THE HAW-PROJECT AND ITS CONTRIBUTION TO THE INVESTIGATION OF RADIOLYTIC EFFECTS IN ROCK SALT

T. Rothfuchs

ABSTRACT

In 1985 activities were started for a test disposal of highly radioactive radiation sources in the Asse salt mine in Germany. The objectives of the HAW-project were to develop and to test a transport and emplacement system for the handling of high-level radioactive waste (HAW) and to investigate the interaction between the radioactive canisters and the surrounding rock salt formation.

Within the framework of the R & D programme of the project special emphasis was given to the investigation of radiation damage in rock salt.

Besides the planned in situ irradiation of the rock salt in the test field and of special rock salt samples contained in dummy canisters in the emplacement boreholes an intensive laboratory irradiation programme was agreed. The laboratory irradiations were carried out at Petten (NL), Saclay (F) and Barcelona (E). In the framework of a Multi Partner Contract with the Commission of the European Communities (CEC) the project was carried out by the "GSF-Institut für Tieflagerung (GSF/IfT) Braunschweig (FRG)", the "Energieonderzoek Centrum Nederland (ECN) Petten (NL)", the "Agence Nationale pour la Gestion de Déchets Radioactifs (ANDRA) Paris (F), and the "Empresa Nacional de Residuos Radiactivos S. A. (ENRESA) Madrid (E).

On December 3rd, 1992, the German Federal Government, represented by Bundesministerium für Forschung und Technologie (BMFT) and Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit (BMU), has decided not to perform the test emplacement of the 30 high-level radiation sources and to stop all preparatory activities.

In order to fulfil the Multiple Partner Contract with the CEC a revised research programme was negotiated among the project partners. This programme involved the termination of the in situ activities in 1993 and the continuation of the laboratory investigations as originally foreseen until the end of 1994.

1. INTRODUCTION

In order to improve the final concept for a high-level radioactive waste (HAW) disposal in salt formations plans were developed a couple of years ago in Germany for a full scale testing of an underground HAW repository.

To satisfy the test objectives thirty isotopic heat and radiation sources containing the radionuclides Cs-137 and Sr-90 in quantities sufficient to cover the bandwidths of heat generation and gamma radiation of real HAW were planned to be emplaced in six boreholes located in two test galleries at the 800-m-level in the Asse salt mine. A duration of testing of approximately five years was envisaged. The project was named the HAW-project. Detailed information on the HAW-project e. g. the layout of the test in the Asse mine can be taken from (CEC, 1991) and (CEC, 1993). An accompanying scientific investigation programme included the observation of the interaction between the isotopic heat and radiation sources and the surrounding rock salt. The project was a joint effort of the "GSF-Institut für Tieflagerung (GSF/IfT) Braunschweig (FRG)", the Energieonderzoek Centrum Nederland (ECN) Petten (NL), the "Agence Nationale pour la Gestion de Déchets Radioactifs (ANDRA) Paris (F)", and the "Empresa Nacional de Residuos Radiactivos S. A. (ENRESA) Madrid (E)".

One of the major objectives of the project was the investigation of the impact of gamma radiation from HAW canisters on natural rock salt which is known from the literature as "Radiation Damage in Rock Salt". The impact of gamma radiation on rock salt is characterized by a radiolytical decomposition of the solid rock salt as well as by a radiolytical decomposition of volatile components contained in the pore space of the rock salt. The generation of gases as a consequence of the decomposition of salt minerals is to be considered too. The most important effects that can be measured and used for the quantification of radiation damage in salt are the formation of collodial sodium, the development of chlorine gas and the storage of energy resulting from the radiolytical decomposition of NaCl and the accompanying deformation of salt crystals.

In order to coordinate the scientific work of this objective within the HAW-project an international test plan was set up by the project partners (Mönig et al., 1990).

The test plan considered in situ irradiation experiments as well as laboratory irradiation experiments. Because of licensing uncertainties, however, the German Federal Government, represented by Bundesministerium für Forschung und Technologie (BMFT) and Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit (BMU), has decided on December 3rd, 1992, not to perform the test emplacement of the 30 high-level radiation sources and as a consequence no in situ-irradiation experiments were performed. Some of the in situ-experiments, however, were compensated by additional laboratory irradiation experiments at the High Flux Reactor (HFR) in Petten.

2. THE TEST FIELD IN THE ASSE MINE AND THE PLANNED IN SITU IRRADIATION EXPERIMENTS

The test disposal in the Asse salt mine was designed to satisfy the following points:

* Representative heat generation rate of the radioactive canisters
* Representative ionizing radiation from the radioactive canisters
* Disposal and thermomechanical conditions representative for a nuclear repository.

According to the German disposal concept the conditions of a HAW repository can be summarized as follows: The permitted maximum salt temperature at the disposal borehole wall is 200 °C and the initial dose rate in air at the canister surface is assumed to be about 0.5 - 1 kGy/h.

Since the licensing conditions do not permit any final disposal of radioactive material in the Asse salt mine, the safe retrievability of the radioactive canisters was to be guaranteed. Therefore, the emplacement boreholes in the test field were lined with high-strength steel tubes to avoid clamping of the radioactive sources. As a consequence the outer surface of the liner would have represented the outer surface of radioactive canisters during testing in the Asse mine. Accordingly, the activity of the sources was increased compared to high-level waste canisters to compensate the shielding of the steel liner.

The radioactive canisters necessary to carry out the test disposal were fabricated by the Battelle Pacific Northwest Laboratories (PNL) in the framework of a contract between the US Department of Energy and the BMFT. Three different types of canisters were fabricated to enable an irradiation of the rock salt with different gamma dose rates at different temperatures. The average values of the radioactive canisters as reported by PNL in June 1988 (Holton et al., 1988) are presented in Table 1. It can be seen that the surface dose rates of the canisters are considerably above those to be expected from real waste canisters and the shielding by the borehole liner would have been compensated as desired.

Mining of the test field was started in March 1985 and finished in October 1985. The configuration and dimensions of the test field (Fig. 1) were mainly determined by the number of boreholes that were required to perform the test. The test field consists of a set of eight boreholes located in two parallel galleries (A and B). The distance between the boreholes is 15 m in the direction of the gallery and 19 m in the direction perpendicular to the gallery. The test galleries can be reached by two access drifts (also for reasons of ventilation) coming from the south and the west.

Two types of emplacement boreholes are distinguished and named type A and type B (see gallery designation in Fig. 1).

Table 1: Average values of the highly radioactive canisters; canisters filled between February 1986 and March 1987
Canister dimensions: height 1154 mm; outer diameter 298.5 mm;
Contents: 60 l borosilicate glass

Type of Can-ister	Average Cs 137 Content* (PBq)	Average Sr 90 Content* (PBq)	Average Gamma Dose Rate (in Air)* (kGy/h)	Average Heat Output of Canister** (Watts)	Maximum Salt Tempera-ture (°C)**
I	2.83	5.34	0.93	1335 (≈ 22 W/l)	160
II	7.15	3.10	2.28	1490 (≈ 25 W/l)	180
III	7.64	4.83	2.51	1860 (≈ 31 W/l)	230

* Measured during canister fabrication
** Calculated on basis of the measured radionuclide content

In the boreholes of type A the annulus between the liner and the borehole wall was backfilled with ceramic alumina beads to keep the annulus porous thereby representing the early stage in a repository. In the boreholes of type B the salt was permitted to creep onto the liner surface thereby representing the long-term conditions in a repository.

As a result of the heat output of the radioactive canisters two boreholes in each gallery would have reached a maximum salt temperature of 230 °C and two of 180 °C. For reasons of comparison two of the 230-°C-boreholes (A1 and B1) were heated only electrically whereas each of the two others should have been charged with five radioactive canisters of different activity and heat power. In this manner it would have been possible to investigate the impact of the gamma radiation at different maximum salt temperatures.

A dummy canister would have been placed on top of the five radioactive canisters (see Fig. 2). This dummy was not only meant to serve for radiation shielding upwards, but it was also designed to contain chambers for placing salt specimens which should be exposed to radiation at elevated temperatures as shown in Table 2.

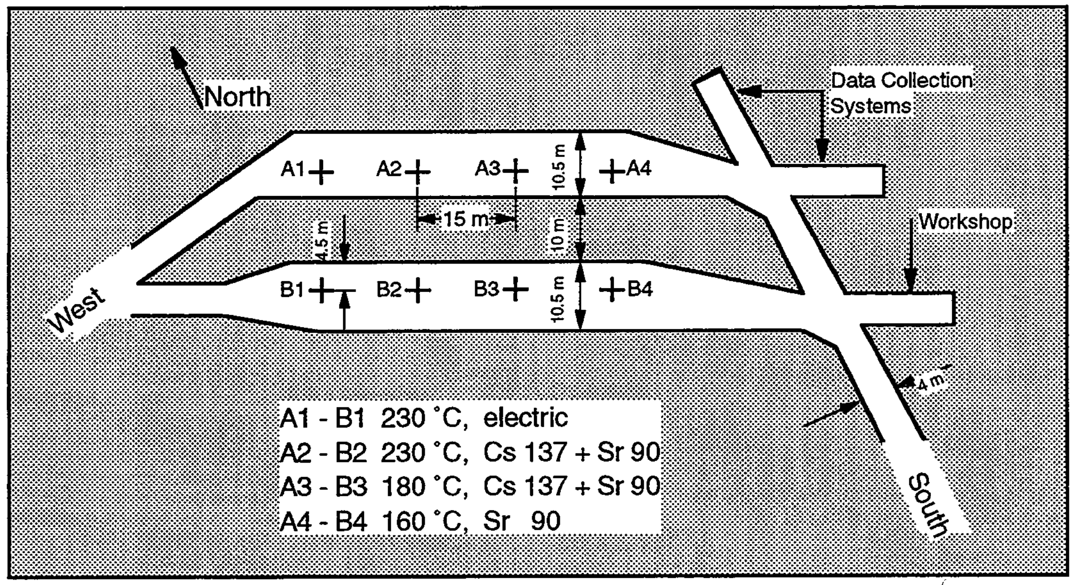

Figure 1: *Layout of the test field and arrangement of the emplacement boreholes*

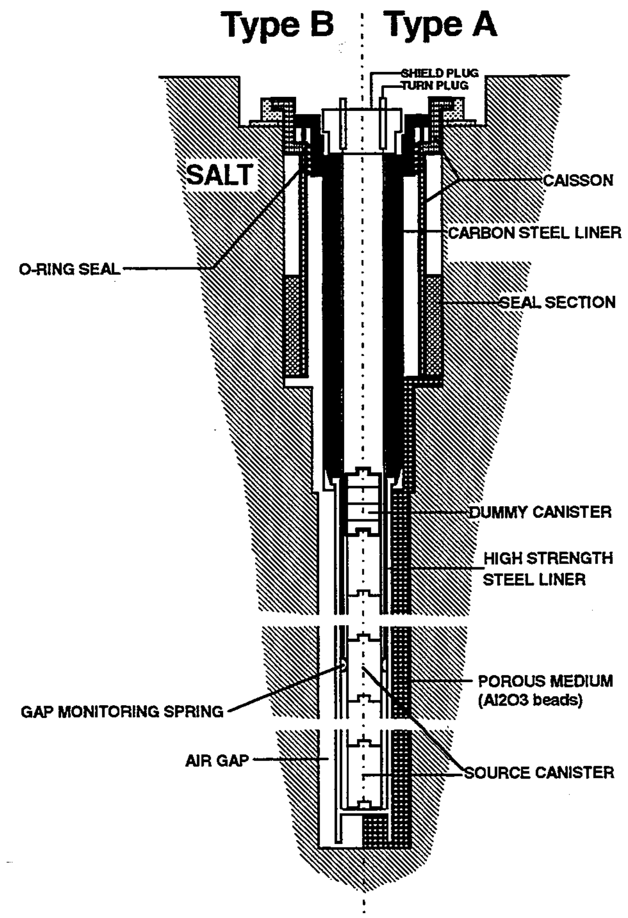

Figure 2: *Emplacement borehole; right side: type A, left side: type B*

Table 2: Calculated dose rates and temperatures at the various levels inside the dummy canister

Level	Dose Rate [Gy/h]	Temperature
L1	10^3	150 °C for A2 and B2 125 °C for A3 and B3 120 °C for A4 and B4
L2	10^2	125 °C for A2 and B2 108 °C for A3 and B3 100 °C for A4 and B4
L3	10	100 °C for A2 and B2 90 °C for A3 and B3 85 °C for A4 and B4

3. LABORATORY IRRADIATION EXPERIMENTS

Three different laboratory irradiation programmes were agreed to by the project partners.

3.1 Investigation of radiolytical gas generation in natural rock salt

This irradiation programme was performed by the commissariat à l'energie atomique (CEA) in Saclay. The irradiations were performed using spent fuel elements from the OSIRIS reactor. In the irradiation experiments ground salt samples from the Asse salt mine were irradiated. The samples were encapsulated in pyrex glass ampoules under an atmosphere of 80 % N_2 and 20 % O_2. The resulting amounts of radiolytically generated gases were determined.

3.2 Investigation of colloid development and stored energy

This irradiation programme was performed in two gamma irradiation facilities GIF A and GIF B at the High Flux Reactor in Petten (the Netherlands) and aimed to determine the degree of radiation damage in rock salt by the determination of the amount of stored energy and of the concentration of radiation induced defects. The programme was performed by the Energieonderzoek Centrum Nederland (stored

energy) and by the Laboratori d'Investigació en Formacions Salines at the Univercity of Barcelona (radiation defects).

3.3 Investigation of the radiation induced development of collodial sodium, chlorine gas, and of stored energy in natural rock salt

An irradiation programme was performed by the GSF-Institut für Tieflagerung (IfT) on the relationship of collodial sodium, chlorine gas and stored energy in natural rock salt from the Asse mine. The irradiated samples consisted of ground salt from Asse with different mineralogical composition.

The samples were encapsulated in glass ampoules under normal and helium atmosphere and were irradiated with different doses at different temperatures. The irradiations were performed at the HFR in Petten and the samples were evaluated at the IfT in Braunschweig.

Table 3 summarizes the most important test parameters of the three laboratory irradiation programmmes. A similar programme has been performed in irradiated samples of ground salt from the Sallent mine in the CESAR irradiator in Granollers, Spain.

4. CONCLUSIONS

Within the HAW-project an irradiation programme for the investigation of radiolytic effects in rock salt was performed. The programme was subdivided into three major tasks. Task 1 performed by CEA/France considered the generation of radiolytic gases. Task 2 performed by ECN and ENRESA considered the storage of energy in salt crystals and the formation of colloidal sodium. Task 3 performed by GSF considered the equivalent formation of colloidal sodium, chlorine gas and the storage of energy. In all irradiation experiments total doses between 10^4 - 10^9 Gy as expected in a HAW repository have been reached. The maximum irradiation temperature was 250 °C.

In spite of the cancellation of the experiments in the HAW test field many irradiation experiments took place in laboratory irradiation facilities. Even if carried out at dose rates higher than those expected under in situ conditions the most relevant parameters and their relationships could be studied. The objectives of the HAW-project regarding the development of radiation induced defects in rock salt have been satisfactorily accomplished.

Table 3: Test parameters of the laboratory irradiation experiments

CEA-irradiations at Saclay for the investigation of radiolytical gas generation in natural rock salt	
Type of salt	Ground natural rock salt from the 800-m-level of the Asse mine encapsulated in glass ampoules with normal atmosphere (80 % N_2, 20 % O_2).
Radiation source used	Spent fuel elements with different cooling times from a couple of days up to 3 years after they have been discharged from the reactor.
Dose rate	from 1E3 Gy/h to 1E5 Gy/h
Total dose	from 1E3 Gy to 1E7 Gy
Irradiation temperature	from 30 °C to 250 °C
Analyzed gases	H_2, CH_4, other small hydrocarbons, H_2S, Cl_2, HCl, CO, CO_2, NOx
ECN-ENRESA irradiations at the HFR in Petten for the investigation of the colloid development and stored energy	
Type of salt	Solid synthetic and natural salt samples; the natural salt samples originating from the Asse mine and from Spain
Radiation source used	Spent fuel elements from the HFR in Petten
Dose rate GIF A-0 GIF A-1 GIF B-1 GIF B-2 GIF B-3	 2.4E5 to 4E4 Gy/h 2.1E5 to 2E4 Gy/h 1.5E4 Gy/h 4E3 Gy/h 1.5E4 Gy/h
Total dose GIF A-0 GIF A-1 GIF B-1 GIF B-2 GIF B-3	 max. 5.7E8 Gy max. 1.2E9 Gy 2E4, 2E5, 3E5, 4E5, 2.6E6, 4E6, 5.4E4, 1.6E7, 2.4E7 and 4.5E7 Gy 2.2E4, 1E5, 2.1E5, 4.4E5, 1.1E6, 2.2E6 4.6E6, 1.1E7, 2.3E7 and 4.4E7 Gy 2.2E4, 1.1E5, 2.2E5, 4.2E5, 1.1E6, 2.2E6 4.5E6,1.1E7, 2.1E7 and 4.1E7 Gy
Irradiation temperature	100 °C

Table 3 continued

GSF-irradiations at the HFR in Petten for the investigation of the radiation induced development of colloidal sodium, chlorine gas and stored energy	
Type of salt	Ground salt samples consisting of 94.5 wt% halite, 5 wt% anhydrite, 0.45 wt% polyhalite, 0.03 wt% water. The samples are encapsulated in glass ampoules with synthetic air or helium.
Radiation source used	Spent fuel elements from the HFR in Petten
Dose rate	~ 4 · 1E4 Gy/h
Total dose	1E6 Gy, 1E7 Gy, 1E8 Gy
Irradiation temperature 140 °C 150 °C	100 °C 160 °C 130 °C 170 °C 200 °C 250 °C

5. REFERENCES

CEC, 1991: *The HAW-Project: Demonstration Facilit for the Disposal of High-Level Waste in Salt*, Synthesis Report 1985 - 1989, Commission of the European Communities, Nuclear Science and Technology, EUR 13263 EN

CEC, 1993: *The HAW-Project: Test Disposal of Highly Radioactive Canisters in the Asse Salt Mine*, Fed. Rep. Germany, Activity Report May 1990 - December 1991, Commission of the European Communities, Nuclear Science and Technology, EUR 14531 EN

HOLTON, L. K., BURKHOLDER, H. C., MCELROY, J. L., KAHL, L., KROEBEL, R., ROTHFUCHS, T., STIPPLER, R., 1989: *Fabrication of vitrified isotopic heat and radiation sources for testing in the Asse mine*, In: Proc. Waste Management 89, Tucson, Arizona/USA, 26.02. - 02.03.1989 (Eds. Post/Wacks), Arizona Board of Regents/USA, Vol. 1, 575 - 586

MÖNIG, J., GARCIA CELMA, A., HELMHOLDT, R. B., HINSCH, H., HUERTAS, F., PALUT, J. M., 1990: *The HAW-Project - Test Disposal of High-Level Waste in the Asse Salt Mine - International Test-Plan for Irradiation Experiments*, Commission of the European Communities, EUR-Report 12946 EN