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A new kinetic rate equation for silica polymerization in geologically-relevant aqueous solutions

SAMUEL SCOTT[1], IWONA GALECZKA[2], INGVI GUNNARSSON[3], STEFÁN ARNÓRSSON[1] AND ANDRI STEFÁNSSON[4]

[1]Nordic Volcanological Center, Institute of Earth Sciences, University of Iceland

[2]Iceland Geosurvey

[3]Reykjavik Energy

[4]University of Iceland

Presenting Author: samuelwarrenscott@gmail.com

Aqueous silica is among the major components of natural hydrothermal fluids. In solutions that are supersaturated with respect to amorphous silica, monomeric silica polymerizes and forms nanocolloidal particles that aggregate, settle out of solution, and undergo diagenesis to form amorphous silica polymorphs such as opal. As silica polymerization is kinetically-controlled, understanding the physio-chemical factors controlling the mechanism and rate of silica polymerization is crucial to understanding a variety of processes including biosilicification, silica sinter formation, silica scaling, and silica diagenesis. Although previous studies have shown that the rate of silica polymerization depends on factors like pH, temperature and ionic strength, the reported reaction rate equations are unable to predict the rate of silica polymerization over a wide range of conditions. In this study, the polymerization of silica was studied at temperature 23-80ºC, pH 2-10, and ionic strength 0.01-0.5 M. Consistent with previous studies, we fit measured changes in the concentration of monomeric silica ($SiO_{2(mono)}$) assuming the polymerization reaction is fourth-order with respect to the difference between the concentration of orthosilicic acid ($H_4SiO_{4(aq)}$) and its concentration at equilibrium. In addition, the rate law incorporates a term describing how the number and surface area of silica polymers changes during nucleation, growth, and Ostwald ripening of formed polymers. The experimentally-determined rate constants were fitted using an Arrhenius equation, with additional empirical terms for pH and ionic strength. At a given pH, increasing temperature from 20-80ºC or increasing ionic strength from 0.1-0.5 M causes the rate of silica polymerization to increase by roughly one order of magnitude. In comparison, increasing pH from 2 to 10 causes the rate constant to increase by roughly six orders of magnitude. However, the rate of silica polymerization decreases at pH above ~8 due to the decrease in the activity of $H_4SiO_{4(aq)}$ and the increasing solubility of amorphous silica. The developed rate equation is able to predict rates of silica polymerization across a wide range of fluid compositions. This project has received funding from the European Union’s Horizon 2020 under Grant Agreement #851816 (GeoPro).