SUMMARY

Rock salt formations are candidates for the geological disposal of High Level (radioactive) Waste (HLW). The HLW is that category of radioactive waste obtained, usually in a vitrified form, from the reprocessing of the spent fuel of light water reactors where more than 99 % of the radionuclides from these reactors are included. HLW is therefore a heat producing vitrified radioactive waste.

The existing concept designs for radioactive waste repositories in rock salt can be divided in two main groups: those based on including the HLW in thick walled containers, and those which consist of vertical boreholes where the HLW canisters are placed without additional shielding. In the first case it is obvious that radiolytic effects are negligible, provided the shielding of the containers is sufficient. In the second case radiolytic effects have to be expected as the rock salt is directly confronted with the radiation of the waste. Two types of effects can be expected in this last case, the radiation damage in the salt crystals of the rock and the radiolytic and thermal gas production and/or release.

This document deals with the general question whether these radiolytic effects can endanger the containment of the waste in a repository. To answer this question was one of the objectives of the R&D programme of the C.E.C. on Management and Storage of radioactive Waste-part B which was started in 1985. Initially within this programme an in situ irradiation experiment in the Asse salt mine in Germany (HAW project) and a supplementary laboratory irradiation programme were planned. Because of licensing uncertainties the in situ experiment was cancelled in late 1992. However, as a consequence the laboratory experiments were extended. The results of these experiments are described in this document.

Part I of this document starts with an outline of the R&D programme regarding the investigation of radiolytic effects in rock salt. To allow an evaluation of the relevance of the experimental work, this outline is followed by a discussion of the main parameters (temperature, dose rate, total dose), resulting from various conceptual repository designs, which would affect the radiolytic effects in a rock salt repository. In the last two papers of part I the general question is worked out into a number of research theses/questions, which formed the basis of the research programme. The methodology followed to answer these questions is briefly explained and the obtained results are summarily presented.

The laboratory irradiation experiments which were performed at Saclay (F), Petten (NL) and Barcelona (E) are extensively described in part II of this document. At Saclay ground salt samples were mainly γ-irradiated with spent fuel elements from the Osiris reactor to study the radiolytic gas production. Also some irradiation experiments using ^{60}Co sources or an X-ray generator have been performed. At Petten three irradiation facilities using γ-radiation from spent fuel elements of the High Flux Reactor (HFR) were used. Two of these facilities, GIF A and GIF B were used to study the radiation damage development in non-ground salt samples, the third facility was used to study both the radiolytic gas production as well as the radiation damage development in ground salt samples. At Barcelona some irradiation experiments using ^{60}Co sources to study radiolytic gas production have been performed.

In the irradiation experiments not only synthetic but also a numerous amount of natural rock salt samples have been irradiated. A detailed characterization of two rock salt formations where these samples were taken from is given in part III of this document.

Part IV of this document deals with the results of the experiments that were performed to investigate the radiolytic gas production. The papers provide detailed experimental data on the gas yields that are to be expected in a final repository. Since these investigations covered the whole range of experimental boundary conditions with respect to dose, dose rate and temperatures, that are encountered in the vicinity of an emplacement borehole, meaningful conclusions can be drawn.

Radiolysis of the borehole air leads to the formation of N_2O and some NO_x . In this process the oxygen in the borehole is consumed, so that anaerobic conditions prevail after some time. In the absence of oxygen, H_2, CH_4 and CO_2 were observed as gaseous products. Only small amounts of corrosive, toxic and explosive gases will be formed. Hydrogen is formed via radiolytic decomposition of water. The amount of chlorine gas corresponds to the amount of colloidal sodium formed. However, most of the chlorine remains trapped in the halite crystals and appears not to be released into the borehole.

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 are evidently much more important for safety considerations. Since the gas production resulting from corrosion processes was not the subject of the papers in this report some investigations remain to be performed.

In Part V of this document the results of the measurements performed to analyze the radiation damage in the irradiated salt samples and the results of new theoretical developments on the models used to describe the damage formation are given. The radiation damage has been studied by various techniques such as: Light Absorption measurements, measurements of the amounts of H_2 evolved and OCl^- formed upon dissolution of the irradiated samples, Microstructural analysis and Differential Thermal Analysis.

In some cases the experimental results have been compared with calculated results obtained from computer simulations of the experiments using the Jain-Lidiard or Jain-Lidiard related model. These comparisons show that for some low dose experiments the theoretical results underestimate the damage, but generally, and certainly for all high dose ($\geq$ 22 MGY) experiments, the models overestimate the damage. The Jain-Lidiard model predicts that low dose rates enhance damage formation, this has been qualitatively and quantitatively confirmed by our experiments.

One of the most important results of the performed analyses is that a saturation of damage has been found for natural rock salt samples irradiated in GIF A at a level that corresponds to 1.7 mol% of decomposition of the salt.

It is also shown that the conversion factor between stored energy and defect concentrations should be around 80 J/g per mol% defects.

Another important result is that the microstructural observations show that various creep processes, removing radiation damage, have been active not only after but also during irradiation of the samples. Neither these creep processes nor the anneal associated with them are taken into account in the models which, probably due to this, produce an overestimation of damage for high dose and moderated dose rates. In one of the papers it is shown that some of the microstructural observations can be explained by introducing a heterogeneous dislocation distribution into the Jain-Lidiard model.

The Jain-Lidiard model has been extended to include the nucleation stage and the effect of impurities. Computer simulations of the damage expected in a repository using this model show that the damage levels in the rock salt, even very close to the HLW containers, will be limited to a few mol%. The specific stored energy in the rock salt in a repository is therefore expected not to be higher than 300 J/g. At this moment there is no scientific basis to assume that this value will be an underestimation. On the contrary, as indicated above, there are many arguments to assume that the reverse, an overestimation, is very probable.

Finally, in part VI of this document the safety consequences of an eventual release of radiation induced stored energy in a repository are evaluated. The general conclusions and recommendations regarding the consequences of radiolytic effects due to storage of HLW in salt formations are also given.

The general conclusion concerning radiation damage development in the crystals of the rock salt is that it will not constitute a safety problem in the eventuality of geological disposal of HLW in deep boreholes (even without a thick shielding).

PART I

FRAMEWORK