HYDROTHERMAL STABILITY OF LAYERED SILICATES IN NEUTRAL AND ACIDIC MEDIA: EFFECT ON ENGINEERED-BARRIER SAFETY

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Abstract—Many environmental applications in the inorganic remediation field are based on the swelling and ion-exchange capacities of smectites, even though these can be affected by hydrothermal treatment in water and acidic media. Here a systematic study of the properties of layered silicates that could affect their hydrothermal stability at different pH is described: type of layers, octahedral occupancy, layer charge, and origin of the layer charge. The silicates studied were selected on the basis of their different characteristics associated with these properties. Kanemite (1:0 phyllosilicate), kaolinite (1:1 phyllosilicate), and pyrophyllite and talc (2:1 phyllosilicates with no-layer charge) were examined in order to determine the effect of layer structure, whereas the hydrothermal reactivity of silicates with different layer charge was analyzed by comparing the talc- Hectorite-Laponite and talc-saponite-trioctahedral vermiculite series. Samples were treated hydrothermally at 300°C for 48 h in pure water and in a 0.01 M HNO3 solution and the final products were analyzed by X-ray diffraction, scanning electronic microscopy, and solid-state nuclear magnetic resonance spectroscopy. All layered silicates, except for kanemite, were found to remain intact after hydrothermal treatment in water and acidic media, with only minimal short-range structural changes observed. The extent of the structural changes depended on the octahedral sheet occupancy (greater extent) and the number of isomorphic substitutions (lesser extent), both of which weaken the structure.

Key Words—Bentonite, Engineered-barrier, Hydrothermal Stability, Layered Silicates,

INTRODUCTION

Natural and modified bentonites, which consist mainly of smectites, have been used in many applications (Grim, 1968; Grim and Güven, 1978; Murray, 1991, 1999, 2000; Bergaya et al., 2006). Bentonite, especially Na-bentonite, is used extensively either on its own or as a component in soil mixtures in many geoenvironmental engineering applications, including the construction of waste-containment barriers. The design of such barriers requires the compatibility of the soil barrier with the waste. The main hurdle to overcome during the design and construction of safe nuclear-waste repositories is that of water, which can transport the radionuclides in dissolved form or as colloids or small particles. If water reaches the canister and corrodes it, the emplacement should be able to retain the radionuclides in the water and avoid their transport away from the repository (Cuadros, 2008). The interaction of this barrier material with groundwater can, however, modify its physical and chemical properties, thus decreasing its groundwater isolation capacity or, in the case of canister failure, decreasing the radionuclide retention capacity of clay materials (Pusch et al., 2007).

Because accidents can modify the behavior of such barriers, the hydrothermal stability of clay in water and in basic and acidic media is of great importance, particularly where radioactive-waste confinement is concerned. In many countries (such as Belgium, Germany, France, Japan, Switzerland, etc.), deep clay formations are considered to be potential host rocks for radioactive-waste disposal and so the clay buffer is of special concern because even moderate mineralogical changes can lead to the reduction and, eventually, to the loss of the critical sealing and mechanical properties of the buffer (Pusch and Yong, 2003; Pusch et al., 2003). Precise knowledge of the clay’s properties in the various domains concerned with construction feasibility, such as the long-term integrity of the bentonite as it comes into contact with either acidic tailings pore water (in the case of slurry wall, filter cake) or acidic, stored, mine water (in the case of acid-mine drainage, collection ponds), is, therefore, of crucial importance when assessing the performance and safety of radioactive-waste disposal concepts. Furthermore, concrete and cement, which have been proposed for use as matrix material, backfill material, and structural components of radioactive-waste repositories (Glasser, 2001), are likely to produce solutions with a high pH which will also modify the clay’s properties.

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