LONG-TERM STABILITY OF BITUMEN PRODUCTS

- A JOINT NORDIC PROJECT

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ABSTRACT

Scandinavian disposal concepts for reactor wastes are based on mined geological repositories, e.g. at the depth of -50, -100 m in hard crystallinex rock such as granite. The concepts proposed include the efficient use of engineered barriers. In addition to concrete bentonites and in some cases bentonites mixed with local minerals may be used as construction materials. The main objective of the joint investigation is under nordic final disposal conditions to evaluate the long-term e.g. 1000 A, stability of bituminized wastes and the mechanisms which might with long-term lead to release of radonuclides.

Then joint investigation comprises studies on leaching of bituminized decontamination waste products in cement conditioned water, swelling phenomena of the waste products, release mechanisms for nuclides and long-term properties of bitumen matrix and bituminized waste.

INTRODUCTION

The Nordic Liaison Committee for Atomic Energy NKA) was founded in 1957 to promote joint Nordic co-operation in the nuclear field[1]. Under the coordination of the NKA several joint Nordic Activities have been realized including research projects on nuclear waste management. Recently a project dealing with a generic approach to the safety evaluation of an integrated management scheme for solidified reactor waste was completed[2].

Presently four waste research projects are under way:

- AVF-1 Management of reactor wastes resulting from nuclear fuel damage
- AVF-2 Long-term stability of bitumen products
- AVF-3 Project catalogue on Nordic national research projects
- AVF-4 Comparative studies of hydrological models

The main objective of the AVF-2 project is to evaluate the long-term stability of bituminized reactor wastes under Nordic final disposal conditions. The proposed concepts[3-5] are based on the disposal of wastes in mined geological repositories, e.g. at the depth of 50 m...100m in hard crystalline rock such as granite. The concepts also include the use of engineered barriers such as concrete and bentonites. Bentonites mixed with local minerals may in some cases be used as construction materials.

Main emphasis is put on the identification and investigation of mechanisms and phenomena which within a short term or longer term might lead to changes in the natural properties or in the entire system of waste and surroundings and thereby result in the release of radionuclides. Further knowledge on the changes in the natural properties of bitumen is produced. An essential part of the investigation is devoted to developing general research methodology and instrumentation, which can be used in the participating countries for the evaluation of their specific final disposal concepts.

PROGRAM

The main objective of the AVF-2 project is to evaluate the long-term stability of bituminized reactor wastes under Nordic final disposal conditions including:

- definition of the waste type
- characterization of the chemical conditions in the repository
- basic studies concerning the long-term properties of bituminized waste.

Definition of the waste type

Bituminized waste products containing ion exchange resins, Co, Cs, Sr and Ni isotopes, with an without decontamination chemicals such as EDTA, citrate and ascorbic acid were chosen for the study[6]. The bituminization of reactor wastes is practiced at some nuclear power plants in Sweden as well as in Finland. The incorporation of decontamination waste is closely connected to the AVF-1 research project (Management of radioactive waste resulting from nuclear fuel damage).

The stability of the waste products defined above is investigated by a special leach test procedure[7] using cement conditioned water, a procedure deviating somewhat from the conventional leaching methods (ISO, Hespe, etc.). It combines the measurement of water uptake, swelling and leaching of the bituminized waste with precipitation/adsorption on concrete particles. The formation of neutral complexes and other mobile species is of interest, too.

The proposed procedure is supposed to secure a reasonably constant water chemistry in the system i.e. one which is not widely fluctuating in connection with the sampling as is typical for the ISO and Hespe methods. The presence of a solid cement phase is supposed to stabilize the pH of the solution by supplying new OH^- groups if these are used by the sample. Furthermore, the relatively large area of the granulated cement is expected to serve as a precipitation and adsorption surface for some of the radionuclides which otherwise in a high pH solution might have precipitated on the beaker or on the sample itself.

The possible correlation between leaching and swelling mechanisms will also be investigated.

Characterization of the Chemical Conditions in the Repository

The repositories for reactor wastes, including bituminized wastes, are planned to be located in granite bedrock, at a fairly small depth, both within the Swedish and Finnish waste programs.

The groundwater conditions and chemical parameters affecting the leachability of bitumen matrix and the subsequent release of the incorporated radionuclides are defined[8]. The engineered barrier system shown in Fig. 1 controls the chemical composition of the groundwater within the repository affecting the release of radionuclides. One of the principal chemical parameters is the pH of the water. Since concrete is used as a construction material, the water will be strongly alkaline and have a high content of cations and carbonates. The leaching studies will thus be carried out in a compatible alkaline environment.

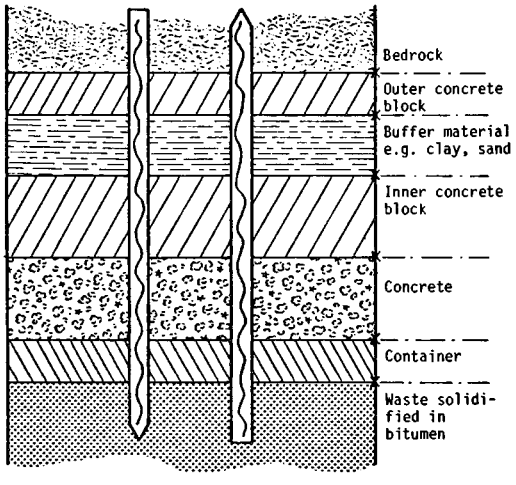

Fig. 1. The engineered barrier system in the repository pathways for transport of radionuclides and groundwater through the barriers with mechanical and chemical interactions between waste and surroundings.

Basic Studies Concerning the Long-Term Properties of Bituminized Waste

During 1982 the main mechanisms and phenomena which might influence the long-term stability and behavior of bituminized waste products were defined. Basic studies in order of priority on the long-term behavior of bituminized waste were identified (Table I[9]). The AVF-2 program in 1983 is mainly based on the priority list given in Table I.

Influence of Bituminized Waste on Surrounding Barriers in a Repository

Bituminized waste has mechanical and chemical influence on the surrounding media which might disturb the function of the engineered barriers. Two physico-chemical phenomena which are of major importance are swelling and gas generation.

A well known fact is that dry ion exchange resins swell in contact with water. Investigations have postulated forces in the order of 30-40 MPa, acting on surrounding media. An evaluation of older leach test samples, which have been in contact with water up to 1600 days, has indicated higher swelling of irradiated resins[10]. National projects for measuring swelling pressures of different resin types and effects of heat or other treatment on the swelling behavior of resins are in progress.

In addition to a model proposed for the swelling of bituminized salt concentrates[11] two different swelling models for bituminized resins on the basis of theoretical studies are under development.

The pressure on surrounding barriers may also increase due to gas evolution caused by radiolysis or microbial attack. In connection with the proposed disposal system for reactor wastes only the anaerobic microbial attack on bituminous material is of importance. However, the process is slow since the conditions are highly unfavorable for bacterial growth (high pH, lack of essential nutrients, etc.). Hence, any resulting gas (mainly H_2, CH_4) would have ample time for being transported away from the repository without any buildup of pressure. Though these phenomena at present are not included in the joint Nordic research, the international development on this and related areas is being followed.

One of the chemical effects to be studied is the degradation of bitumen in an alkaline environment. The degradation of bitumen might lead to the production of surface active agents and complexants, which have chemical influence on the barrier integrity. The present knowledge of degradation of bitumen is covered in two reports[12,13] and further experimental studies on the behavior of bitumen in an alkaline environment are underway. Special attention in the studies is paid to two phenomena. The possible adsorption of solubilized bitumen on the walls of pores and microfissures of the surrounding barrier may affect the transport of radioactive species. Furthermore, the presence of surface active agents may lead to a considerable increase of the barriers permeability.

Long-Term Changes in Matrix, Waste and Waste Products

The long-term alterations in the properties of bituminized waste, such as the viscoelastic properties will be investigated. The physical aging of bitumen is accompanied by structural changes, most of which tend to increase its viscosity. Methods for quantification and examination of these changes are being evaluated.

SUMMARY

The results obtained, hitherto, have indicated that under unfavorable conditions swelling might considerably change the properties and function of barriers. The groundwater conditions expected to prevail under final disposal conditions have been

TABLE I

LONG-TERM BEHAVIOR OF BITUMINIZED REACTOR WASTE. SUGGESTIONS FOR BASIC STUDIES IN ORDER OF PRIORITY.

1.	Influence of bituminized waste on surrounding barriers in a repository
1.1	Mechanical influence
1.1.1	Estimation of swelling pressures
1.1.2	Development of a theory for swelling dynamics
1.2	Chemical influence
1.2.1	Degradation of bitumen and behavior of solubilized bituminous substances in a repository
1.2.2	Behavior of radioactive species in the presence of solubilized bituminous substances
2.	Long-term properties of bitumen and bituminized waste
2.1	Rheological properties
2.2	Sedimentation
2.3	Comparative studies of aged and fresh bitumen
3.	Investigation of a possible correlation between the leaching and swelling mechanisms

characterized. The swelling and leaching tests are performed under simulated conditions in according with the procedure development. Present knowledge of the long-term properties of bitumen is collected from non-nuclear sources and an experimental investigation on the properties of bitumen in alkaline groundwater conditions is underway. On these bases, models for the long-term behavior of bitumen products are being developed.

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