J. PRIJ, 1995: "*Radioactive waste repository relevant parameters*", Article in this volume.

D. RAWSON et al, "Post-Explosion Environment Resulting from the Salmon Event," *Journal of Geophysical Research*, **71**, No. 14, 3507, (1966).

W.J. SOPPE, H. DONKER, A. GARCIA CELMA, J. PRIJ, 1994: "*Radiation induced stored energy in rock salt, a review*". J. Nucl. Mat. **217**, 1-31, (1994).

H.W. WILD, 1984: "*Sprengtechnik in der Bergbau, Tunnel- und Stollenbau sowie in Tagebau und Steinbrüchen*", Verlag Glückauf, Essen.

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

A. García Celma, C. de las Cuevas, H. Donker, J. Mönig, J. Prij, T. Rothfuchs & L. Vons.

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 consequence 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.

5. REFERENCES

C. DE LAS CUEVAS and L. MIRALLES, 1995: *"Colour Centres Development by Gamma-Irradiation of Natural and Synthetic Rock Salt Samples"*. Article nr. 16, this volume.

H. GIES, A. GARCÍA CELMA, J.B.M. DE HAAS, L. PEDERSEN and T. ROTHFUCHS, 1990 : *"Radiation Damage in Natural Rock Salt: Results of the Investigations on Irradiated Salt from the Asse Brine Migration Test*. Final Report. GSF- IfT. 8/90 München, Germany.

J. PRIJ, 1991: *"On the Design of a Radioactive Waste Repository*. Ph D. Thesis. Enschede University. The Netherlands. ISBN: 90-375-0261-X.

J. PRIJ, 1995 a: *"Radioactive Waste Repository Relevant Parameters"*. Article nr. 2 in this volume

J. PRIJ, 1995 b: *"Evaluation of the Safety Consequences of Stored Energy in a Repository in Rock Salt"*. Article nr. 23 in this volume

H. DONKER and A. GARCÍA CELMA, 1995: *"Stored energy in irradiated natural rock salt as compared to synthetic halite of different characteristics"*. Article nr.17, this volume.

LISTS OF PARTICIPATING INSTITUTIONS AND AUTHORS.