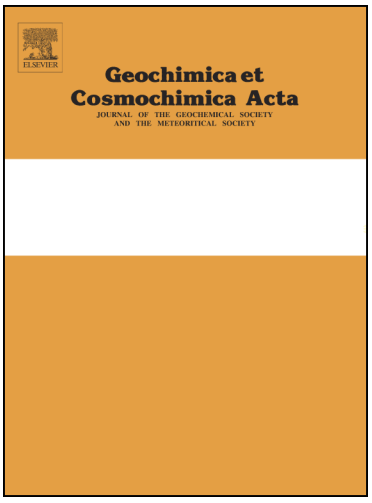

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The Dissolution Behavior of Borosilicate Glasses in Far-From Equilibrium Conditions

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ABSTRACT

An area of agreement in the waste glass corrosion community is that, at far-from-equilibrium conditions, the dissolution of borosilicate glasses used to immobilize nuclear waste is known to be a function of both temperature and pH. The aim of this work is to study the effects of temperature and pH on the dissolution rate of three model nuclear waste glasses (SON68, ISG, AFCI). The dissolution rate data are then used to parameterize a kinetic rate model based on Transition State Theory that has been developed to model glass corrosion behavior in dilute conditions. To do this, experiments were conducted at temperatures of 23, 40, 70, and 90 °C and pH(22 °C) values of 9, 10, 11, and 12 with the single-pass flow-through (SPFT) test method. Both the absolute dissolution rates and the rate model parameters are compared with previous results. Rate model parameters for the three glasses studied here are nearly equivalent within error and in relative agreement with previous studies though quantifiable differences exist. The glass dissolution rates were analyzed with a linear multivariate regression (LMR) and a nonlinear multivariate regression performed with the use of the Glass Corrosion Modeling Tool (GCMT), with which a robust uncertainty analysis is performed. This robust analysis highlights the high degree of correlation of various parameters in the kinetic rate model. As more data are obtained on borosilicate glasses with varying compositions, a mathematical description of the effect of glass composition on the rate parameter values should be possible. This would allow for the possibility of calculating the forward dissolution rate of glass based solely on composition. In addition, the method of determination of parameter uncertainty and correlation provides a framework for other rate models that describe the dissolution rates of other amorphous and crystalline materials in a wide range of chemical conditions. The higher level of uncertainty analysis would provide a basis for comparison of different rate models and allow for a better means of quantifiably comparing the various models.