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PART VI

CONCLUSIONS AND CONSEQUENCES

EVALUATION OF THE SAFETY CONSEQUENCES OF RADIATION INDUCED STORED ENERGY IN A REPOSITORY IN ROCK SALT

J. Prij

ABSTRACT

This article evaluates the possible safety consequences of the radiation induced stored energy which might occur in the rock salt of a radioactive waste repository. An overview is given of the levels of stored energy which might be expected in some generic repositories. Possible release mechanisms of this stored energy are discussed. It is concluded that there are large safety margins between the conditions in a repository and the conditions at which very rapid release mechanisms occur. To judge the safety consequences, an instantaneous release has been postulated and its thermo-mechanical consequences are handled. The final conclusion is that the radiation induced stored energy in rock salt is not a safety problem.

1. INTRODUCTION

Due to the disposal of high level waste (HLW) in a salt formation gamma energy will be deposited in the rock salt. Most of this energy will be converted into heat, whilst a small part (~ 1‰), will create defects in the salt crystals. This energy is used in the decomposition of the NaCl into (colloidal) sodium and chlorine. It has been shown in this volume that the amount of stored energy depends on the total dose and the dose rate of the irradiation, the temperature at which the salt is irradiated, and on the nature and amount of intra crystalline defects (see part V of this Volume). In a repository these variables are not constant in time and space but vary substantially. Directly after disposal of the HLW the dose rate is high, ≤ 0.75 kGy/h, and due to the decay of the radionuclides the dose rate decreases monotonically. In the first 500 years after disposal, the decrease of the dose rate is about 4 orders of magnitude. The total dose increases monotonically from zero to ≤ 277 MGy.

The maximum dose rate and total dose are reduced significantly if an extra shielding container is used or if a long interim storage is applied. In the salt, at some distance from the HLW canister, the dose rates are considerably lower due to the shielding effect of the salt itself. At distances larger than 0.2 m the dose rate and total dose can be neglected [de Haas, 1989; Soppe et al, 1994].

The temperature of the salt rises due to the heat production of the HLW. The temperatures in the salt close to the HLW canister can reach values of more than 200 °C within 10 years. After this time period the temperature decreases slowly due to conductive heat transport. The maximum temperatures strongly depend on the heat production of the waste canisters and on the geometrical details of the repository [Prij, article 2 in this volume]. Hundred years after disposal the maximum temperatures will be below 75°C in the studied repository concepts studied [Jong, 1987].

It is obvious that the amount of radiation induced stored energy cannot be determined in a one to one experiment. This implies that computer simulations are needed to determine the development of the radiation damage and related stored energy. These computer simulations must be based on well understood physical processes. The models for the individual processes must be based on sound theory and tested against well defined experiments. One of the best known theoretical set of models is that based on the theory of Jain and Lidiard [1977]. A review of the theoretical and experimental work is given by Soppe et al [1994]. For some study cases computer simulations of the built up of stored energy in a repository have been made by different authors [de Haas et al, 1989 and Soppe et al, 1994]. The results of these simulations will be discussed in this article. In this discussion the experimental results obtained in the HAW project will also be accounted for. The study cases deal with 16 conceptual design variants of a repository. Disposal in boreholes in a mine as well as disposal in deep boreholes are considered (Prij, article 2 in this volume).

To obtain a soundly based opinion on the safety of HLW disposal in salt it is of importance to investigate whether mechanisms exists through which the stored energy can instantaneously be released. The consequences depend on the amount of stored energy and on the time needed for the release. If the release is instantaneous or explosive, dynamic effects will attenuate the consequences. The release mechanisms will be discussed shortly.

Only the mechanisms which are relevant for the amount of stored energy which can be expected in a repository will be handled.

To judge the most severe consequences of stored energy an instantaneous release has been postulated and its thermo-mechanical consequences are analysed. The results are here presented and discussed. The discussion is concentrated on the question of whether pathways in the salt can be created through which the nuclides can migrate from the canisters to the groundwater around the salt formation.

2. EXPECTED AMOUNT OF STORED ENERGY IN A REPOSITORY

To evaluate the, with respect to the radiation damage, most influential parameters in the design of a repository in rock salt 16 relevant study cases were defined by de Haas [1989] for which detailed analyses of the temperature, dose rate, total doses and built up of radiation damage have been performed. These analyses were performed with two computer models both based on the theory of Jain and Lidiard. The first model is the original Jain Lidiard model while the second model includes modifications made by Soppe [1994]. The modifications are based on the work of Groote and Weerkamp [1990] and deal with the effect of impurities and colloid nucleation.

2.1 Amount of radiation damage [mol%]

Some results are summarized in Table I were the maximum percentage of decomposed salt into sodium colloids and Cl_2 is given for pure rock salt and for rock salt with impurities. Based on the results of De Haas [1989], not all 16 study cases were reanalysed by Soppe but only the representative ones. It can be seen that the model extensions of Soppe lead to a lower level of radiation damage. This is mainly caused because in this model the damage is less dependent on the dose rate than in the original model [Soppe et al, 1994]. For the repository concepts consisting of disposal in deep boreholes after a long interim storage, the dose rate is the lowest and consequently the effect of the model extension is the highest, a reduction of a factor 8. For the repository concepts of disposal in a mine after a short interim

Table I: *Calculated maximum amount of radiation damage for several study cases.*

Case	Burn up [GWd/tU]	Energy [EJ]	t_{pre} [a]	t_{int} [a]	FT	DT	C^H [%]	C^{S1} [%]	C^{S2} [%]
11	33	0.5	3	50	P	D	0.55	0.07	0.07
12	33	0.5	10	50	P	D	0.51	0.07	0.07
13	33	3.5	3	50	P	D	0.56	n.a.	n.a.
14	33	3.5	10	50	P	D	0.52	n.a.	n.a.
15	33	3.5	3	10	P	D	1.06	0.25	0.25
16	33	3.5	10	10	P	D	1.09	0.26	0.26
17	33	0.5	3	50	P	M	2.93	0.60	0.95
18	33	0.5	10	50	P	M	2.88	n.a.	n.a.
19	33	3.5	3	50	P	M	3.24	n.a.	n.a.
20	33	3.5	10	50	P	M	3.13	n.a.	n.a.
21	33	3.5	3	10	P	M	4.17	1.15	1.26
22	33	3.5	10	10	P	M	4.10	1.42	1.60
23	33	3.5	10	10	D	D	0.70	0.26	0.26
24	33	3.5	10	10	D	M	4.11	1.70	2.23
25	40	3.5	10	10	P	D	1.08	n.a.	n.a.
26	40	3.5	10	10	P	M	4.09	n.a.	n.a.

Energy: The amount of waste is related to the electric energy produced in the nuclear power plants. The 0.5 EJ is related to an installed nuclear power of 0.5 GWe and an operational period of 30 years. The 3.5 EJ is related to an installed power of 3.5 GWe.
t_{pre} Pre storage, time between removal of the reactor and reprocessing of the spent fuel
t_{int} Interim storage, time between reprocessing and disposal
FT Formation type.
P: Pillow with an overburden thickness of 800 m.
D: Dome with an overburden thickness of 230 m.
DT Disposal technique
D: Deep boreholes drilled from the earth' surface.
M: Mine with galleries and boreholes drilled from the floor of the galleries.
C^H The maximum colloid fraction [mol %] calculated with the original Jain Lidiard model by de Haas [1989]
C^{S1} The maximum colloid fraction [mol %] calculated with the extended Jain Lidiard model by Soppe for pure rock salt [1994]
C^{S2} The maximum colloid fraction [mol %] calculated with the extended Jain Lidiard model by Soppe for rock salt with an impurity concentration for which the maximum colloid fraction is obtained [1994]
n.a. These cases are not analysed by Soppe.

storage the dose rate is the highest and therefore the effect of the model modification is the lowest, a reduction of about a factor 3. It can further be observed that the effect impurities can have on the total damage in a repository, which is also included in this modified model, can be to increase in the maximum radiation damage by about 50 %. The results clearly indicate that the first important design parameter is the presence or absence of a shielding container. For the 'deep borehole' cases with a 3 cm thick shielding container the maximum colloidal fraction is about a factor 4 lower than for the corresponding 'mine' cases without

a shielding container. The next important parameter is the duration of the interim storage. For the cases with an interim storage of 50 years the maximum colloidal fraction is about 60 % of the fraction obtained for an interim storage of 10 years. The burn up, the pre storage duration and the type of salt formation appear to have a very small influence on the maximum colloidal fraction.

2.2 Amount of specific stored energy [J/g]

The amount of specific stored energy in the salt crystals is directly related to the amount of defects. In the calculations performed by De Haas it has been assumed that the amount of specific stored energy per mol% of colloid present is 70 J/g. This corresponds to 4.25 eV per defect. In the analyses of Soppe it has been assumed that 1 mol% metallic sodium and molecular chlorine corresponds with a stored chemical energy of 125 J/g [Soppe et al. 1994; Groote and Weerkamp, 1990]. Since the parameter values used in the models are calibrated using these conversion factors the same factors will be used here to calculate the

Table II: *Predicted maximum amount of specific stored energy Q_{max} and storage efficiency η_{SE}*

Case	Initial dose rate [kGy/hr]	Original J-L model		Extended J-L model[1)]	
		Q_{max} [J/g]	η_{SE} [‰]	Q_{max} [J/g]	η_{SE} [‰]
11	0.037	38.5	2.2	8.8	0.5
12	0.032	35.7	2.2	8.8	0.6
13	0.037	39.2	2.2	n.a.	n.a.
14	0.032	36.4	2.3	n.a.	n.a.
15	0.116	74.2	1.8	31.3	0.8
16	0.088	76.3	2.2	32.5	0.9
17	0.254	205.1	1.7	118.8	1.0
18	0.215	201.6	1.8	n.a.	n.a.
19	0.254	226.8	1.9	n.a.	n.a.
20	0.215	219.1	2.0	n.a.	n.a.
21	0.750	291.9	1.0	157.5	0.6
22	0.585	287.0	1.2	200.0	0.8
23	0.088	49.0	1.4	32.5	0.9
24	0.585	287.7	1.2	278.8	1.2
25	0.088	75.6	2.1	n.a.	n.a.
26	0.585	286.3	1.2	n.a.	n.a.

1) The maximum specific stored energy corresponds to the values for impure rock salt (C^{S2} from Table I)
n.a. These values are not analysed by Soppe

amount of stored energy. The resulting specific stored energies for the study cases considered are given in Table II. This table also gives the storage efficiency η_{SE} which is defined as the ratio of the specific stored energy (in J/g) to the total dose (note that 1 MGy = 1000 J/g) [Soppe et al, 1994]. It can be observed that the storage efficiency is the highest when the dose rate is the lowest. Since the dose rate effect is less pronounced in the extended Jain Lidiard model the storage efficiency predicted with it is lower than that predicted by the original model. It can further be observed that the storage efficiency calculated with the extended model depends more on the impurity content than on the dose rate. It is important to emphasize that, following these results, only a very small fraction 0.5 to 2.3‰ of the gamma energy is converted into chemical energy. The 'remaining' energy is deposited in the salt in the form of thermal energy.

The build up versus time of the radiation damage in the salt directly in contact with the canister is shown in Figure 1. This figure is based on the analyses with the extended model [Soppe et al, 1994]. A comparison of these results with the temperature profiles {given by Jong, 1987 and Prij, 1995] leads to the conclusion that in the period during which the temperatures are rising the recombination and anneal mechanisms are stronger than the damage building processes. After the temperature has reached the maximum value and is decreasing the damage is built up.

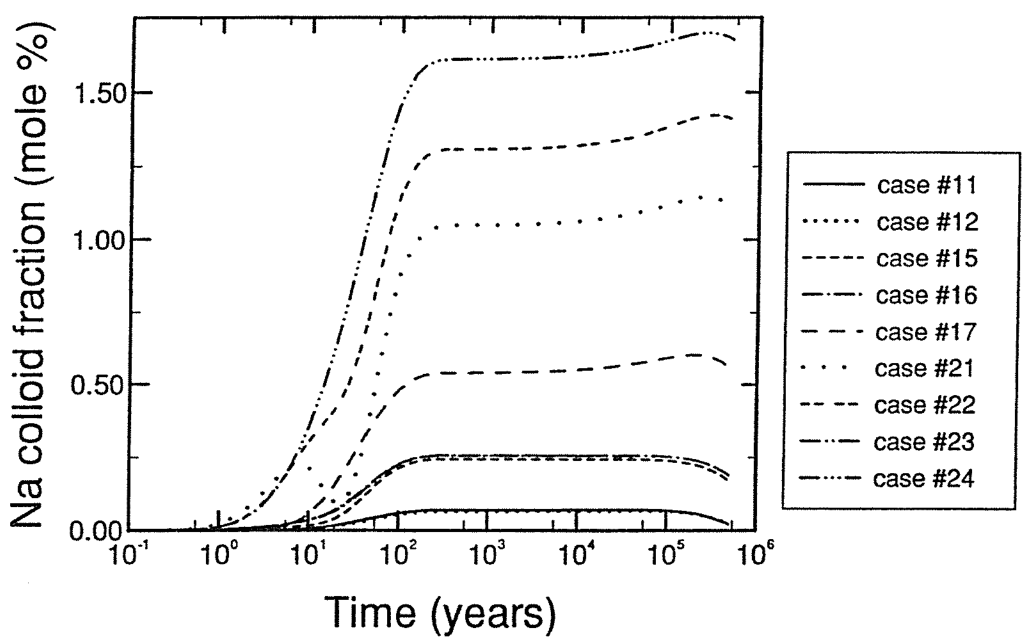

Figure 1. *Calculated colloid fraction in the salt very near to the canister C_{max} for the various study cases.*

A comparison of this build up with the evolution of the dose rate leads to the conclusion that the 'stationary' damage level is already reached at about 100 years after disposal, the period in which the dose rate has decayed about one order of magnitude. The total dose until this moment is about 65 % of the total yield of gamma radiation for the cases with an interim storage duration of 10 years. For the cases with an interim storage of 10 years this percentage is about 80 %.

For the study cases with a short interim storage and no extra shielding the stationary level of damage is higher than the saturation level found in the laboratory experiments [Donker et al., 1995]. This is due to anneal processes which take place in the experiments such as a long nucleation phase, and several creep processes which are not accounted for in the models.

2.3 Amount of stored energy [J]

The total amount of stored energy around one HLW container, Q_t, is related to the amount of specific stored energy and is given by the relation:

$$Q_t = \int_V Q(x,y,z)\,\rho\,dV \tag{1}$$

where ρ is the density [kg/m^3] of the rock salt and V is the volume of decomposed salt [m^3].

The amount of specific stored energy at some distance δ from the container $Q(\delta)$ decreases exponentially:

$$Q(\delta) = Q_o e^{-\frac{\delta}{\lambda}} \tag{2}$$

where Q_o is the specific stored energy in the salt directly at the container wall and λ is the attenuation length. This is also illustrated in Figure 2. The attenuation length for colloid formation and stored energy can be derived from the Jain Lidiard calculation. The analyses with the original Jain Lidiard model result in λ = 6.8 cm [de Haas, 1989] while λ = 5.5 cm according to the Soppe model [Soppe et al, 1994]. This difference is caused by the stronger dose rate dependence in the original Jain Lidiard model as compared to that of Soppe.

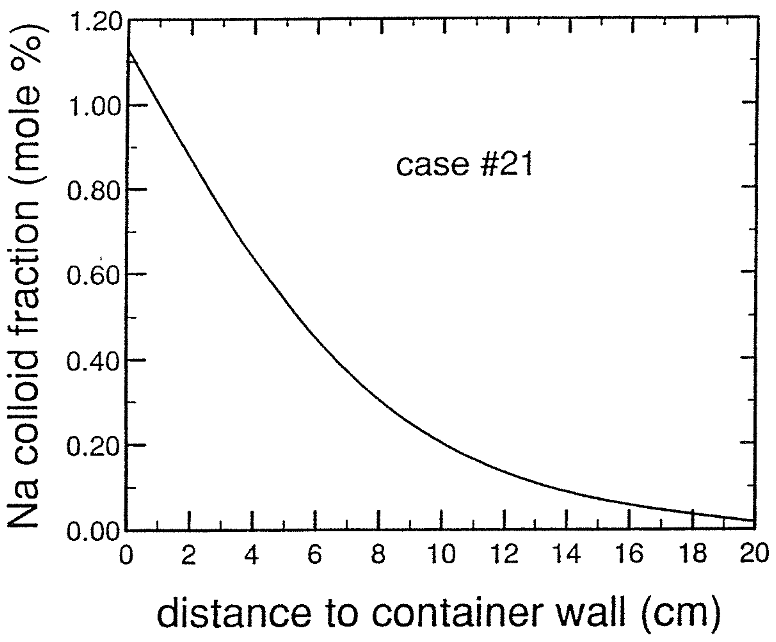

Figure 2. *Calculated maximum colloid fraction in pure salt as a function of the distance to the container wall for study case 21.*

Substitution of eq. 2 in eq. 1 leads to the following relation:

$$Q_t = \int_0^\infty Q_o e^{-\frac{\delta}{\lambda}} \rho \left(2\pi(h+2\lambda)(r_o+\delta)+2\pi r_o^2\right) d\delta \equiv Q_o \rho V_{eq} \qquad (3)$$

where the height of the glass in the waste container, h, is 1.1 m, and the outer radius of the container, r_o, is 0.21 m. Using these parameter values the calculated equivalent volume of decomposed salt, V_{eq}, is 0.166 m^3 for the original Jain Lidiard model and 0.126 m^3 for the Soppe model. The effective mass of decomposed salt is 358 kg and 272 kg respectively. Once these effective masses are known, the total stored energy in the salt around one HLW container can be calculated. The results are summarized in Table III. It can be observed that the amount of stored energy varies between 2 and 104 MJ, depending upon the disposal concept and model used. It is interesting to realize that the electrical energy produced with the fuel elements which constitute the one container of waste is about 1.7 10^{15}J whereas the thermal energy produced by one container of HLW is about 10^{12}J.

Table III: *Predicted amount of stored energy Q_t around one HLW container*

Case	Original J-L model	Extended J-L model[1)]
	Q_t [MJ]	Q_t [MJ]
11	13.7	2.4
12	12.8	2.4
13	14.0	n.a.
14	13.1	n.a.
15	26.6	8.5
16	27.3	8.9
17	73.4	32.3
18	72.1	n.a.
19	81.1	n.a.
20	78.4	n.a.
21	104.4	42.9
22	102.7	54.5
23	17.6	8.9
24	102.9	75.9
25	27.1	n.a.
26	102.4	n.a.

1) The maximum specific stored energy corresponds to the values for impure rock salt (C^{S2} from Table I)

n.a. These values are not analysed by Soppe

3. POSSIBLE RELEASE MECHANISMS OF THE STORED ENERGY

The energy which storage was discussed above will be released if the Na and Cl recombine. Recombination takes constantly place and therefore the damage level is not continuously increasing but reaches a saturation level as long as the radiation continues. After radiation the gradual recombination 'removes' the damage. This recombination process is not a safety issue as the related heat production is about 1‰ of the amount of heat already accounted for in the design and safety analyses. Of direct importance for the safe disposal of radioactive waste is the question whether mechanisms exist by which the stored energy can be released suddenly. If these mechanisms cannot be totally excluded it has to be investigated whether the release of the stored energy can create a pathway for the nuclides from the containers to the ground water.

Two mechanisms are known: a thermally activated process, and a spontaneous backreaction. The stored energy can be released through the recombination of sodium and chlorine via a *thermally activated process* which can be initiated if the temperature is high enough. In the differential scanning calorimetry (DSC) experiments, which were performed to measure the stored energy, the energy release occurred at temperatures between 200 and 300 °C [Den Hartog et al. 1990; Groote and Weerkamp, 1990; Garcia Celma et al. 1988, 1992; Opbroek et al. 1985]. Once this release mechanism starts, the released energy will cause a rise in temperature, thus leading to a faster release of the remaining stored energy. In this way all stored energy will be released quickly. Considering the temperatures at which the radiation damage remains permanently present (< 75 °C [Bergsma et al. 1985; De Haas et al. 1989; Prij, 1991], there is a large safety margin for this release mechanism because an external heat source which rapidly heats the salt to 200 °C or more would be necessary. Such a heat source cannot in all reasonableness be imagined.

It is expected that a *spontaneous back reaction* will occur if the so-called percolation barrier is exceeded. This will be the case if the concentration of decomposed salt reaches a certain level. In the beginning of the HAW research project it was thought that this level was 20 to 30 mol%, later the value of 12 mol% was considered to be more realistic [Soppe et al., 1994]. This implies that in cases where the sodium and chlorine segregations are not uniformly distributed the stored energy can be spontaneously released even if the average concentration is slightly less than 12 mol%. During laboratory experiments some samples with an extremely high gamma energy dose were completely shattered after the DSC measurement and sometimes after the irradiation [Groote and Weerkamp, 1990]. In the fragments still a significant amount of energy was found to be stored. It is not completely clear whether this shattering of the samples is caused by a violent recombination of sodium and chlorine or by mechanical stresses due to the high pressure in the chlorine segregations. Considering the levels of radiation damage to be expected in a repository, about 1 mol%, there is a large safety margin for this release mechanism.

Based on these considerations one can conclude that there are large margins between the conditions in a repository and the conditions needed for the instantaneous release mechanisms.

4. THERMO-MECHANICAL CONSEQUENCES OF A POSTULATED INSTANTANEOUS RELEASE OF STORED ENERGY

To judge the most severe consequences of stored energy an instantaneous release has been postulated and the thermo-mechanical consequences were analysed [Prij, 1991]. Given the uncertainties in the release mechanism detailed numerical analyses of the thermo-mechanical consequences have not yet been performed and analytical approximations were considered to be more appropriate [Prij, 1991]. These analytical approximations are based on solutions for point and line sources in a homogeneous elastic medium. The solutions for the thermal stresses due to a point source were used to estimate the consequences of the release of stored energy in the rock salt around *one* container, and the solutions for the thermal stresses due to a line source were used to approximate the consequences of the release of stored energy in the rock salt around *all containers* in one borehole at exactly the same moment [Prij, 1991]. The approach for the point source will be summarized briefly.

4.1 Quasi-static solutions

The temperature distribution $T(r,t)$ due to an instantaneous heat source Q [J] acting at the origin of a spherical coordinate system is known [Nowacki, 1962; Parkus, 1959]:

$$T(r,t) = \frac{Q}{8\rho c\left(\sqrt{\pi\kappa t}\right)^3} e^{-\psi^2} \tag{4}$$

with:

$$\psi(r,t) = \frac{r}{2\sqrt{\kappa t}} \tag{5}$$

where κ is the heat distributivity $\lambda/\rho c$, λ is the heat conductivity coefficient, c is the specific heat, and t is the time.

Assuming that the acceleration effects can be neglected, the radial and tangential stresses are [Nowacki, 1962; Parkus, 1959]:

$$\sigma_r(r,t) = -2A\left(erf\psi - \frac{2\psi}{\sqrt{\pi}}e^{-\psi^2}\right)$$

$$\sigma_{tg}(r,t) = A\left(erf\psi - \frac{2\psi}{\sqrt{\pi}}(1+2\psi^2)\,e^{-\psi^2}\right) \tag{6}$$

with:

$$A = \frac{E\alpha}{4\pi(1-\nu)\rho c}\frac{Q}{r^3} \tag{7}$$

where E is the Youngs Modulus, α is the linear thermal expansion coefficient, and υ is the Poisson's ratio. The stresses at small distances are rather high and decrease rapidly with increasing radius or time, and after a short time the radial as well as the tangential stresses approach the value $-2A$ resp A. This implies that after this initial period the stresses are proportional with r^{-3}, see eq. 7.

In order to assess these stresses, the total state of stress has to be considered. The total stress components can be found by superimposing the lithostatic pressure p on the thermal stresses. The stresses can cause fracture when fracture criteria for shear or tensile failure are met. Two criteria will be considered [Prij, 1987]:

$$\text{i)}\quad \tau_{fr} = \sqrt{7T_0(\sigma_n + T_0)}$$

$$\text{ii)}\quad \sigma_3 = T_0 \tag{8}$$

where τ_{fr} is the shear strength [MPa], σ_n is the mean normal compression [MPa], T_0 is the tensile strength (2.4 MPa [Prij, 1987]), and σ_3 is the maximum principal stress [MPa].

The maximum shear stress is:

$$\tau_{max} = 0.5(\sigma_{tg} - \sigma_r) = \frac{A}{2}\left(3\,erf\psi - \frac{2\psi}{\sqrt{\pi}}(3+2\psi^2)\,e^{-\psi^2}\right) < \frac{3A}{2} \tag{9}$$

The mean normal compression σ_n is:

$$\sigma_n = p - \frac{2\sigma_{tg} + \sigma_r}{3} = p + \frac{8A}{3\sqrt{\pi}}\psi^3 e^{-\psi^2} > p \tag{10}$$

Combining eqs. 7, 8, 9 and 10 leads to the following relation for the region where the stresses do not cause shear fracture:

$$r > r_s \equiv \left(\frac{3E\alpha Q}{8\pi(1-\nu)\rho c\sqrt{7T_0(p+T_0)}}\right)^{1/3} \Rightarrow \tau_{max} < \tau_{fr} \tag{11}$$

The tensile criterion is determined by the maximum principal stress σ_3 which equals the sum of σ_{tg} and $-p$. Realizing that the tangential stress is always less than A, the following relation for the region where the stresses do not cause tensile fracture is obtained:

$$r > r_t \equiv \left(\frac{E\alpha Q}{4\pi(1-\nu)\rho c(p+T_0)}\right)^{1/3} \Rightarrow \sigma_3 < T_0 \tag{12}$$

When fracture occurs energy will be needed to create the free surfaces. It can be anticipated, however, that this will be less than the elastic energy assumed in the analysis. This implies that the real fracture zone can be slightly larger than indicated with eqs. 11 and 12. As indicated above the neglect of dynamic effects may also lead to an underestimation of the fracture zone.

4.2 Dynamic amplification

The problem of the dynamic stress distribution due to an instantaneous heat source concentrated in a point has been handled by Nowacki [1962]. As can be imagined the difference between the dynamic and the quasi-static solution, is noticeable only in a limited period of time. This dynamic effect also strongly depends on the time needed to release the energy and the area or volume of the source. If this time is set to zero, and the heat source is concentrated in a point, the dynamic solutions for the stresses and displacement have a discontinuity. To numerically investigate whether this discontinuity will occur in reality the release mechanism has to be known. As the precise release mechanism is not known it is not possible to make an accurate dynamic analysis. There is, however, a method which can provide a reference point with respect to the importance of the dynamic effects. This method is based on the use of explosives in civil engineering and mining technology. The following procedure has been applied [Prij, 1991]:

a) The quasi-static solution for a point source is compared with an empirical relation for a subsurface explosion. Based on the similarity of this relation and the quasi-static solution, a dynamic amplification factor is determined and an estimate for the dynamic solution is obtained.

b) This estimate of the dynamic solution is validated with some results of large nuclear explosions deep in salt formations.

An empirical relation exists for the burial depth h needed to obtain a 'contained' explosion of yield L [N.N. 1975; Parker, 1970; Wild, 1984]:

$$h > c_1 \, L^{1/3} \tag{13}$$

where h is the burial depth of the explosive [m], L is the yield of the explosive in kt (1 kt = 10^{12} cal = 4.2 10^{12}J [N.N. 1975]), and c_1 is a constant which generally depends on the type of explosive and the type of rock formation. The highest factor has been found for a nuclear explosion: 120 [N.N. 1975, Parker, 1970]. These values are based on experiments in different rocks including rock salt.

A contained yielding implies that the burial depth h is large enough to ensure that the fracture criteria are not met at the earth surface where $p = 0$. The quasi-static solutions given above then give the following prediction for the burial depth:

$$h_{Q.S} > \left(\frac{E\alpha}{4\pi(1-\nu)\rho c T_0} \right)^{1/3} Q^{1/3} \equiv c_2 \, Q^{1/3} \tag{14}$$

The parameter c_2 depends upon the thermo-mechanical properties and appears to be 54 m/(kt)$^{1/3}$ [Prij, 1994]. A comparison between the quasi-static solution and the empirical relation leads to the conclusion that the relation between the yield and the depth only differs in the factor c_1 and c_2. It can be concluded that for rock salt an amplification factor of 2.0 to 2.4 on the quasi-static solution is needed to predict the condition for 'contained' yielding.

A reasonable estimate for the dynamic solution of the instantaneous point source is the product of the quasi-static solution and the dynamic amplification factor. According to this dynamic estimate the region where the stresses do not cause shear fracture is bounded by:

$$r_s = 2.4 \left(\frac{3\,E\,\alpha}{8\pi(1-\nu)\rho c\sqrt{7\,T_0(p+T_0)}} \right)^{1/3} \cdot Q^{1/3} \tag{15}$$

And the region where the stresses do not cause tensile fracture is bounded by:

$$r_t = 2.4 \left(\frac{E\alpha}{4\pi(1-\nu)\rho c(p+T_0)} \right)^{1/3} \cdot Q^{1/3} \tag{16}$$

The dynamic estimates have been validated with the results of two underground nuclear explosions in rock salt formations viz. the Gnome shot and the Salmon Event [Prij, 1991].

The Gnome shot was the first Plowshare experiment. It was performed on 10 December 1961 at a depth of 360.9 m in the bedded salt formation 25 miles southeast of Carlsbad New Mexico. The fission product analyses showed that the actual yield was 3.1 ± 0.5 kt. Post explosion inspection showed that a cavity was created with a volume of nearly 1 million cubic feet, equivalent to a sphere with diameter of 38 m [N.N. 1963]. It was noted that the walls of the cavity and the vent path had a blue colour which is attributed to the radiation damage of the salt crystals. Radial fractures about the cavity extended up to 60 m. Some of these fractures had melt injected into them 12 m from the walls [N.N. 1963]. The dynamic estimates of eqs. 15 and 16 predict maximum crack lengths of 80 - 100 m. The observed crack length after the Gnome shot is shorter than predicted. This is assumed to be explained by the fact that the seal of the adit was not functioning as it should be [Prij, 1991].

The Salmon Event was a 5.3 ± 0.5 kt nuclear detonation at a depth of 827.8 m in the Tatum salt dome in Mississippi on October 22, 1964. The explosion created a nearly spherical and stable cavity of radius 17.6 ± 0.6 m. After the explosion two boreholes were drilled in the direct surrounding of the cavity providing samples for geophysical investigation. Radioactive melt injected into cracks was observed as far as 37 m from the shot point, and radioactivity increased above background as far as 64 m. The salt in the direct surrounding was highly microfractured and contained some macrofractures. It was found that beyond 90 to 120 m from the shot point, the rock salt approached the pre-shot condition [Rawson, 1966]. The dynamic estimates of eqs. 15 and 16 predict maximum crack lengths of 100 - 120 m. These predictions and observations for the Salmon event agree very well.

It has been concluded that the dynamic effects of the postulated instantaneous energy release can be bounded with the quasi static solution and the dynamic amplification factor of 2.4 [Prij, 1991]. For the line source similar solution have been obtained and the same dynamic amplification factor has been assumed.

4.3 Thermo-mechanical results

The thermo-mechanical consequences of the postulated instantaneous energy release for the study cases are summarized in Table IV. The maximum temperature rise ΔT_{max} is the instantaneous rise in temperature directly at the container wall and r_s is the radius of the region around which cracks will not occur. The values for r_s are presented here while they gave the largest cracks for the disposal concepts considered. The values for the line source are also smaller [Prij, 1991 and 1994]. It can be seen that the temperature rises are restricted to 320 °C while the region where cracks could occur is restricted to 2 m.

To obtain an upper bound of the consequences of the stored energy the consequences were considered of a radiation damage of 20 mol% and the instantaneous release of the induced stored energy, 0.66 GJ [Prij, 1991]. The crack length was estimated to be max. 4.8 m. It can be concluded that the thermo-mechanical consequences of the postulated instantaneous release of radiation induced stored energy is restricted to a small zone around the waste container. The dimensions of this zone are not strongly dependent on the level of radiation damage.

The zone in which possibly cracks occur is small, some meters, compared to the distance between the waste container and the boundary of the salt formation which is at least 200 m in the conceptual designs considered. The isolation capacity of the salt formation is therefore not significantly reduced. This implies that as a consequence of the postulated

instantaneous release of stored energy no pathway is created for the nuclides to migrate from the waste to the boundary of the salt formation.

Table IV: *Predicted thermo-mechanical consequences*

Case	Original J-L model		Extended J-L model[1)]	
	ΔT_{max} [°C]	r_s [m]	ΔT_{max} [°C]	r_s [m]
11	42.3	1.0	9.7	0.6
12	39.2	1.0	9.7	0.6
13	43.1	1.0	n.a.	n.a.
14	40.0	1.0	n.a.	n.a.
15	81.5	1.2	34.4	0.8
16	83.8	1.3	35.7	0.9
17	225.4	1.7	130.5	1.3
18	221.5	1.7	n.a.	n.a.
19	249.2	1.8	n.a.	n.a.
20	240.8	1.8	n.a.	n.a.
21	320.8	2.0	173.1	1.5
22	315.4	1.9	219.8	1.6
23	53.8	1.1	35.7	0.9
24	316.2	1.9	306.4	1.8
25	83.1	1.2	n.a.	n.a.
26	314.6	1.9	n.a.	n.a.

1) The maximum specific stored energy corresponds to the values for impure rock salt (C^{S2} from Table I)

n.a. These values are not analysed by Soppe

5. SUMMARY AND CONCLUSIONS

In this article the consequences of radiation damage and induced stored energy in rock salt have been estimated for some repository concepts. The models used for the calculation of the amount of damage as well as for the thermo-mechanical consequences have been calibrated against experiments and have been validated with other experiments. The values for the different parameters are conservative or best estimate values. A summary of the main results is:

1. The radiation damage is restricted to some mol% and concentrated in a region around the waste containers with a thickness less than 0.2 m. For about 100 years after disposal a saturation is predicted at a level about a factor of two higher than observed in the laboratory

experiments. This shows that the models are somewhat conservative. The damage can be reduced if a large interim storage period and an extra shielding container is used.

2. Related to the radiation damage, chemical energy is stored in the rock salt around the container. For the cases with a very short interim storage and without any shielding container the maximum level of specific stored energy is less than 300 J/g. At distances from the container the specific stored energy is lower. An exponential decay with an attenuation length of about 6 cm has been predicted. The total stored energy around one HLW container ranges from 2 to 100 MJ.

3. There are two mechanisms through which the stored energy can be released in a very short time. The thermally activated process which requires temperatures of more than 200 °C, and the percolation process which requires damage levels of more than 12 mol %. As the temperature at which the damage is established is below 75 °C and the damage level is only some mol% these mechanisms will not occur. The only remaining mechanism is a gradual recombination of the colloidal sodium and molecular chlorine.

4. To judge the safety consequences, an explosive release of the radiation induced stored energy has been postulated and the consequences have been assessed. It appears that cracks can occur in a zone around the waste containers. The outer radius of this zone is some meters. Outside this zone the salt is undisturbed. Consequently the isolation shield of more than 200 m around the repository is not significantly affected. This means that a pathway for nuclides from the waste containers to the groundwater will not be induced in case of a postulated explosive release of stored energy.

These results underline the huge margins of safety in the contemporary repository designs with respect to the consequences of radiation induced stored energy.

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GENERAL CONCLUSION AND RECOMMENDATIONS REGARDING RADIOLYTIC CONSEQUENCES OF DISPOSAL OF HIGH LEVEL RADIOACTIVE WASTE IN SALT FORMATIONS.

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ABSTRACT

On the basis of the research presented in this volume, it is concluded that the radiation damage (of the halite crystals of the rock salt) which would be produced by gamma-rays in radioactive waste repositories, where containers of high level waste would be disposed off (with or) without a thick steel overpack in dry-drilled deep boreholes, will not constitute a safety problem. Some recommendations are made for further research in the field of radiolytic gas development.

1. INTRODUCTION

As described by Prij [1995 a], several concepts for the final disposal of radioactive waste in rock salt are available. Two main differences exist in these concepts, the duration of the interim storage (before definitive disposal), and the application or not of a thick steel overpack to the High Level Waste (HLW) canisters. The longer the interim storage time, and the thicker the steel overpack in the canisters, the lower the amount of energy which will be emitted as gamma rays by the canisters, and of course the lower the radiolytic effects in the rock salt.

Our studies and conclusions are mainly based on short interim storage times, and on disposal of the vitrified HLW-canisters without an overpack, in deep dry-drilled boreholes. The conclussion are also applicable to the repository concepts based on the direct disposal of spent fuel, but it has to be taken into account that only the damage produced by gamma rays has been studied by us and therefore no conclussions can be drawn regarding the consequences of other possible emmisions (e.g.neutrons).

The radiolytic effects of such types of waste disposal will, to ease this exposition, be divided into two groups, the radiation damage in the form of crystal defects and the radiolytic gas production. First we will give the conclusions of our study of the radiation damage (stored energy) in the crystals of the salt rock and then those of the study of the radiolytic gas production.

2. CONCLUSIONS REGARDING RADIATION INDUCED STORED ENERGY IN ROCKSALT

Gamma irradiation can damage the lattice of the halite crystals which constitute a rock salt leading to partial decomposition of the halite (NaCl) into very small particles (colloids) of Na-metal and interstitial chlorine atoms. The energy stored in these defects can be liberated when the sodium and chlorine recombine to reconstitute the undamaged lattice.

As calculated, and experimentally shown in the Brine Migration Test (BMT-experiment [Gies et al.1990]) at the Asse Mine, the radiation damage in halite is a local phenomenon in rock salt repositories. The damaged area is located in the direct vicinity of the boreholes in which the vitrified High Level Waste is placed. As a conseqeunce of gamma ray absorption by the rock salt, the area affected by radiation damage, is limited to the first half meter radius around the container. Within this area maximum damage is found in the salt directly in contact with the container, but the damage decreases radially away from the canisters following a very steep gradient.

The safety consequences of an explosive back reaction which would suddenly liberate all the energy stored in a rock salt were calculated. In these calculations an hypothetical decomposition of the salt crystals (into Na-colloids and Cl) of 20 mol % was assumed. The results of these calculations [Prij 1991] show that such an explosion will not threaten the containment of the waste. Anyway, at present these calculations have lost their actuality since we have shown that the damage in the salt will not reach the required levels for explosions (a minimum 12 mol % decomposition). This level cannot be reached because damage saturation which we have proven to occur in natural rock salt samples, will be attained at a very low level

of damage.

The processes which, by enhancing damage anneal, produce this radiation damage saturation, are expected to be more efficient in a repository where dose rate will even be lower than that used by us. Moreover, the total dose applied by us to show the existence of this saturation, is a factor 4 higher than that which could possibly be reached in the considered repositories. Therefore it is very unlikely that a maximum value of damage higher than that of our experimentally obtained saturation, will be reached in a repository. It could very well be that, taking intercrystalline processes into account (e.g. fluid assisted recrystallization), radiation damage development would stabilize even before nucleation of Na-metal colloids would be completed .

The existing mathematical models used to predict damage build up in prospective repositories were, at the beginning of this research, tested against values of damage measured in our experiments [De las Cuevas and Miralles, 1995] and found to qualitatively reproduce the stored energy build up, but quantitatively yield an overestimation. However, the dependence of damage efficiency on dose rate which was found in our experiments is also quantitatively the same as simulated using the Jain-Lidiard model in the version published in 1985 [Donker and García Celma, 1995]. The models have further been improved on the basis of our experimental results, and now all the intracrystalline phenomena observed by us can also qualitatively be reproduced by them. However, the values of damage calculated for the experiments and for prospective repositories using the different versions of the models are very similar to each other. We are therefore satisfied to accept that the models, even as they were at the beginning of this research, can reasonably be used to calculate worse cases in a repository situation.

We can therefore say that *the radiation damage in the rock salt of a repository as a consequence of the emplacement of vitrified high level waste canisters without a thick steel overpack in dry-drilled deep boreholes does not constitute a safety problem.*

3. RADIATION-INDUCED GAS FORMATION

The γ-radiation-induced gas production from rock salt was investigated in several laboratory studies using Co-60-sources and spent fuel. In these experiments γ-radiation doses of up to 10^8 Gy were employed. The temperature varied between ambient temperature and 250 °C. In some experiments the rock salt samples were also subjected to confining pressures of up to 200 bar. To estimate the significance of the experimental parameters they are compared with those of the envisaged disposal concept : while the temperature range for the borehole disposal concept was fully covered by the experiments, the experimental radiation doses do not include the high dose region. The following conclusions can be drawn.

The gas atmosphere in which the rock salt is irradiated has a significant influence on the product composition. When the irradiations were carried out in synthetic air, N_2O, CO, CO_2 as well as some H_2 were detected. But in the absence of oxygen H_2, CH_4 and CO_2 were observed as gaseous products, while no CO nor N_2O was detected.

Radiolysis of the borehole air leads to the formation of N_2O, a relatively innoxious gas, and some NO_x. Radiolysis can be expected to consume the oxygen in the borehole so that anaerobic conditions will prevail after some time.

Small amounts of corrosive, toxic and explosive gases will be formed. Hydrogen is formed via radiolytic decomposition of water and also as a product of the reaction of methane with oxygen. The hydrogen yields are decreased by one order of magnitude in the presence of oxygen.

Methane, CH_4, is formed by thermal and probably also by radiolytic decomposition of organic matter. In the presence of oxygen, carbon monoxide and carbon dioxide are formed.

The amount of chlorine gas corresponds to the amount of colloidal sodium formed. However, most of the chlorine seems to remain trapped in the halite crystal and is not released into the borehole.

The influence of gamma dose rate on the gas production has not been investigated in any great detail. However, the existing data do not point to any significant effect.

All data on the γ-radiation-induced gas formation show, that the contribution of this process to the total source term of gas production in an emplacement borehole for vitrified high level radioactive waste should be small and does not represent a safety problem.

Other processes such as the formation of hydrogen via corrosion and the pressure increase due to the creeping of the salt are much more important for the safety considerations.

4. RECOMMENDATIONS.

4.1. Radiation damage in the halite crystals

Usually, as in our case, at the end of a project of a technical/scientific nature, and due to the new insights developed by the research work itself, some loose ends can be identified. In our case, our reasonable contentedness with the results regarding the stored energy saturation could be improved by showing experimentally that at lower dose rates damage saturates at values even lower than those actually found. Besides that, starting from our confidence in the qualitative correctness of our improved mathematical models for radiation damage simulation, further refinement of the model parameters themselves would produce a better quantitative simulation of damage, instead of the till now produced overestimation. And, of course, we still miss the knowledge on the effect of low temperatures at low dose rates which was planned to be obtained from the HAW-test field experiment. However, a continuation of research work in this area cannot be justified by arguments regarding safety aspects of radiation damage in repositories of the type considered.

4.2. Thermal and Radiolytic gas production and/or release.

The release and/or the in situ trapping of the gases generated in an underground nuclear repository is mostly determined by the hydraulic properties of the host rock and of the geotechnical barriers used to seal disposal areas (boreholes, drifts and chambers) in the repository. For the long-term prediction of the safety-consequences of the gas production (e.g. pressure and concentration) coupled mechanical / hydraulical computer models are needed.

To produce the computer models suitable constitutive equations are required. These equations have to describe the mechanical and hydraulical behaviour of the materials through which the gases are transported. The parameters used in the constitutive equations have also to be known. In the case of the transport of gases in a repository the important parameters are the porosity and permeability of the host rock and of the sealing materials used in the geotechnical barriers and the dependency of both parameters on the time dependent temperature and stress fields in the repository.

These coupled computer models are not yet available and only a few data are available regarding mechanical/hydraulical behaviour of the disturbed zones around the emplacement openings in the repository and regarding the behaviour of candidate sealing materials in the geotechnical barriers. Therefore, it is strongly recommended to intensify the research effort directed to provide these tools and data in a reasonable period of time.

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