

# RADIOACTIVE WASTE REPOSITORY RELEVANT PARAMETERS

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## ABSTRACT

This article explains which are the main parameters affecting radiation damage development in radioactive waste repositories in rock salt formations. Information is given on the values these parameters would take in some generic conceptual designs of a radioactive waste repository. The intention of this article is to provide a reference framework to allow evaluation of the relevance of the experimental work on radiation damage presented in this volume.

## 1. INTRODUCTION

Studies of radiation damage in rock salt show that the amount of damage in the salt depends on the total dose and dose rate of the irradiation, on the temperature at which the salt is irradiated, and on the nature and amount of intra crystalline defects. The production of gases due to the irradiation depends, besides the above given parameters, on the content of inter-crystalline impurities of the salt e.g. brine and organic matter contents.

The dose rate and temperature rise in a repository with radioactive waste canisters decrease with increasing distance to the emplaced waste. The total dose obviously increases with increasing time. These variations act in different ways on the halite and other components of the rock salt to finally produce damaged salt and gases. The way in which the different values adopted by these parameters act on the salt is subject of other articles in this volume [de las Cuevas et al, Mönig et al, Donker et al, Garcia Celma et al.]. In this article only the direct manifestations of the activity of the waste: heat and radiation output producing temperature rise and gamma dose rates will be considered.

The evolution of the heat and dose rate output of the nuclear waste are related to each other since they both depend on the nuclide inventory (mass or activity of the different nuclides) of the waste. The nuclide inventory of the high-level waste can be calculated rather accurately using burn-up calculations as the high-level waste originates from the fuel elements used in nuclear power plants. The nuclide inventory varies depending on the characteristics of the reactor where the fuel was used.

At the moment of planned emplacement, the inventory of a canister also depends on the time elapsed since the fuel was taken from the reactor. Usually this time is considered as consisting of two different periods, the pre-storage period, and the interim storage period. The pre-storage period is the time elapsed between removal of the fuel from the reactor and its reprocessing. The interim storage period is the time elapsed between the reprocessing and the final disposal. Obviously the inventory of a canister will also depend on the reprocessing method and on the amount of waste packed together within it.

The heat and gamma radiation output of the canisters will produce a temperature, dose rate and total dose gradients in the salt surrounding the canisters. These gradients will change in time together with the evolution of the respective nuclide inventories and the mutual interaction between individual canisters. The estimated changes in time and space for all these parameters will be described for some generic repository concepts. First the geometric and strategic elements of the different concepts will be summarized, and then, after discussing the evolution of the temperature rises for different study cases, the evolution of the dose rate and that of the total dose will be considered.

### 1.1 The repository concepts and strategies: definition of study cases

To evaluate the ranges of the parameters some repository designs have been studied. Generic concepts have been selected as the final designs are not yet fixed in the European countries. The concepts consist of a combination of geometries and waste strategies. The geometries refer to the characteristics of the geological formation (bedded salt, salt pillow or salt dome), to the engineered geometry itself, galleries and boreholes, and to the type of canister. The strategies refer to different reactor characteristics and various combinations of

pre-storage and interim storage periods. Sixteen relevant case studies have been considered, see Table I where each case is numbered with a two digit number. These case studies were first defined and analyzed by Heijboer et al. [1988], and by de Haas et al. [1989]. In Table I the characteristics are: the burn up of the fuel in GWatdays per ton uranium (GWd/tU), the amount of energy produced in the nuclear power plants in EJ ( $10^{18}$  J), the pre-storage period and the interim storage period in years, the type of salt formation (FT), and the disposal technique. In both formation types the waste is disposed at the same depth, viz. from 900 m to 1200 m.

Table I: Cases considered for the determination of the repository parameters.

case	Burn up [GWd/tU]	Energy <sup>1)</sup> [EJ]	Pre-storage [a]	Int. storage [a]	FT <sup>2)</sup>	DT <sup>3)</sup>
11	33	0.5	3	50	P	D
12	33	0.5	10	50	P	D
13	33	3.5	3	50	P	D
14	33	3.5	10	50	P	D
15	33	3.5	3	10	P	D
16	33	3.5	10	10	P	D
17	33	0.5	3	50	P	M
18	33	0.5	10	50	P	M
19	33	3.5	3	50	P	M
20	33	3.5	10	50	P	M
21	33	3.5	3	10	P	M
22	33	3.5	10	10	P	M
23	33	3.5	10	10	D	D
24	33	3.5	10	10	D	M
25	40	3.5	10	10	P	D
26	40	3.5	10	10	P	M

<sup>1)</sup> 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.

<sup>2)</sup> FT is formation type.  
P: Pillow with an overburden thickness of 800 m.  
D: Dome with an overburden thickness of 230 m.

<sup>3)</sup> DT is disposal technique  
D: Deep boreholes drilled from the earth' surface.  
M: Mine with galleries and boreholes drilled from the floor of the galleries.

In the mine concepts the boreholes are drilled, using a dry-drilling technique, in the floor of the galleries at regular distance (pitch) from each other. In these boreholes the canisters with vitrified HLW are placed without shielding container. Consequently the impact

of the radiation on the rock salt must be regarded as a maximum. In the mine concepts studied, a salt plug with a length of 0.65 m is placed between adjacent canisters. The plug is used to avoid leakage of the canisters caused by the weight of the overlaying canisters which might lead to an exposure into the borehole. The salt plugs also act as additional barriers against intruding groundwater.

The impact of the radiation can be reduced to almost zero by applying large steel containers in which the vitrified HLW canister is placed as suggested in the PAE study in Germany. The effect of shielding on the radiation damage can clearly be seen in the backup concept in the dutch programme. This so called deep-borehole concept consist of long boreholes drilled from the surface reaching into the salt formation. The drilling technique used is a standard technique using drilling liquid. The HLW canisters are placed in the deep boreholes. Also in this case the canisters are separated by a salt plug. Furthermore the canisters would be provided with an extra steel overpack having a wall thickness of 3 cm. This overpack is used to withstand the hydraulic pressure due to the liquid collum in the hole. This overpack has a considerable effect on the dose rate and total dose.

## 2. TEMPERATURES

Temperature rises in the salt of a repository due to the heat generated by the waste can be calculated by solving the heat conduction equations, provided the proper material parameters are taken into account. For the salt surrounding a canister, the resulting temperature rise depends, besides on the heat output of the individual canister, on its location in the repository and on the size of the repository.

The calculated maximum dose rate and temperature for the 16 cases described in 1.1 are given in Table II. Considering Table I and Table II together it can be concluded that the maximum temperature in the salt directly in contact with the canister,  $T_{\max}$  in Table II, is determined by the duration of the interim storage and quite independent of the pre-storage. The same independence on the pre-storage time is displayed by the maximum dose rate i.e. the initial dose rate in the salt at the canister wall,  $DR_{\max}$  in Table II.

The temperature evolution is qualitatively the same for all cases studied. Therefore two illustrative examples will be considered, viz. cases 19 and 21 presented in Figs. 1 and 2 respectively. In these figures, each line represents the evolution of the temperature in the salt at a radial distance,  $d$ , varying between 0 to 0.2 m to the wall of the waste canister.

Table II: Some resulting temperatures and dose rates

case	$T_{\max}^{1)}$ [°C]	$t_{\max}^{2)}$ [a]	$DR_{\max}^{3)}$ [kGy/h]
11	64.6	5	0.037
12	63.8	6	0.032
13	64.7	5	0.037
14	63.9	6	0.032
15	118.2	5	0.116
16	109.1	5	0.088
17	67.5	11	0.254
18	67.6	14	0.215
19	69.8	14	0.254
20	70.2	17	0.215
21	128.7	13	0.750
22	118.3	13	0.585
23	116.2	5	0.088
24	113.2	9	0.585
25	109.1	5	0.088
26	118.3	13	0.585

- 1)  $T_{\max}$  is the maximum temperature in the salt direct to a canister at the corner of the repository in the middle of the vertical stack of canisters [de Haas et al., 1989] and [Jong, 1987].  
 2)  $t_{\max}$  is the time at which the maximum temperature is reached.  
 3)  $DR_{\max}$  is the maximum, i.e. the initial, dose rate in the salt directly at the canister wall.

It can be seen (in Figs. 1 and 2) that after emplacement of the waste the temperature in the salt close to the waste canisters rapidly increases. This increase leads to a temperature maximum, at about 10 years after emplacement,  $t_{\max}$  in Table II. This maximum is followed by a gradual decrease until the undisturbed initial salt formation temperature is reached, more than thousand years after disposal.

The temperature rise as calculated for the salt surrounding a given borehole is caused by the heat generation of the waste emplaced in the considered borehole and by that of the waste emplaced in the neighbouring boreholes. The latter is referred to as the field effect. It takes some time before the heat from one borehole reaches the neighbouring borehole, the

field effect is noticed after a period of time has elapsed. It would be possible to design a repository where the temperature maximum would have been reached before the field effect is considerable. It would suffice to allow for a pitch between the boreholes larger than about 100 m [Jong, 1987]. In that case the maximum temperature would be reached after 5 years. The mine concepts consider a pitch of 50 m, and therefore the maximum temperatures are affected by the neighbouring boreholes, and the maximum temperatures are reached after 10 years. In the deep borehole concepts the pitch is 100 m which implies that the maximum temperature is reached after 5 years.

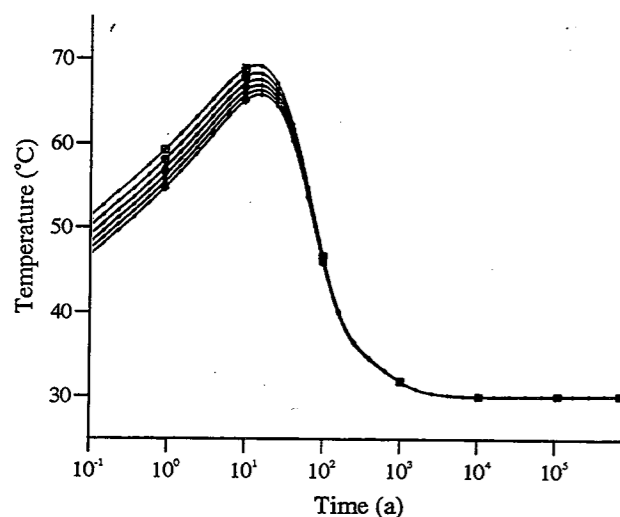


Fig. 1. Evolution of the temperature (case 19; interim storage 50 years)

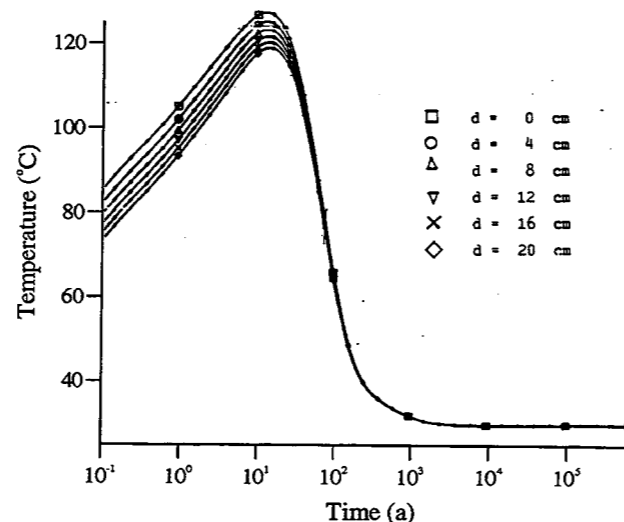


Fig. 2. Evolution of the temperature (case 21; interim storage 10 years)

The temperatures given in Table II correspond to those of the salt at the boundary of a repository where the field effect is limited. In the centre of a repository the field effect is the highest as there are the highest number of neighbouring boreholes. At that location the maximum temperature which would be obtained for case 21 would be about 150 °C, and for case 19 the maximum would be about 80 °C [Jong, 1987].

In the mine concepts, a salt plug with a length of 0.65 m, is placed between the individual canisters, as a consequence the average heat production per meter borehole is 67.5 % of that produced by a meter of canister. The temperature analyses have been made

with average heat production values, which implies that locally the temperatures can be somewhat higher or lower than the values given. The deviation from the maximum values given is  $\pm 3.5$  °C for case 19 and  $\pm 9$  °C for case 21 [Jong, 1987].

Due to the interacting effects of the canisters in the stack and due to the initial geothermal gradient there is a vertical temperature gradient along the boreholes. The temperature rise in the salt around the top canister of the stack has been calculated to be about 50 % of the temperature rise in the salt around of the middle canister of the stack [Jong, 1987]. A geothermal gradient of  $\sim 0.02$  °C/m was taken into account in the calculations.

From all the above described vertical and horizontal gradients along and around a borehole, and the field and geothermal gradients as well, it turns out that the temperature at a given canister can vary considerably depending on its position in a repository and in the stack within the borehole. Table III gives the intervals within which the maximum temperatures can vary.

Table III: Spatial variation in the maximum temperature [°C]

location in the repository	case 19	case 21
	$t_{\text{int}} = 50$ [a]	$t_{\text{int}} = 10$ [a]
corner; top of the stack	50	90
corner; middle of the stack	75	140
centre; top of the stack	60	100
centre; middle of the stack	80	160

In the German mine concepts there is no salt plug between the individual canisters. This implies that the temperature rise at all locations would be about 50% larger than in the concepts with the 0.65 m plug. Due to the absence of the salt plug the range of the maximum temperature for an interim storage period of 10 years will be 120 to 220 °C while the range in the maximum temperature is 60 to 105 °C for an interim storage period of 50 years. These ranges can be slightly larger due to the smaller pitch between the boreholes used in the German concepts as compared to the other concepts.

### 3. DOSE RATE

Maximum dose rate values at the wall of the canister for different study cases have been given in Table II. The evolution of the dose rate for some study cases is represented in Figures 3 to 6. Figures 3 and 4 represent the study cases 13 and 15 of the deep borehole concept while Figures 5 and 6 represent the study cases 19 and 21 of the mine concept. Just as in the case of the temperatures presented in Figs. 1 and 2, the dose rates received by the

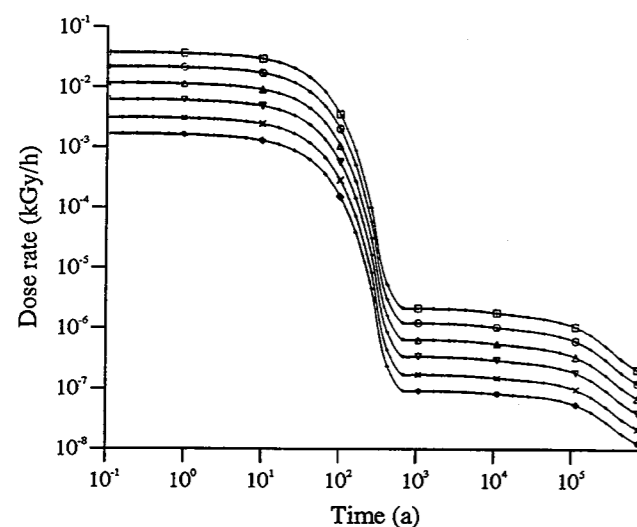


Fig. 3. Evolution of the dose rate (case 13; deep boreholes,  $t_{int} = 50$  years).

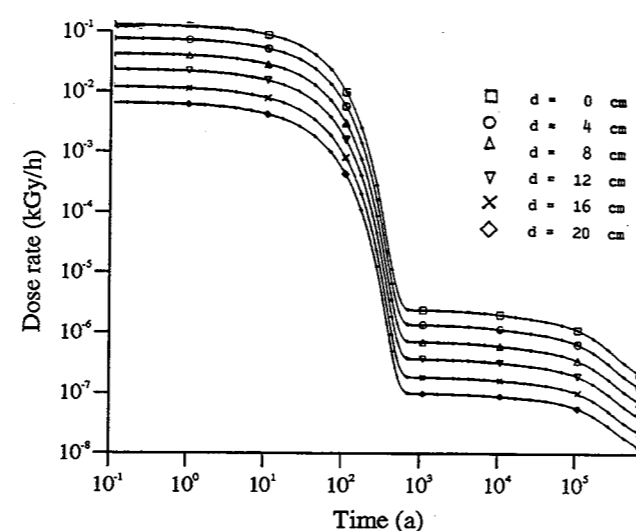


Fig. 4. Evolution of the dose rate (case 15; deep boreholes,  $t_{int} = 10$  years).

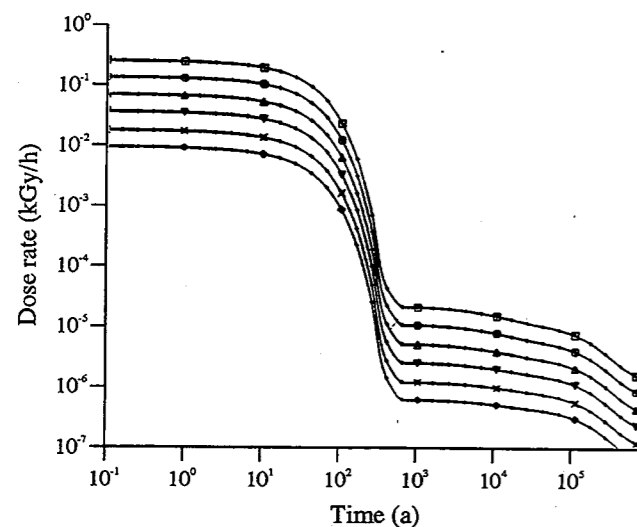


Fig. 5. Evolution of the dose rate (case 19; mine,  $t_{int} = 50$  years).

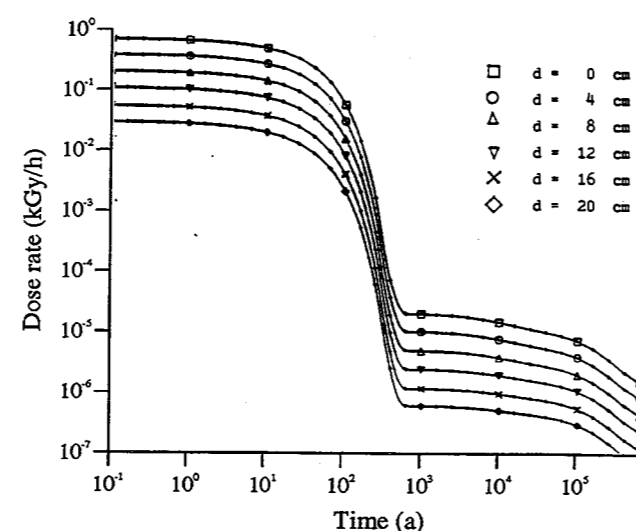


Fig. 6. Evolution of the dose rate (case 21; mine,  $t_{int} = 10$  years).

salt are given for different distances from the canister wall. Considering these figures and Table II it can be concluded that the 3 cm thick steel overpack reduces the dose rate with a factor of 6.8. The temporal and spatial distribution is qualitatively the same for all case studies. In the first 500 years after disposal the evolution of the dose rate is controlled by the decay of the nuclides  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , nuclides which have a half life value of 30.0 respectively 29.1 years. During these first 500 years the dose rate will decrease by about 4 orders of magnitude. Afterwards, the evolution of the dose rate will be controlled by the long lived fission products and actinides.

In the German concepts, the dose rate is not expected to essentially deviate from that calculated for the Dutch concepts, because the dose is not determined by the repository design but only by the inventory of each individual canister. There is no interaction between boreholes or canisters as the radiation is much more absorbed and not conducted as the heat.

### 4. TOTAL DOSE

The total dose evolution logically depends on that of the dose rate. The expected values of the total dose in the salt close to the canister are given in Table IV. Just as was the case for the dose rate, the most important variations on its value are introduced by the

Table IV: Evolution of the total dose [MGy]

case	time after disposal [a]					
	$10^1$	$10^2$	$10^3$	$10^4$	$10^5$	$6 \cdot 10^5$
11	2.9	12.6	14.0	14.1	15.4	17.6
12	2.5	10.7	11.9	12.0	13.3	16.0
13	2.9	12.6	14.0	14.1	15.4	17.6
14	2.5	10.7	11.9	12.0	13.3	16.0
15	8.4	33.5	37.0	37.2	38.4	40.7
16	6.7	27.8	30.8	31.0	32.4	35.0
17	19.8	86.0	95.5	97.1	106.1	122.5
18	16.8	73.0	81.1	82.8	92.3	112.1
19	19.8	86.0	95.5	97.1	106.1	122.5
20	16.8	73.0	81.1	82.8	92.3	112.1
21	55.3	225.8	249.6	251.2	260.2	276.6
22	44.9	188.8	209.1	210.7	220.2	240.1

overpack and the variations in interim storage duration. The maximum total dose would be about 120 MGy (or 12 Grad) for an interim storage of 50 years, while it will be about 270 MGy (27 Grad) for an interim storage of 10 years. In cases where an overpack of 3 cm is applied the values are about 18 MGy and 40 MGy for the interim storage of 50 and 10 years respectively.

The total dose that the salt would receive during the first thousand years after emplacement would be 70% of the total yield of gamma radiation in the cases in which an interim storage of 50 years is applied. If the interim storage is only 10 years, the dose received in the first thousand years after emplacement would be about 90 % of the total yield of gamma radiation. The missing 20% is mainly due to the decay of the  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  during the interim storage period.

## 5. CONCLUSIONS

The results of the case studies calculations show that the pre-storage duration and the burn-up only have a very limited influence on the temperature rises in the salt around the canister. The temperature rise is most affected by the interim storage and the location in and the size and design of the repository. For an interim storage of 10 years the maximum temperature rise varies between 90 and 160 °C in the conceptual designs with a salt plug between the individual canisters. In the conceptual designs without such a salt plug the variation in the maximum temperature rise will be between 120 and 220 °C. In case of an interim storage of 50 years the temperature rises are smaller, the maximum varies between 50 and 80 °C in the designs with a salt plug and between 60 and 105 °C in the designs without a salt plug. The temperature rise is not influenced by the overpack.

The pre-storage time and burn-up have a small effect on the dose rate. The dose rate in the salt surrounding the canister is not dependent on the location of this canister in the considered designs of a repository. Dose rate and total dose are strongly influenced by the interim storage and the overpack thickness. For the cases without an overpack the initial dose rate varies between 0.21 and 0.75 kGy/h, for the cases with an overpack of 3 cm the initial dose rates vary between 0.03 and 0.11 kGy/h. The low values are for the long interim storage

and long pre-storage period while the higher values refer to the short interim and pre-storage period.

The total dose varies between 16 and 41 MGy for the overpack cases while the variation in the total dose in the mine cases is from 112 to 277 MGy. Here again the low values refer to the long interim storage period while the high values refer to the short interim storage. The duration of the pre-storage has only minor influence on the total dose.

Besides the wide ranges in the maximum values for the temperature rise, dose rate and total dose the study cases have made clear that these parameters are strongly time dependent. Furthermore they are strongly interrelated which means that for accurate mathematical simulation of the built-up of radiation damage this interrelation and time dependency should be included. At each location around a canister the conditions of the salt are characterized by a unique time dependent set of parameter values for the temperature, dose rate and total dose. These dependencies imply that one must be very careful in extrapolating the experimental results obtained for constant dose rate and constant temperature to a repository situation with the same total dose. These dependencies can be elucidated with 'trajectories' in the temperature-dose rate domain, see Figures 7 and 8. In these figures the trajectories start at the

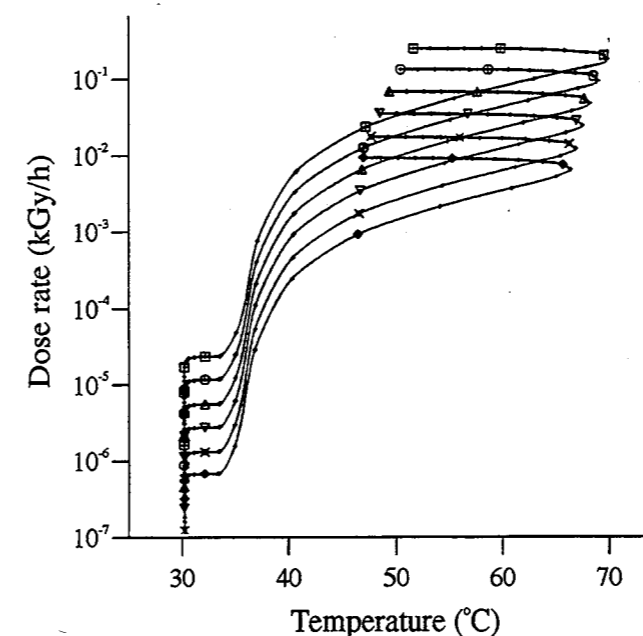


Fig. 7. Trajectory in the temperature and dose rate domain (case 19; mine,  $t_{int} = 50$  years).

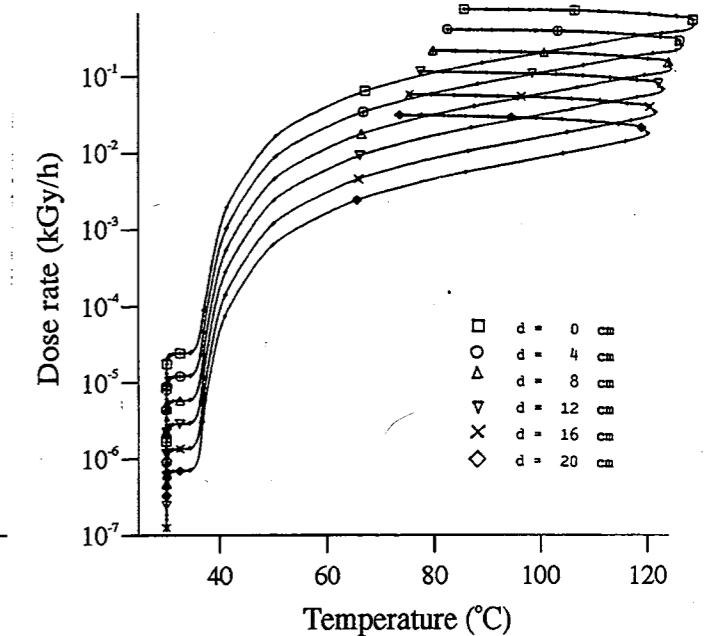


Fig. 8. Trajectory in the temperature and dose rate domain (case 21; mine,  $t_{int} = 10$  years).

'top of the figure', with the high dose rate and stops at the 'bottom' with a very low dose rate and low temperature. Along each trajectory the time is indicated with markers starting with  $10^{-1}$  [a]. The 'distance' between the markers is a factor 10.

## REFERENCES

- CUEVAS, C. de las, & L. MIRALLES, 1995: "Colour centres development by gamma irradiation of natural and synthetic rock salt samples", article in this volume.
- DONKER, H & A. GARCIA CELMA, 1995: "Stored energy in irradiated natural rock samples as compared to synthetic halite of different characteristics". Article in this volume.
- GARCIA CELMA, A., W. SOPPE, & H. DONKER, 1995: "The effect of crystal defect density gradients on radiation damage development and anneal". Article in this volume.
- HAAS, J.B.M. de, HELMHOLDT, R.B., 1989: "Stralingschade rond KSA containers in steenzout". ECN report ECN-89-23, Petten, The Netherlands.
- HATTUM van, and BLANKEVOORT, 1986: "Location-independent study into the excavation, operation, and connection of possible facilities for the definitive disposal of radio-active waste in rock salt formations in the Netherlands".
- HEIJBOER, R.J., HAAS, J.B.M de, DALEN, A. van, BENNEKER, P.B.J.M., 1988: "Nuclide-inventaris, warmteproductie en gammastraling van kernsplijtingsafval". VEOS eindrapportage deelrapport 11, maart 1988, ECN Petten, The Netherlands.
- JONG, C.T.J., 1987: "Temperatuurberekeningen". VEOS eindrapportage deelrapport 11, juli 1987, ECN Petten, The Netherlands.
- MÖNIG, J., N. JOCKWER, H. GIES, 1995: "Colloid formation and stored energy deposition in irradiated natural salt samples". Article in this volume.

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## THE THEORY OF RADIATION DAMAGE IN SALT CRYSTALS AND ROCKS, THE LEADING QUESTIONS AND UNDERLYING RESEARCH THESES

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## ABSTRACT

Taking into account repository concepts for final disposal of radioactive waste in salt formations consisting of long vertical boreholes where the vitrified HLW-canisters devoid of an extra overpack are emplaced, a methodology was developed to prove or disprove some essential theses. Some of these theses constituted the basis of pessimistic estimations regarding the safety of these concept repositories. The theses refer to the accumulation of stored energy in the form of lattice defects in the crystals of the rock salt due to  $\gamma$ -irradiation. Following these estimations accumulation of crystal defects could bring about spontaneous explosive back reactions which would threaten the containment of the waste. This article shortly introduces these theses, explains the methodology applied to be able to prove or disprove them, summarily presents the experimental and theoretical results obtained following the described methodology and, after discussing the veracity of each of the theses concludes that no scientific basis has been found to assume that spontaneous explosive back reactions would take place in eventual repositories built according to the considered concepts.

## 1. INTRODUCTION

In 1989 an "International Test Plan" [Mönig et al., 1990] was formulated, which coordinated the different ways in which some remaining questions regarding radioactive waste disposal in rock salts would be tackled and which settled the responsibilities of the different partners. The "International Test Plan" document establishes a series of irradiation experiments directed to answer remaining questions regarding on the one hand the problem of gas development in repositories and on the other hand the problem of radiation damage development.