids C_c increases with increasing doses. If C_c becomes very large, almost all F and H centers will be trapped by colloids before they can participate in other processes like trapping by dislocations. Thus, the actual driving force for net colloid growth: the differences in the drains of F and H centers to dislocations becomes negligible. The colloid fraction would finally drop to zero if C_c could grow without restraints.

In Figure 2 the colloid fraction as a function of radiation dose is shown for two different dose rates at $T = 100$ °C. It appears that the threshold dose for colloid growth is smaller for smaller dose rates. This leads to a higher efficiency of colloid formation at smaller dose rates as already observed in Figure 1. The saturation level of the colloid fraction at this temperature, however, appears to be higher for higher dose rates. Figure 2 also shows the rather drastic effect of the presence of trapping impurities on the colloid growth. For a dose rate of 10 kGy/hr, an impurity concentration of 10 ppm leads to a maximum colloid fraction that is about 4 times larger than for pure NaCl. This impurity effect increases with increasing concentrations but finally saturates. For a dose rate of 100 kGy/hr, saturation of the impurity effect is obtained at a concentration of about 100 ppm. If the dose rate decreases, the impurity effect also decreases and saturation sets in at higher concentrations (cf. Figure 2). For dose rates relevant for HLW disposal strategies (1 - 10^{-4} kGy/hr) the maximum theoretical enhancement of colloid growth by impurities appears to be about 50 %[Soppe and Prij, 1994].

Using the extended model, we have calculated the colloid fraction which will be formed in the salt very close to radioactive waste containers for some typical (Dutch) rock salt repository concepts. These concepts not only differ in the mining techniques, but also in the so called 'cooling time' and 'interim storage time' of the HLW. The cooling time is the period between the moment in which the HLW leaves the reactor and the moment in which it is reprocessed. The interim storage time is the period that the reprocessed and vitrified waste is stored overground until it is finally disposed in a rock salt formation. Characteristics of the investigated concepts (cases) are given in Table III. In the borehole concept, the waste containers are reinforced by a 3-cm steel overpack which leads to a reduction of the dose rate by about a factor 10 with respect to the mine concepts. The storage depth determines the geothermal background temperature of the salt. Heat from the waste containers gives rise to an extra temperature increase in the salt near to the containers. The maximum increase (which is gradually developed after about 10 years) is approximately 60 centigrades.
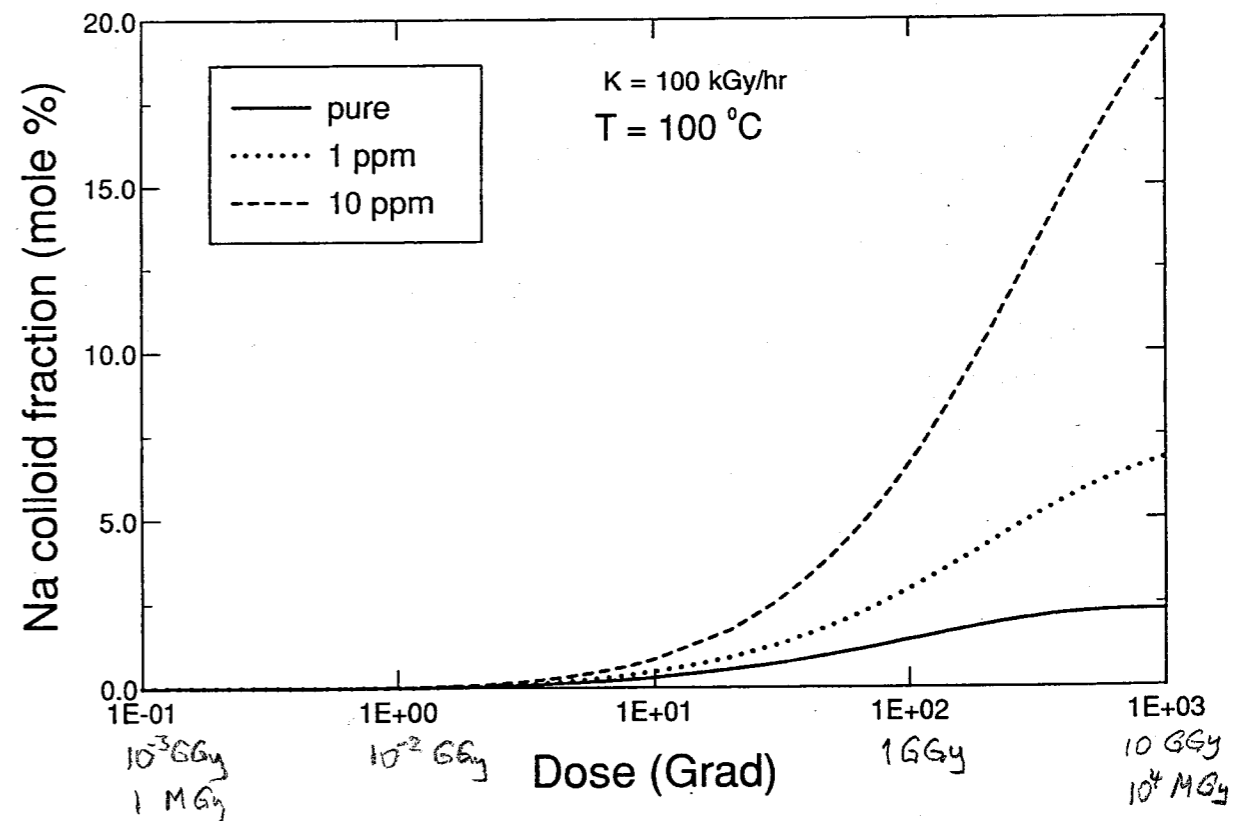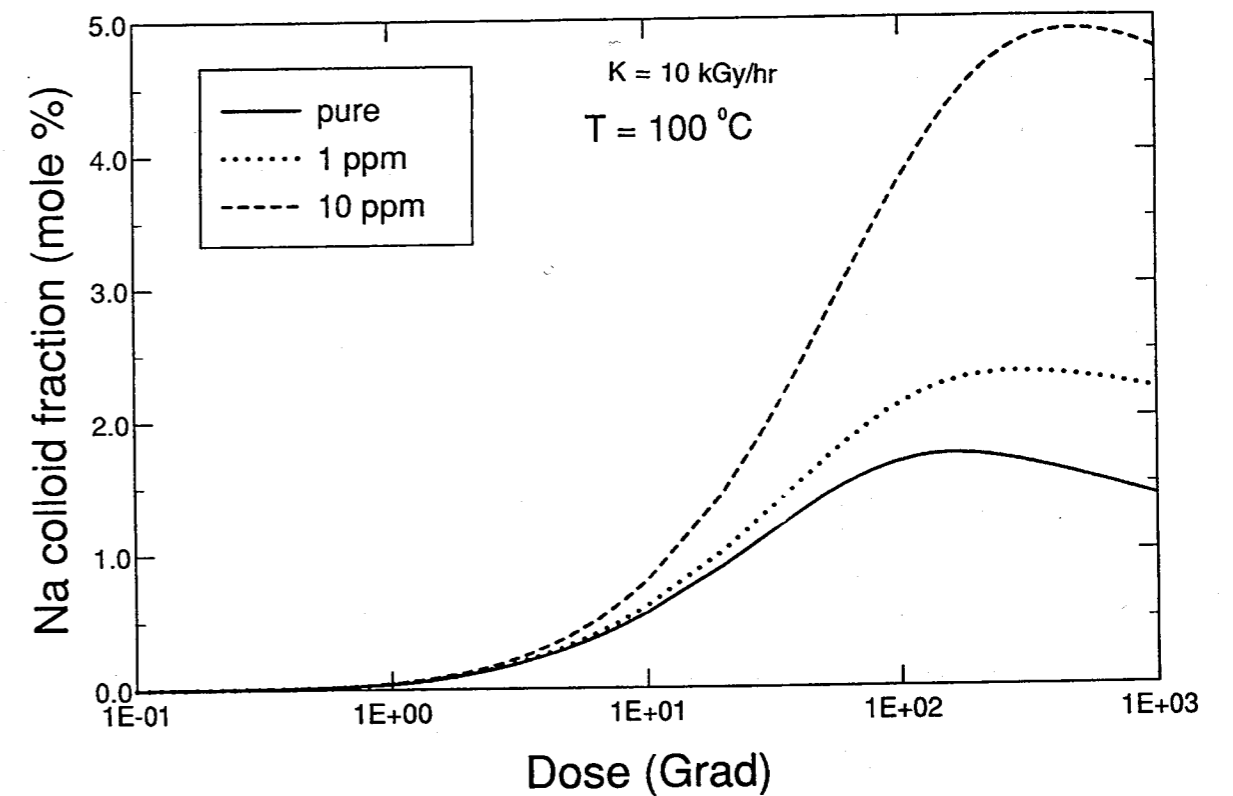

Figure 2   *Colloid fraction as a function of the radiation dose at T = 100 °C.*
(a) dose rate = 10 kGy/hr; (b) dose rate = 100 kGy/hr.

Table III. Specifications of the cases evaluated in this study.

Case No.	Formation Type	Disposal Technique	Cooling Time (y)	Interim Storage (y)	Storage depth (m)
11	pillow	borehole	3	50	616
12	pillow	borehole	10	50	616
15	pillow	borehole	3	10	619
16	pillow	borehole	10	10	619
17	pillow	mine	3	50	654
21	pillow	mine	3	10	654
22	pillow	mine	10	10	654
23	dome	borehole	10	10	957
24	dome	mine	10	10	1048

In all the cases, colloid growth appears to start after about one year (see Figure 3). The dose rate during this period is typically about 0.1 kGy/hr, *i.e.* the threshold dose is about 10 kGy. The largest increase of the radiation damage takes place in the period between 1 and 100 years. In this period the dose rate is more or less constant but then the dose rate drops fastly by about 2 orders of magnitude in about 500 years due to the decay of ^{90}Sr and ^{137}Cs. In the period between 10^2 and 10^6 years, the colloid fraction remains more or less constant and then starts to decay. For pure NaCl a maximum colloid fraction of 1.7 mole % is calculated. If the effect of trapping impurities is also taken into account, we expect a maximum colloid fraction of 2.3 mole %.
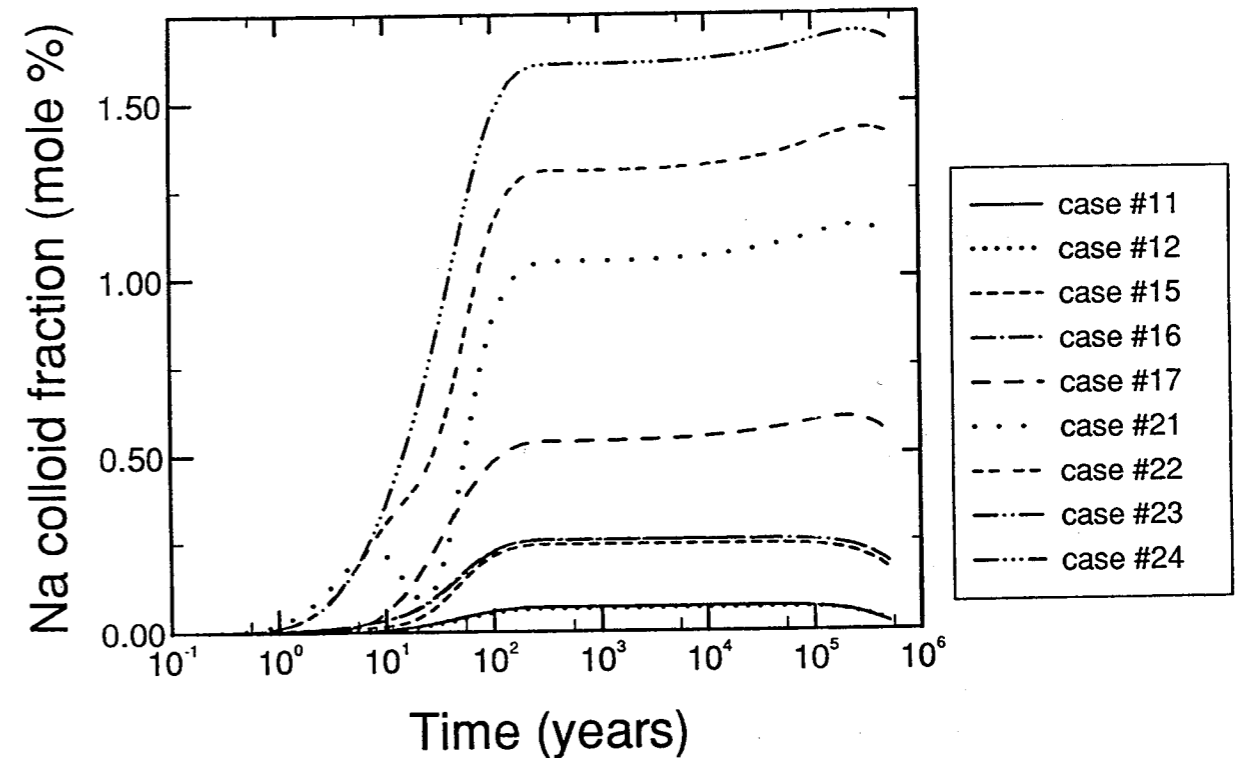

Figure 3   *Colloid fraction in rock salt close to container walls as a function of time under the assumption that the salt is free of impurities (The results for cases #16 and #23 are overlapping.)*

Radiation damage is limited to salt regions very near to the waste containers. In Figure 4 the maximum colloid fraction in rock salt as a function of the distance to the container wall is shown for one typical disposal strategy. It appears that the colloid fraction decays exponentially with increasing distances. The exponential decay length is rougly 5 cm, and in practice the amounts of radiation damage at distances larger than 20 cm are negligible.
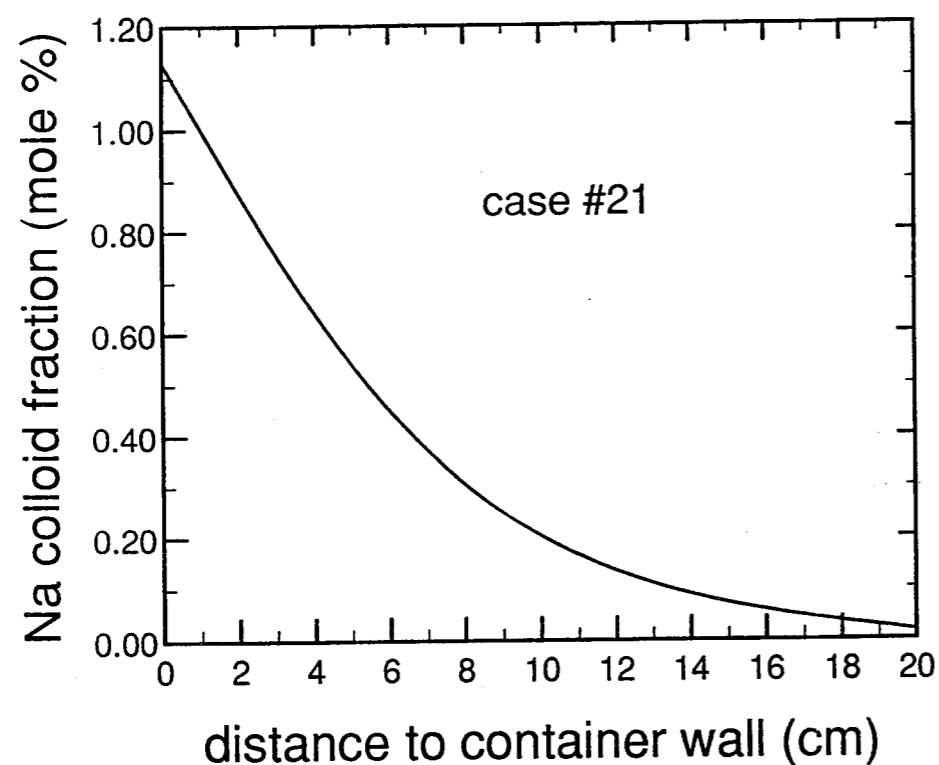
Figure 4   *Maximum colloid fraction in pure NaCl as a function of the distance from the container for case #21.*

4. CONCLUSIONS

The accumulation of radiation damage in rock salt is a complex process, involving a manifold of interactions between the various types of defects created by radiation. In order to meet with the practical demand for a theory describing the long-term behaviour of rock salt nuclear waste repositories, we have developed a new model based on kinetic rate reactions. This new model incorporates basic elements of the theory developed by Jain and Lidiard but also describes the nucleation stage of colloids and the effect of the presence of impurities on colloid growth. The present model, therefore, is expected to provide more realistic estimations of the amounts of radiation damage in rock salt under irradiation than

calculations with the original Jain-Lidiard model. Calculations performed for representative HLW repository designs indicate that, near a waste container, the maximum amount of NaCl that will be converted into metallic Na and molecular Cl_2 will not exceed a few mole percent.

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PART VI

CONCLUSIONS AND CONSEQUENCES