Diffusion and reaction of water in rhyolitic glasses and melts

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Quantitative study for dehydration of silicate glasses and melts is essential to understand bubble formation processes in magmas and magma flow dynamics in a volcanic conduit. Because hydroxyl group (OH) and molecular water (H2Om) are stable species in silicate glasses and melts, the dehydration processes include interconversion reaction between them as well as their diffusion. To study experimentally the diffusion and reaction processes during the dehydration, we performed the dehydration experiments at 723–823 K for rhyolitic glasses with 4wt% total water and measured directly OH and H2Om contents during the dehydration at high temperature using in situ infrared (IR) spectroscopy. This in situ technique is a powerful tool to study the reaction kinetics at high temperatures, because it has been known that water speciation in experimental samples can relax during the sample cooling from experimental temperatures to room temperature. The obtained temporal variations of the OH and H2Om contents were first modeled by assuming the H2Om diffusion and the reaction of 2OH $\leftrightarrow$ H2Om + O. This model successfully reproduced the experimental results at the initial stage of dehydration. Due to the rapid interconversion reaction between OH and H2Om, the equilibrium water speciation was reached at this stage. On the other hand, with increasing dehydration degree, the measured OH content became relatively larger than that at equilibrium and deviated from the results calculated based on the above diffusion and reaction model. This result indicates that the consumption rate of the OH group, i.e. formation rate of H2Om, became smaller than the rate of diffusive H2Om loss with decreasing water contents. To explain this change in the reaction rate, we assumed a phenomenological reaction model of 2OHsingleton $\leftrightarrow$ OHpair $\leftrightarrow$ H2Om + O, where OHsingleton and OHpair represent the singleton and encountered pair of the OH groups. In this model, the encountered OH pair rapidly reacts to form H2Om, but the consumption rate of OH is controlled by a slow migration of OH singleton at low OH contents. By combining this reaction model with the H2Om diffusion, we numerically simulated the dehydration, which successfully reproduced the experimental data. Based on these results, we conclude that the dehydration kinetics can be primarily understood by the H2Om diffusion and equilibrium reaction between OH and H2Om when there are abundant OH pairs, but by the H2Om diffusion and the reaction taking into account slow migration of OH bound to silicate structure at low water contents.