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**Uncertainties, Issues, and Problems Related to the Use of
Basalt/Smectite as Backfill Material for a High-Level
Nuclear Waste Repository in Basalt**

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The following is a brief synopsis of major uncertainties, issues and problems relating to the use of basalt/smectite as a backfill material, based in part on research carried out in the Mineralogical Laboratory, the University of Michigan.

I. Definition of the Problem.

Smectite is a clay mineral rich in Si, Al, Na and H₂O which is common in low-temperature (less than - 200°C) geological environments. Experimental research results imply that it is a metastable phase in such environments, forming only as an intermediate product to more stable products, to which it eventually transforms. Furthermore, exhaustive geologic research confirms that it is unstable in geologic environments and transforms to phases such as illite, a more stable clay mineral. Illite itself is unstable and it changes to the more stable phase muscovite. However, this transformation involves only minor changes in the chemistry, structure and properties of illite compared to the major changes attendant to the smectite to illite transformation.

Because smectite is a metastable phase relative to illite and/or other transformation products, it must eventually undergo transformation. The rate of transformation, as determined by kinetic factors, is therefore the critical factor. The rate of transformation is primarily dependent on (1) temperature and (2) water/rock ratio.

Temperature is critical as shown especially in geologic systems in which untransformed smectite occurs in rocks only up to temperatures of approximately 150°C. Indeed, the ratio of transformed illite clay layers to those of untransformed smectite is commonly used as a measure of temperature by geologists, under the assumption, in part, that other factors have a minimum effect. Insofar as a temperature of 300°C was set as a limit for backfill materials the metastability of smectite is a crucial factor.

Water/rock ratio is at least as significant as temperature in its effect on rate

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of transformation. Water is an important component of the phases, serves to catalyze the reactions, and acts as a vehicle for transport of essential reactants (e.g. K, Al) and products (e.g. Si). In geologic systems which are impermeable to water, mixed-layer smectite/illite may occur at temperatures up to 250°C over periods of millions of years. In geologic systems such as sandstones which are permeable to water, clay minerals react rapidly at relatively low temperatures. The geologic environment serves as a model for synthetic systems, and is a natural laboratory for backfill materials involving time periods which are unrealistic in laboratory experiments. Conclusions derived from such systems are basic to an understanding of the properties and stability of backfill materials.

Summary statement. Smectite is metastable relative to illite and muscovite. The rate of reaction (and therefore the rate of change in physical properties) is primarily a function of temperature and water/rock ratio. Changes in the chemistry, structure, texture and properties of smectite in the backfill environment are inevitable, but the rates of reaction may be contained within useable limits, in part by utilizing data from analogous geologic systems.

Backfill materials may be chosen, and their physical states defined, so as to minimize the smectite/illite transformation rate. Several factors to be considered are described below.

II. Specific Problems

(I) Glass content of basalt.

The Umtanum basalt and associated basalt flows contain up to 25% glass which is intersitital to the crystalline phases such as plagioclase and pyroxene. Furthermore, we have shown that the glass consists of two separate, exsolved glass phases. One of these is rich in K. In the presence of H₂O, glass is especially reactive and can be expected to go into solution long before the crystalline components are affected.

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One of the principal components necessary for conversion of smectite to illite is K. In its absence, the smectite transformation occurs to other phases and is apparently retarded. Even if smectite is subjected to pore water and elevated temperatures, transformation to illite cannot occur without K.

The presence of highly soluble glass will therefore significantly affect the general porewater chemistry and specifically provide K necessary for conversion of smectite. The glass is also rich in Al, a second component necessary to the smectite to illite reaction. The rate of conversion of smectite may therefore be significantly affected by the use of a basalt which contains glass.

Crystalline, igneous rocks which have chemistry, mineralogy and texture which are identical, for these purposes, to basalts containing significant glass are readily available. A rock such as diabase, for example, should be substituted for any backfill basalt which is shown to contain glass, such as the Umtanum Basalt. In any event, it is essential that a glass-free rock be chosen as the non-clay component of the backfill material.

(2) Pore space

Much research has been carried out on geologic systems where smectite, mixed layer smectite/illite, or illite are retained as metastable phases, even over time periods of tens of millions of years. High resolution transmission electron microscopy shows that the clays form, in part through compaction and recrystallization, a continuous, anastomosing framework which is molded to and surrounds, the disconnected, separate, approximately equi-dimensional grains of other phases (the latter is analogous to crushed basalt fragments; the natural clays are analogous to smectite in the basalt/smectite backfill material). Such systems have apparently been virtually closed systems; that is, no chemical components (including water) have been added or lost following the compaction/recrystallization process, due apparently to the barrier created by the

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framework of clay minerals. Because such rocks are virtually impermeable to water, and because migration of cations and anions occurs principally through water, other chemical components remain unaffected. Such rocks retain their original mineral phases, with a minimum of change.

By contrast, in systems where porosity and permeability are high (as in coarse-grained sandstones) fluid flow readily occurs, rock/water ratios are high and water serves as reactant in clay mineral reactions, a catalyst to such reactions, and as a vehicle for transport of necessary chemical components. Smectite, for example, reacts readily, in direct contrast to its behavior in clay-rich systems (shale).

It is therefore essential to produce a physical (textural) state for the backfill material which minimizes porosity and permeability. By analogy to those geologic systems where this has been achieved, the following three factors are critical:

a. proportions of smectite vs. basalt.

A high proportion of equi-dimensional basalt grains, relative to clays will prevent the clays from forming the continuous network which is the primary barrier to fluid transport. High clay/basalt ratios are essential. Data must be presented which optimizes this factor.

b. Compaction

The barrier network of clays can be generated only with significant compaction/recrystallization. The swelling properties of smectite are, we believe, insufficient in promoting the barrier network. Significant compaction is essential. If a system such as blowing the smectite into a cavity is used, rather than physical compaction, then high permeability with consequent high reaction rates is inevitable. Whatever system is used, it must be based on hard, specific data relating to those variables, rather than engineering convenience.

c. The basalt grain size, relative to that of smectite (1000's of Angstroms) is a critical factor in the formation of a clay barrier network. Grain size must be chosen, in combination with a method of compaction, to produce the impermeable membrane.

(3) Smectite vs. Illite.

Because the reactions of smectite to mixed-layer smectite/illite (S/I), of S/I to illite, and illite to muscovite involve phases which are closer to equilibrium phases as the reactions proceed, the rates of reactions should generally be smaller for those phases nearest to muscovite in chemistry and structure. On the other hand, it is the swelling property of smectite which contributes to its value as a backfill material even though it is the most reactive phase in this sequence.

Mixed layer smectite/illite should therefore be strongly considered as a backfill clay mineral in place of smectite. It retains some limited swelling ability and has the much greater advantage of increased relative stability. Its properties are far more likely to be retained, relative to smectite, and it is the lack of change in backfill properties which is essential. If smectite is proposed as a backfill material, then those who would promote its use should be required to justify it relative to other clay minerals.

(4) Limestone as a minor component. Experiments show that solutions enriched in Ca relative to K cause smectite to react to illite at much smaller rates. This is because the smectite structure is stable with interlayer Ca, while illite requires the larger K ion. Calcite, the principal component of limestone, dissolves readily at the low temperatures proposed for the repository and therefore acts to increase Ca activity and to retard the rate of transformation of smectite. Given that the backfill is to be subjected to pore fluids, consideration should be given to the inclusion of a small proportion of limestone as a backfill component. We emphasize that this factor is a secondary one, relative to the others described