

BEHAVIOR OF SMECTITE IN STRONG SALT BRINES UNDER CONDITIONS RELEVANT TO THE DISPOSAL OF LOW- TO MEDIUM-GRADE NUCLEAR WASTE

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Abstract—Two industrial bentonites, IBECO SEAL-80 and TIXOTON TE, have been proposed as potential backfill material in the German Asse salt dome, a test field for the disposal of low- to medium-grade active nuclear waste. Considering the unlikely but possible case of a barrier breakdown with infiltration of a highly concentrated salt brine, the physicochemical stability and material behavior of these bentonites in a saturated salt brine (predominantly MgCl₂) at 25°C were studied over the time period of 150 days. The results show that no mineral transformations occurred throughout the duration of the experiments and minor dissolution was only active during the first days. Some chemical properties, namely sorption capability and swelling, were reduced during contact with the salt brine, but could be reversed by removing the salt after treatment. Despite restriction of the CEC in the presence of salt solution, interlayer cation exchange reactions are still active in this environment. The long-term chemical stability of smectite in salt brine is predicted under these low-temperature conditions, but the increased permeability during aggregate formation could lead to physical breakdown of the backfill component.

Key Words—Cation Exchange Capacity, Layer Charge, Nuclear Waste, Salt Brine, Smectite.

INTRODUCTION

The secure long-term storage of nuclear waste in underground repositories requires an effective combination of both natural and technical barriers to prevent radionuclides from migrating into the groundwater and finally from coming into contact with the biosphere (Kim *et al.*, 2001). In Germany, deeply buried salt formations are being considered as appropriate host rocks due to their fracture and self-healing properties (Pusch and Alstermark, 1985; Pusch, 1992), and the low groundwater flow over geologically relevant time scales (Schmidt, 1995). This scenario is currently being investigated at the test site of the German Asse salt dome (Gömmel, 1997). Here, 126,000 containers of low- to medium-grade radioactive waste have been stored between 1967 and 1978, and over the past few years, a highly concentrated salt brine has entered the mine. Two bentonites, 'TIXOTON TE' and 'IBECO SEAL-80', are being evaluated for use as backfill which should prevent the waste canisters from coming into contact with the corrosive salt brine and thus prevent radionuclide release.

Although bentonite is a well established sealant used in many disposal sites, its application as a backfill in nuclear waste repositories is not yet fully understood. The main constituent of bentonites, the smectite clays, are considered to be ideal due to their swelling/sealing ability and high cation exchange capacity (CEC) (see Bauer *et al.*, 2001, for review). The high CEC of smectite enables sorption of cations and polar substances

onto particle surfaces and into interlayer spaces (Zachara *et al.*, 2002), and therefore has the potential to retain dangerous radionuclides (Sylwester *et al.*, 2000; Coppin *et al.*, 2002).

Despite the beneficial properties of the smectite group minerals, the stability of these minerals in strong salt solutions is a question of concern. A number of experiments have been conducted to evaluate the stability and behavior of smectite in salt solutions. These experiments show that a number of dissolution and precipitation reactions can occur at elevated temperatures (>50–60°C) and under strong acidic or alkaline conditions (Bauer *et al.*, 2001; Huertas *et al.*, 2001; Cama *et al.*, 2000; Kasbohm *et al.*, 2000; Metz, 2001; Zysset and Schindler, 1996; Eberl *et al.*, 1993; Inoue, 1983; Komarneni and Roy, 1983; Komarneni and White, 1983; Eberl and Hower, 1977). There is, however, a notable lack of experimental data for lower-temperature, near-neutral-pH brines, conditions which are directly relevant to the Asse test site. As the low and medium grade radioactive waste produces no heat, the temperature in the repository chambers at a depth of 510–750 m is not expected to exceed 25°C. Thus to fill this niche, this study evaluates the stability and behavior of the bentonite backfill under conditions specific to this repository and also examines the influence of the salt brine on the CEC and layer-charge properties of the clays.

MATERIALS AND EXPERIMENTAL SETUP

The two industrial bentonites used for experimental work are described as follows: 'IBECO SEAL-80' is a Na-activated bentonite distributed by IBECO Bentonit-

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