RADIATION-INDUCED GAS FORMATION.
LEADING QUESTIONS

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One of the concepts for the disposal of high level vitrified radioactive waste is based on the emplacement of waste canisters in boreholes drilled from underground galleries into rock salt formations. The rock salt surrounding such an emplacement borehole will be heated up to 200 °C and will be exposed to a γ-radiation dose of some 10^8 Gy within the first 100 years. In addition to the development of radiation damage in rock salt, another effect being of importance for the repository safety is the generation of and the release of gases from both the rock salt and the waste.

The potential consequences of gas formation processes are the build up of gas pressure, the generation of corrosive gas mixtures, and the generation of explosive gases. The implications of these processes for the repository safety are the following:

Build up of gas pressures can lead to fracturing of the rock surrounding the borehole. The integrity of the borehole seal could also be impaired. High gas generation rates and gas pressures provide a potential means of transportation for volatile radionuclides.

Corrosive gases, e.g. HCl or H_2S, can degrade the waste canisters and reduce their lifetime. In the presence of brine this would facilitate leaching of the waste matrix and the mobilization of radionuclides.

The generation of explosive gas mixtures has to be prevented at any account by the conceptual design. It has to be guaranteed that under no circumstances the concentration of certain gases in the gas phase ever exceeds pre-defined safety levels.

Therefore, the generation of gases and the release of gases has to be taken into account for an assessment not only of the operational phase but also of the long-term safety of a repository. This problem has recently been addressed by several scientific work programmes as well as by some conferences [NEA-OECD, 1992].

Several different processes account for the gas generation and contribute to the total source term for gas production in the emplacement of high level radioactive waste, including:

- the gas release from the rock salt at ambient and elevated temperatures,

- the gas generation from the emplaced waste, e.g. via corrosion of the waste canisters, and

- the radiation-induced gas generation.

These processes can be summarized by the following general expression [Brewitz and Mönig, 1992]:

$$rate_{total} = rate_{host} + rate_{waste} + rate_{radiol} \tag{1}$$

with:
$rate_{total}$ = total gas generation rate
$rate_{host}$ = gas release from the host rock
$rate_{waste}$ = gas generation resulting from the emplaced waste
$rate_{radiol}$ = radiation-induced gas generation

This represents a simplified approach, since there are interrelations and the parameters affect each other. However, it facilitates a comparison of these processes for the different disposal concepts.

Table 1: *Overview of the various processes contributing to the total source term for gas generation at ILW and HLW disposal*

contribution to source term	ILW	HLW
host rock	outgassing at ambient tempereatures	outgassing at ambient and elevated temperatures
waste	corrosion, microbial degradation	corrosion, radioactice decay
radiolysis	radiolytic hydrogen generation inside the waste forms	radiolysis of borehole atmosphere, radiolytic decomposition of the host rock (Cl_2-formation)

The extent to which each of these processes contributes to the total gas generation rate depends on various factors, i.e. the waste type, the host rock and the specific disposal conditions. A general overview of the various mechanisms causing gas generation at the disposal of intermediate level waste (ILW) and high level waste (HLW), respectively, is given in Table 1.

Table 1 shows that in case of ILW the gas generation is mainly governed by the waste itself, whereas in case of HLW also the host rock may contribute to the gas generation. In order to be able to design repositories and to assess their long-term safety, it is necessary that all sources contributing to the generation and release of gases are known and that quantitative data on the various processes are available.

In case of vitrified high level waste the most important mechanism for waste-related gas generation is the corrosion of the steel containers, resulting in the generation of hydrogen. This process has gained considerable attention in the past, since hydrogen is a relevant product with respect to the operational safety of the repository (about 4% of it in the atmosphere make up an explosive mixture). The corrosion rates being reported for waste canisters depend on the container material used and the geochemical conditions (temperature, amount and composition of brine). Maximal values for uniform corrosion rates of up to about 0.2 mm/a have been reported for cast steel specimen in contact with brine [Schwarzkopf et al., 1989]. However, the corrosion rates, in general, are one to two orders of magnitude lower. For some container materials, e.g. Cr/Ni-steel, pit corrosion and stress corrosion prevails [Smailos et al., 1990]. It should also be noted that a radiation-induced enhancement of the corrosion rate has been reported for some materials. From this follows that the concept of uniform corrosion rates is not applicable for these types of corrosion processes. A direct and uncritical comparison of corrosion data is not very informative and may even be misleading.

Microbial degradation of the waste or corrosion processes inside the waste do not have to be considered in case of vitrified high level waste. The generation of helium or krypton by radioactive decay is also of minor importance compared to the generation of hydrogen.

Rock salt may contain significant quantities of various gases (e.g. CO_2, CH_4, H_2S, higher hydrocarbons) which are absorbed on crystal boundaries or which are trapped as intra-crystalline inclusions. Additionally gases may occur in fissures. These gases are part of the entire geological environment and their distribution and concentration is being controlled by geological and geochemical processes. Significant amounts of gases can accumulate, depending on the minerals involved and the geological and chemical conditions of the rock salt

formation [Knabe, 1989]. Since these gases can be released into the emplacement boreholes or drifts they have to be accounted for by an appropriate source term. The occurrence of gases in rock salt in various areas in Germany and in different salt horizons was reviewed by Hermann [1988]. Quantitative data from in situ tests performed in the Asse salt mine are given by Brewitz and Mönig [1992] and in the final report for the HAW project [Rothfuchs et al., 1995]. In these investigations it was observed that gases are heterogeneously distributed even within one stratigraphic horizon. Also, gas release is significantly increased by heating the salt. On the basis of these findings, a determination of the gas content at the actual disposal horizon in the final repository seems necessary, in order to avoid those sections of the salt structure for waste disposal which contain very hgh gas concentrations. The uneven distribution of gases has also to be taken into account in the computer models.

In their paper, Brewitz and Mönig [1992] concluded that the radiation-induced reactions, which lead to additional gas generation are least understood, since experimental data were scarce at that time. In the meantime more irradiation experiments have been carried out so that the situation has changed somewhat. The papers in this volume deal with the γ-radiation induced gas formation and the release from rock salt and should provide the necessary data. Gas production from corrosion processes, on the other hand, have not been studied in the framework of this research.

REFERENCES

W. BREWITZ and J. MÖNIG, 1992: "Sources and Migration Pathways of Gases in Rock Salt with Respect to High Level Waste Disposal", in "Gas Generation and Release from Radioactive Waste Repositories", Proc. NEA-Workshop, Aix-en-Provence, 23-26 Sep. 1991, OECD, 41 - 53

A.G. HERMANN, 1988: "Gase in marinen Evaporiten", PTB informiert, No. 2/88, 32 p.

H.-J. KNABE, 1989: "Zur analytischen Bestimmung und geochemischen Verteilung der gesteingebundenen Gase im Salinar", Z. geol. Wiss. 17, 353-368

NEA-OECD, 1992: Gas Generation and Release from Radioactive Waste Repositories, Proc. NEA-Workshop, Aix-en-Provence, 23-26 Sep. 1991, OECD, 437 p.

T. ROTHFUCHS, C. DE LAS CUEVAS, H. DONKER, H.-K. FEDDERSEN, A. GARCIA-CELMA, H. GIES, M. GOREYCHI, V. GRAEFE, J. HEIJDRA, B. HENTE, N. JOCKWER, R. LEMEUR, J. MÖNIG, K. MÜLLER, J. PRIJ, D. REGULLA, E. SMAILOS, G. STAUPENDAHL, E. TILL, and M. ZANKL, 1995: "The HAW-Project: Test Disposal of Highly Radioactive Radiation Sources in the Asse Salt Mine. Final Report", GSF-Report 6/95, 238 p.

W. SCHWARZKOPF, E. SMAILOS, and R. KÖSTER, 1989: "In situ Corrosion Studies on Cast Steel for a High-Level Waste Packaging in a Rock Salt Repository", in "Scientific Basis for Nuclear Waste Management XII", Eds.: W. Lutze, R.C. Ewing, Mater. Res. Soc. Proc. 127, USA, 411-418.

E. SMAILOS, W. SCHWARZKOPF, R. KÖSTER, B. FIEHN, and G. HALM, 1990: "Corrosion Testing of Selected Packaging Materials for Disposal of High-Level Waste Glass in Rock Salt Generations", KfK 4723, Kernforschungszentrum Karlsruhe