

Molecular dynamics simulation and machine learning investigation on the redox state of silicate melts

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Silicate melts are the dominant constituent of terrestrial magma. The ferrous/ferric ratio of the silicate melts of magmatic rocks is widely used to infer their source regions' redox conditions or the oxygen fugacities. This inference, however, suffers from the poorly understood parameters involved in the thermodynamic relationship between the oxygen fugacity and the ferrous/ferric ratio. Among these, the partial heat capacity and the activity of iron oxides (ferrous and ferric oxides) in the silicate melt are the most critical yet least constrained. Here, we calculate the partial heat capacity of iron oxides over a wide temperature range using the *ab initio* molecular dynamics simulations. The specific heat capacity results agree with previous experiments under 2000 K. The activity of iron oxides, on the other hand, is resolved by two approaches (1) comparing different chemical mixing models and (2) using the machine learning (ML) method by considering all previous experimental results. Using multicomponent symmetrical chemical mixing models with the empirical parameters, the approach (1) calculation shows that the ferrous/ferric activity ratio in the silicate melts deviates from the anticipated results based on ΔG (R square <0.2). In addition, Al_2O_3 shows a minimal contribution to the variation of the ferrous/ferric activity ratio. In approach (2), the activity ratio calculation by ML models (Extra Trees and Random Forest) based on the experimental dataset has higher accuracy (RMSE < 2.7; R square >0.7) and shows a significant improvement over the traditional models. The feature importance values suggest higher importance of iron oxides and temperature followed by MnO and TiO_2 , while the relative importance of Al_2O_3 and SiO_2 is very low. In sum, the acquired heat capacity and activity of iron oxides allow for the calibration of a new model relating the ferrous/ferric ratio to oxygen fugacity, providing an improved and more concrete constraint on the occurrence of volatile species in the terrestrial magma oceans.