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Glass Dissolution Rates From MCC-1 and Flow-Through Tests

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Summary

The dose from radionuclides released from high-level radioactive waste (HLW) glasses as they corrode must be taken into account when assessing the performance of a disposal system. In the performance assessment (PA) calculations conducted for the proposed Yucca Mountain, Nevada, disposal system, the release of radionuclides is conservatively assumed to occur at the same rate the glass matrix dissolves. A simple model was developed to calculate the glass dissolution rate of HLW glasses in these PA calculations [1].

For the PA calculations that were conducted for Site Recommendation, it was necessary to identify ranges of parameter values that bounded the dissolution rates of the wide range of HLW glass compositions that will be disposed. The values and ranges of the model parameters for the pH and temperature dependencies were extracted from the results of SPFT, static leach tests, and Soxhlet tests available in the literature. Static leach tests were conducted with a range of glass compositions to measure values for the glass composition parameter.

The glass dissolution rate depends on temperature, pH, and the compositions of the glass and solution. The dissolution rate is calculated using Eq. 1:

$$\text{rate} = k_o 10^{(\text{pH})\eta} \cdot e^{(-Ea/RT)} \cdot (1-Q/K) + k_{long}$$

where k_0, η, and Eaare the parameters for glass composition, pH, η and temperature dependence, respectively, and R is the gas constant. The term (1-Q/K) is the affinity term, where Q is the ion activity product of the solution and K is the pseudo-equilibrium constant for the glass. Values of the parameters k_0, η and E_a are the parameters for glass composition, pH, and temperature dependence, respectively, and R is the gas constant. The term (1-Q/K) is the affinity term, where Q is the ion activity product of the solution and K is the pseudo-equilibrium constant for the glass. Values of the parameters k_0, and Ea are determined under test conditions where the value of Q is maintained near zero, so that the value of the affinity term remains near 1. The dissolution rate under conditions in which the value of the affinity term is near 1 is referred to as the forward rate. This is the highest dissolution rate that can occur at a particular pH and temperature. The value of the parameter K is determined from experiments in which the value of the ion activity product approaches the value of K. This results in a decrease in the value of the affinity term and

the dissolution rate.

The highly dilute solutions required to measure the forward rate and extract values for k_0, η, and Ea can be maintained by conducting dynamic tests in which the test solution is removed from the reaction cell and replaced with fresh solution. In the single-pass flow-through (SPFT) test method, this is done by continuously pumping the test solution through the reaction cell. Alternatively, static tests can be conducted with sufficient solution volume that the solution concentrations of dissolved glass components do not increase significantly during the test. Both the SPFT and static tests can be conducted for a wide range of pH values and temperatures.

Both static and SPFT tests have short-comings. The SPFT test requires analysis of several solutions (typically 6-10) at each of several flow rates to determine the glass dissolution rate at each pH and temperature. As will be shown, the rate measured in an SPFT test depends on the solution flow rate. The solutions in static tests will eventually become concentrated enough to affect the dissolution rate. In both the SPFT and static test methods, a compromise is required between the need to minimize the effects of dissolved components on the dissolution rate and the need to attain solution concentrations that are high enough to analyze.

In this paper, we compare the results of static leach tests and SPFT tests conducted with simple 5-component glass to confirm the equivalence of SPFT tests and static tests conducted with pH buffer solutions. Tests were conducted over the range pH values that are most relevant for waste glass dissolution in a disposal system. The glass and temperature used in the tests were selected to allow direct comparison with SPFT tests conducted previously. The ability to measure parameter values with more than one test method and an understanding of how the rate measured in each test is affected by various test parameters provides added confidence to the measured values.

The dissolution rate of a simple 5-component glass was measured at pH values of 6.2, 8.3, and 9.6 and 70°C using static tests and single-pass flow-through (SPFT) tests. Similar rates were measured with the two methods. However, the measured rates are about 10X higher than the rates measured previously for a glass having the same composition using an SPFT test method. Differences are attributed to effects of the solution flow rate on the glass dissolution rate and how the specific surface area of crushed glass is estimated. This comparison indicates the need to standardize the SPFT test procedure.