

Supporting Information for

Structures and transport properties of supercritical $\text{SiO}_2\text{--H}_2\text{O}$ and $\text{NaAlSi}_3\text{O}_8\text{--H}_2\text{O}$ fluids

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Introduction

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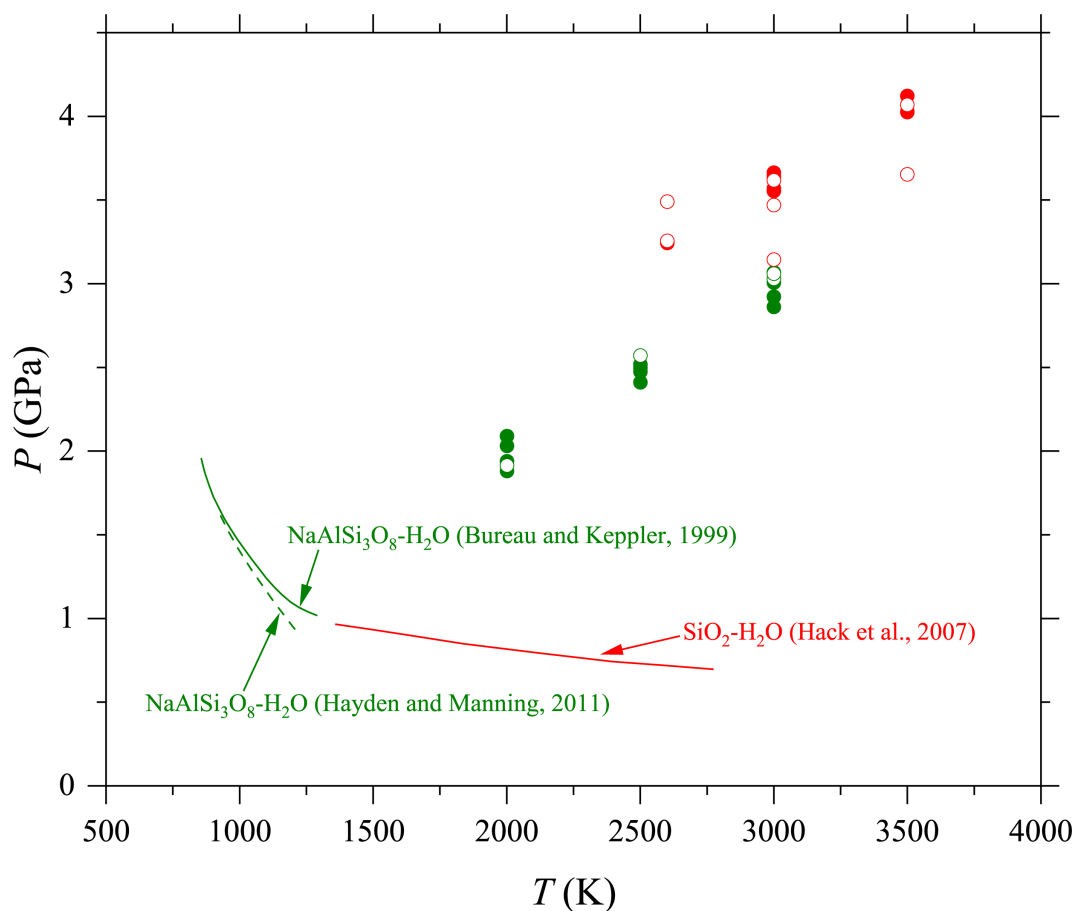


Figure S1. Critical curves of the $\text{SiO}_2\text{--H}_2\text{O}$ and $\text{NaAlSi}_3\text{O}_8\text{--H}_2\text{O}$ systems. The red and green symbols represent the P - T conditions of the $\text{SiO}_2\text{--H}_2\text{O}$ and $\text{NaAlSi}_3\text{O}_8\text{--H}_2\text{O}$ systems, respectively, in this study. The open symbols indicate the $\text{SiO}_2\text{--H}_2\text{O}$ systems with a water content of less than 30 wt% and the $\text{NaAlSi}_3\text{O}_8\text{--H}_2\text{O}$ systems with a water content of less than 20 wt%. Data sources of the critical curves: Bureau and Keppler (1999), Hack et al. (2007) and Hayden and Manning (2011). The original data used by Hack et al. (2007) to constrain the critical curve of $\text{SiO}_2\text{--H}_2\text{O}$ come from Kennedy et al. (1962), Anderson and Burnham (1965), Nakamura (1974), Manning (1994) and Guissani and Guillot (1996). Critical curve is a curve linking critical points of end members in a two-component system, see Manning (2004) and Hack et al. (2007) for details.

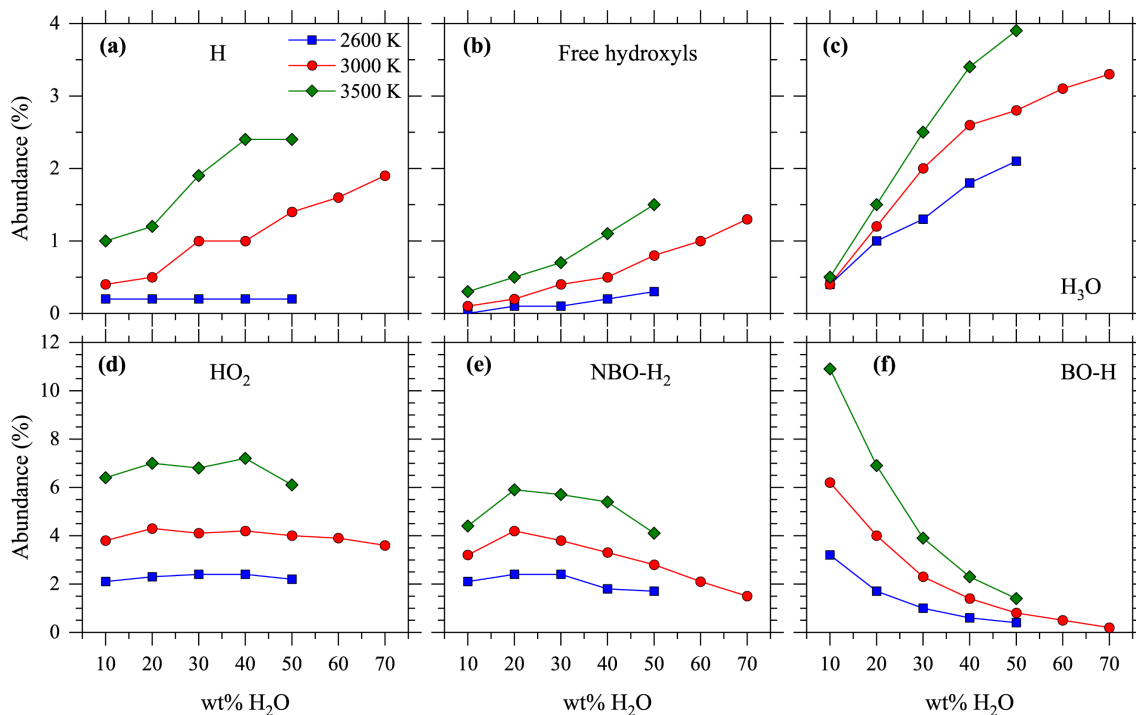


Figure S2. Speciation of hydrogen in the $\text{SiO}_2\text{-H}_2\text{O}$ systems.

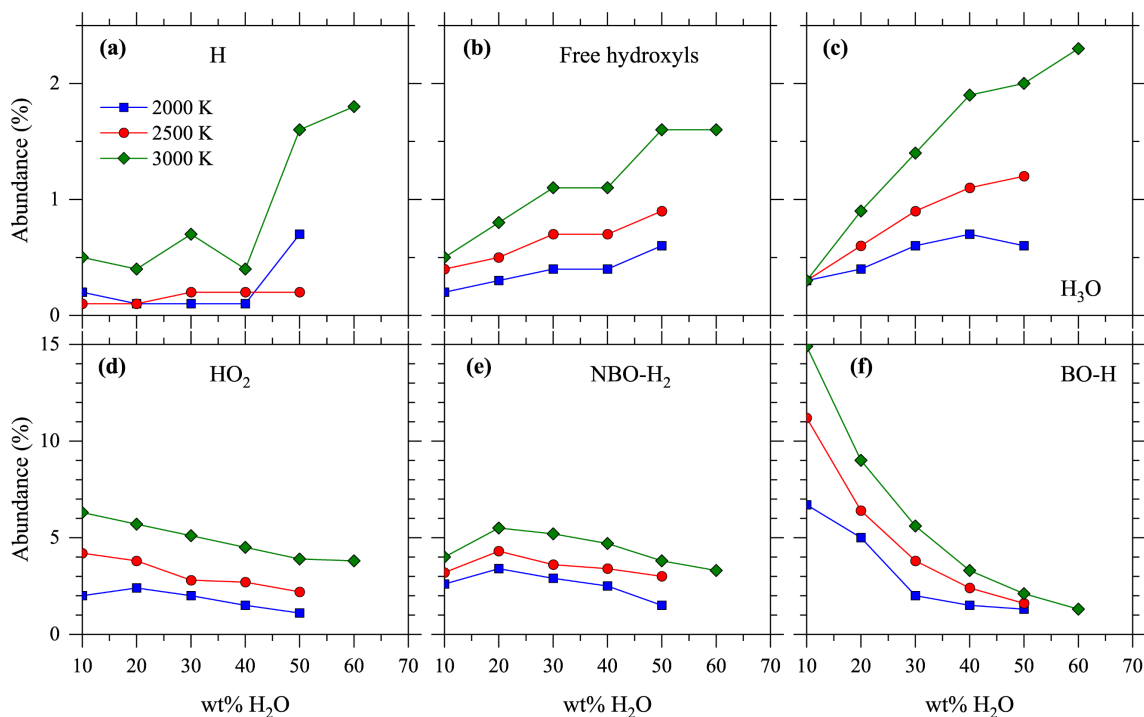


Figure S3. Speciation of hydrogen in the $\text{NaAlSi}_3\text{O}_8\text{-H}_2\text{O}$ systems.

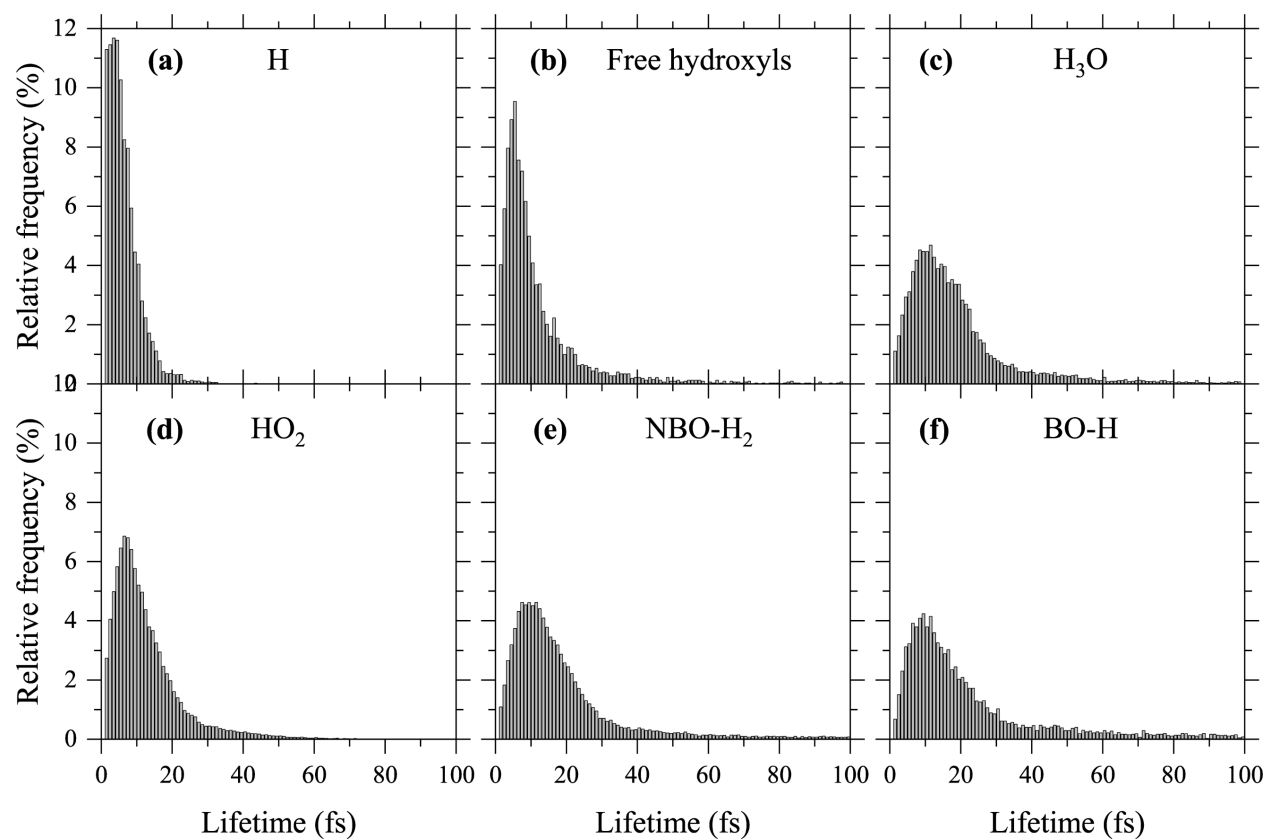


Figure S4. Histograms of the hydrogen species lifetime in the $\text{SiO}_2\text{-H}_2\text{O}$ system with 30 wt% H_2O at 2600 K.

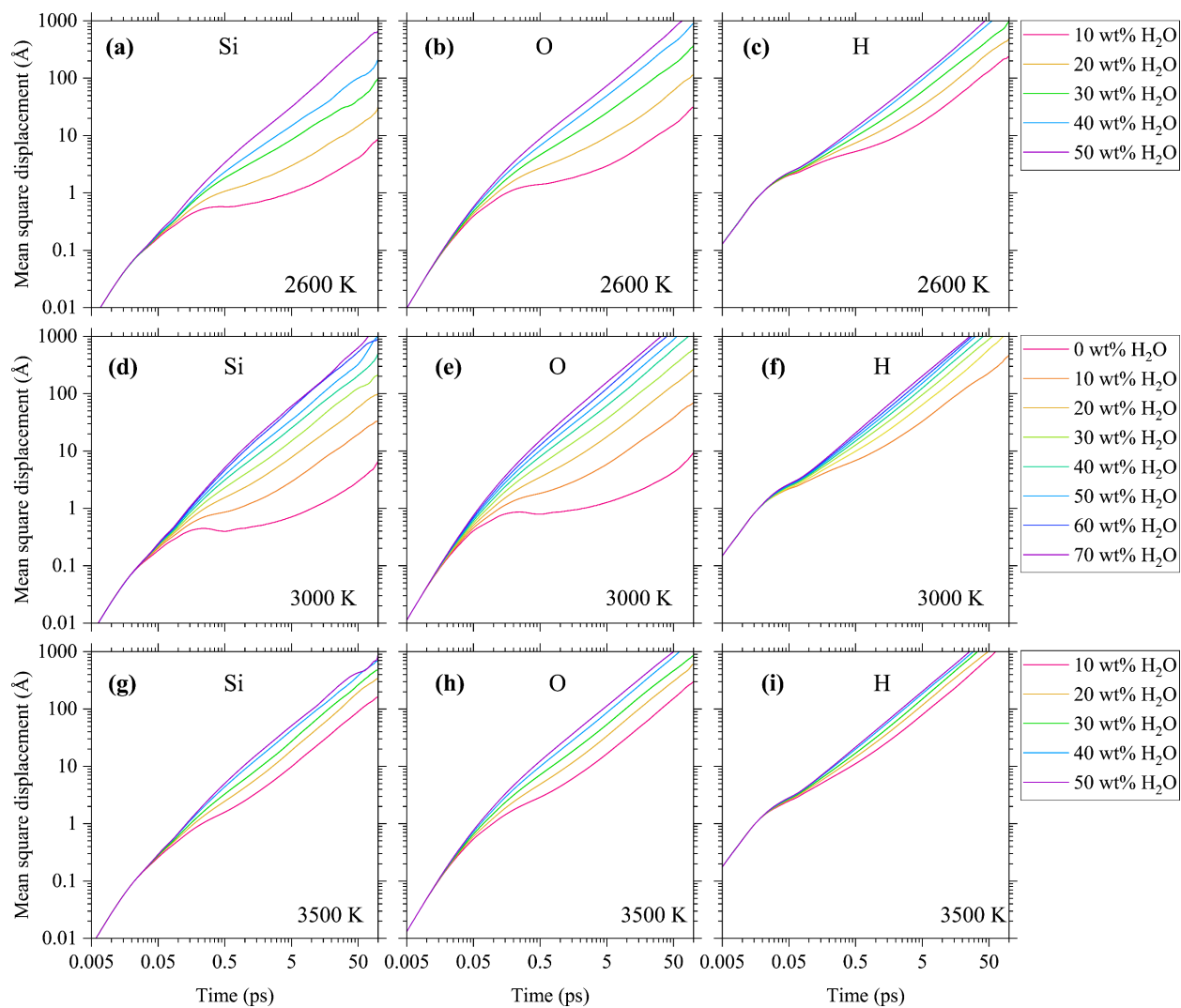


Figure S5. Mean square displacement (MSD) of all elements (Si, O, H) in the $\text{SiO}_2\text{-H}_2\text{O}$ systems at 2600 K, 3000 K, and 3500 K.

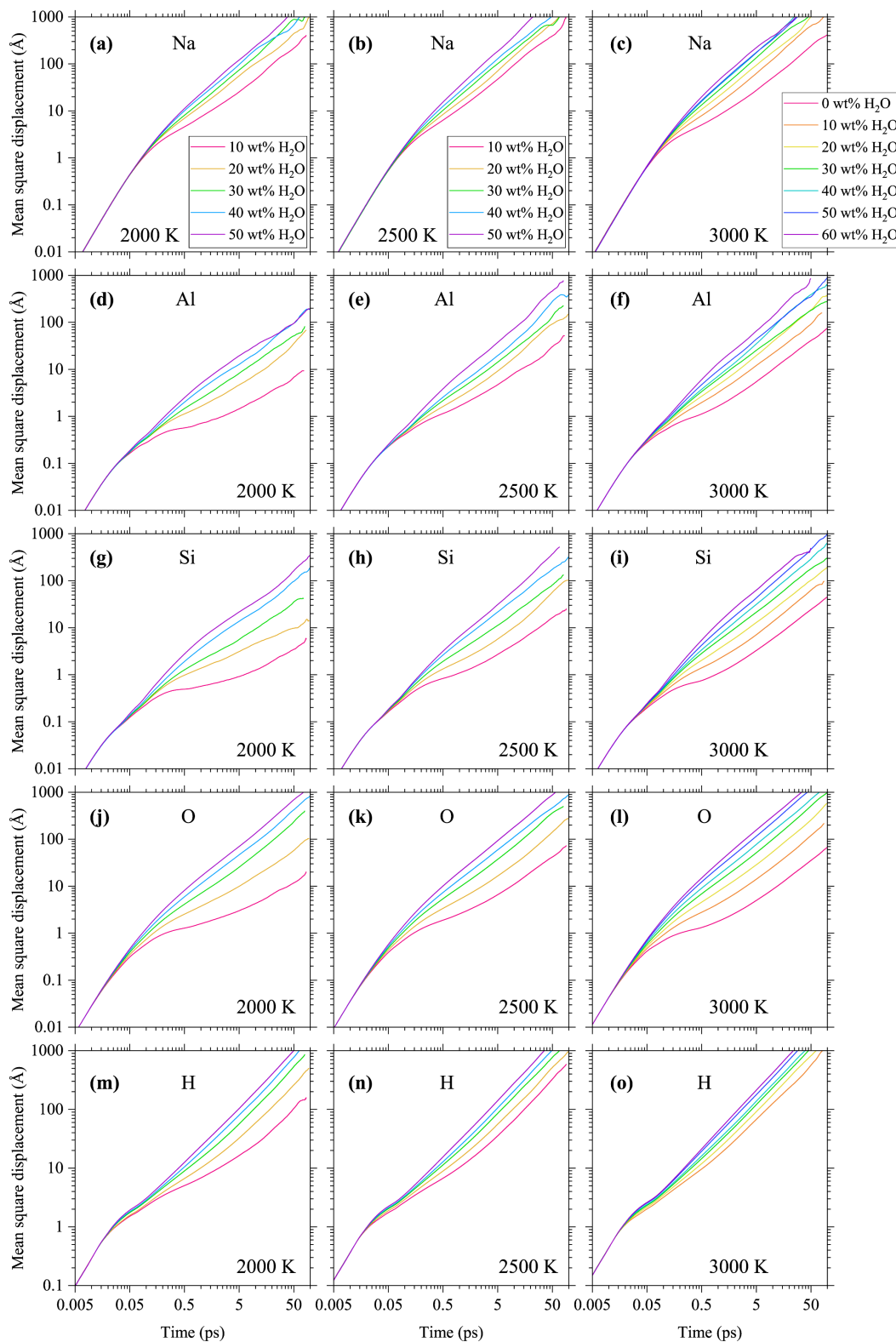


Figure S6. Mean square displacement of all elements (Na, Al, Si, O, H) in the NaAlSi₃O₈-H₂O systems at 2000 K, 2500 K, and 3000 K.

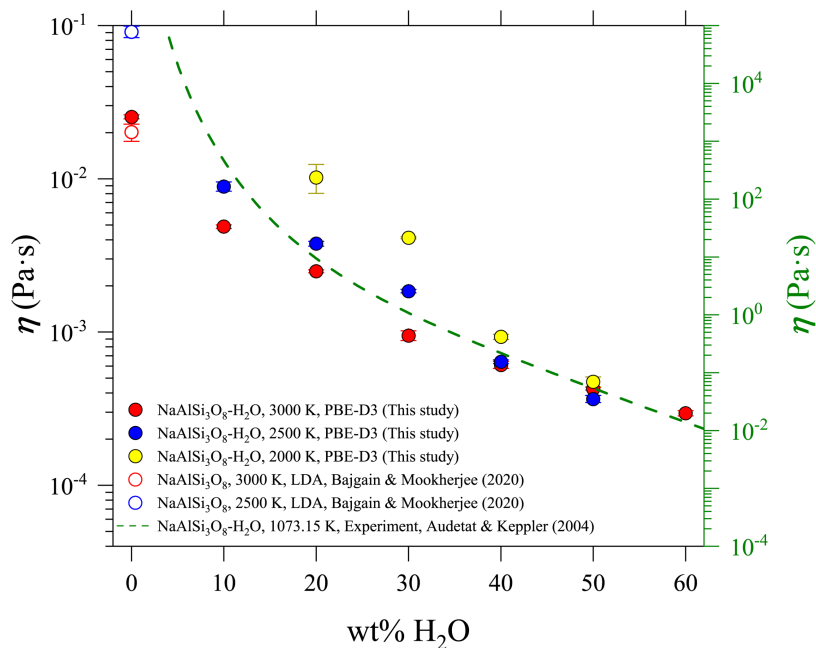


Figure S7. The viscosities of the $\text{NaAlSi}_3\text{O}_8\text{--H}_2\text{O}$ systems as a function of water content. The calculated viscosities of the $\text{NaAlSi}_3\text{O}_8\text{--H}_2\text{O}$ systems in this study at 2000 K (yellow filled circles), 2500 K (blue filled circles) and 3000 K (red filled circles) are shown. The FPMD results for the $\text{NaAlSi}_3\text{O}_8$ melt (Bajgain and Mookherjee, 2020) at 2500 K (2.78 GPa) and 3000 K (3.31 GPa) are included for comparison. The green dashed curve represents the results of Audétat and Keppler (2004) at 1073.15 K, and please note that the values correspond to the right y-axis in green. The insets show our viscosity data plotted with a linear y-axis.

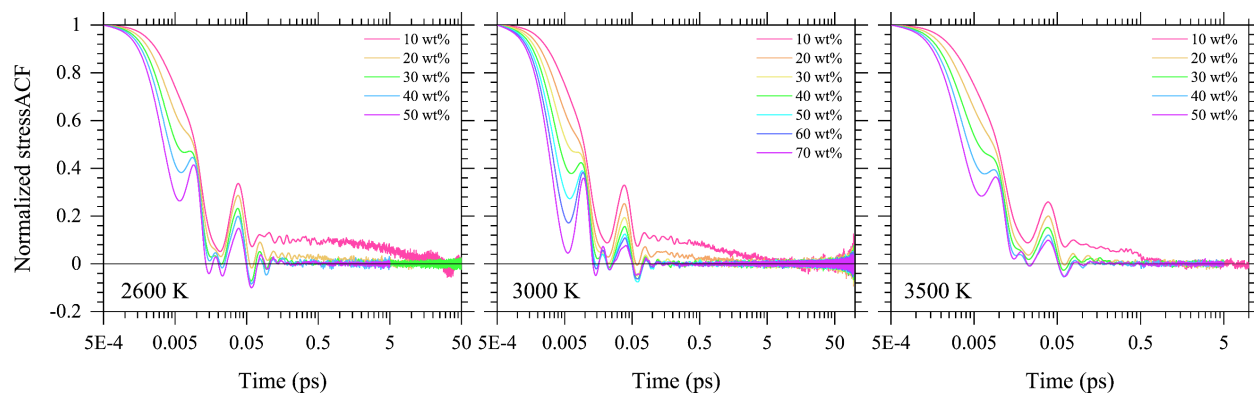


Figure S8. The normalized stress autocorrelation function (ACF) of the $\text{SiO}_2\text{-H}_2\text{O}$ systems at 2600 K, 3000 K, and 3500 K.

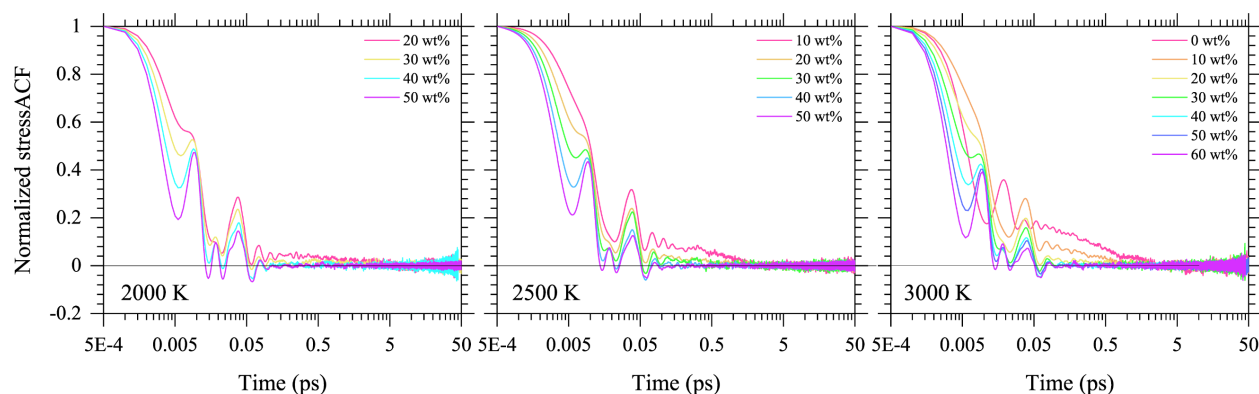


Figure S9. The normalized stress autocorrelation function of the $\text{NaAlSi}_3\text{O}_8\text{-H}_2\text{O}$ systems at 2000 K, 2500 K, and 3000 K.

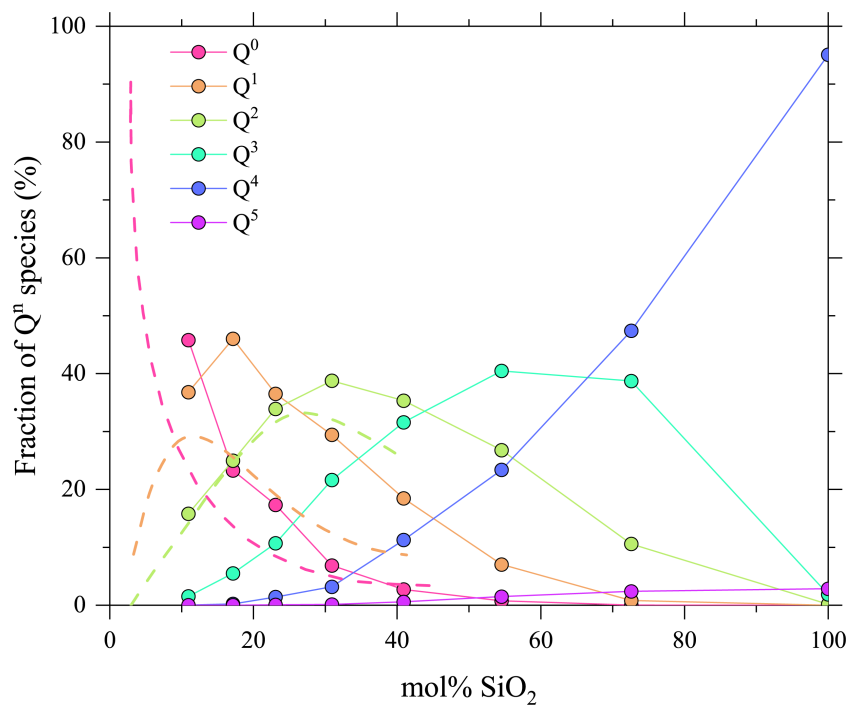


Figure S10. Fraction of Q^n species in the $\text{NaAlSi}_3\text{O}_8\text{--H}_2\text{O}$ systems at 3000 K as a function of SiO_2 content. The dashed curves represent the experimental Q^0 , Q^1 and Q^2 species in $\text{H}_2\text{O--Na}_2\text{O--SiO}_2$ fluids up to 873 K and 2 GPa (Steele-MacInnis and Schmidt, 2014).

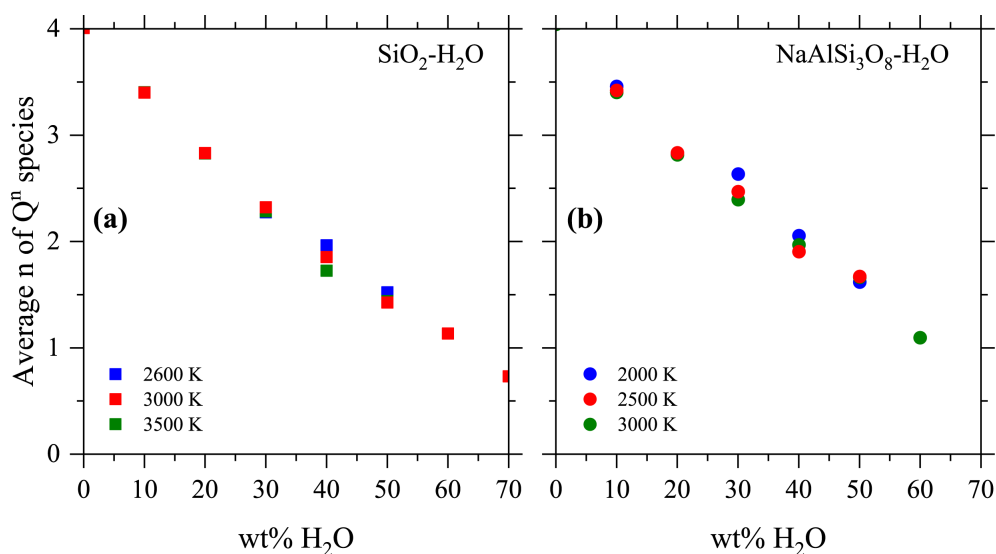


Figure S11. Average n of Q^n for the $\text{SiO}_2\text{--H}_2\text{O}$ and $\text{NaAlSi}_3\text{O}_8\text{--H}_2\text{O}$ systems.

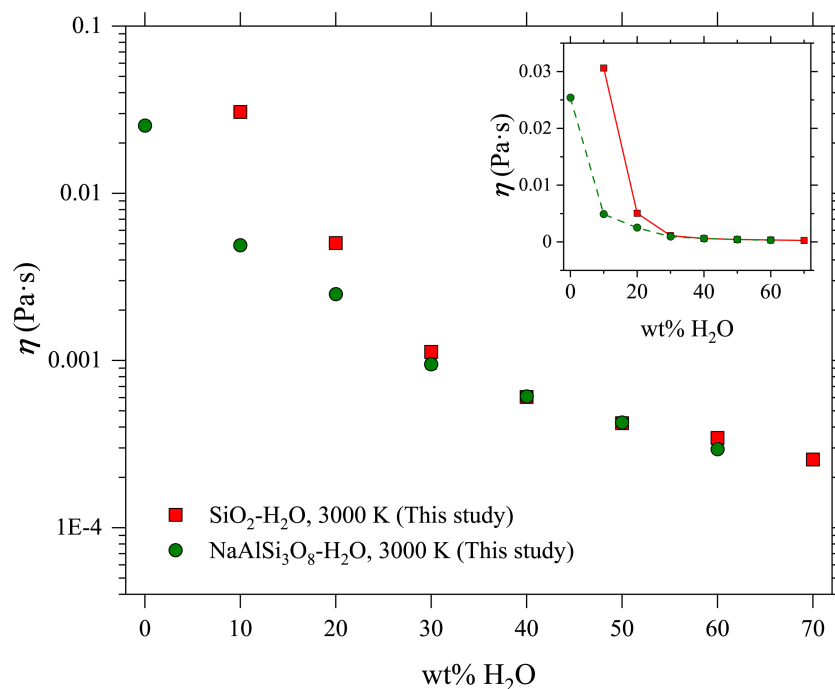


Figure S12. Viscosities of the $\text{SiO}_2\text{-H}_2\text{O}$ and $\text{NaAlSi}_3\text{O}_8\text{-H}_2\text{O}$ systems.

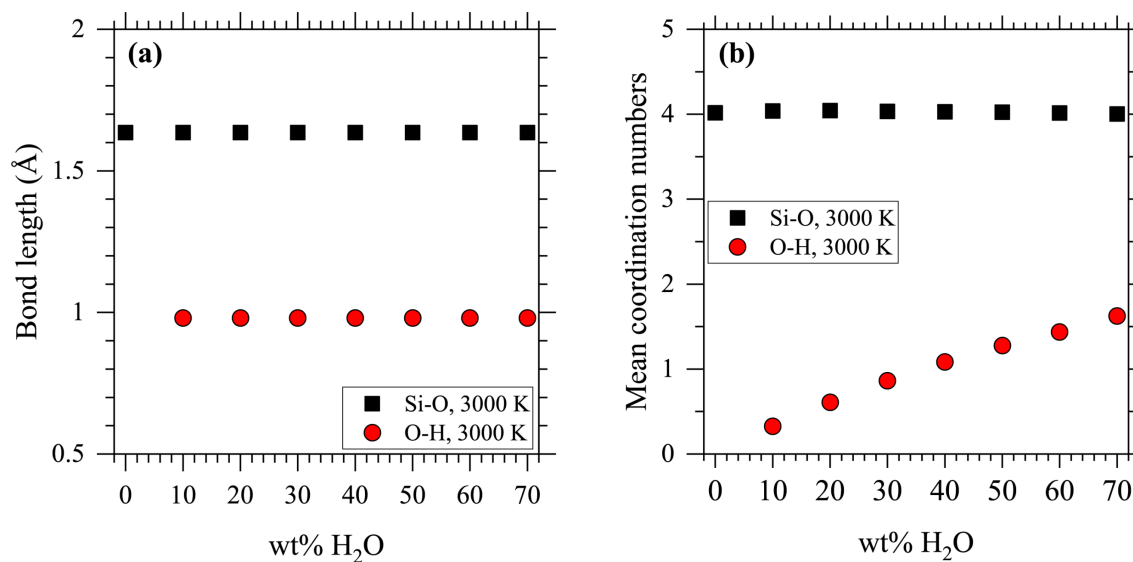


Figure S13. Si-O and O-H bond lengths and mean coordination numbers of the $\text{SiO}_2\text{-H}_2\text{O}$ systems.

Temperature has no effect on bond lengths and mean coordination numbers.

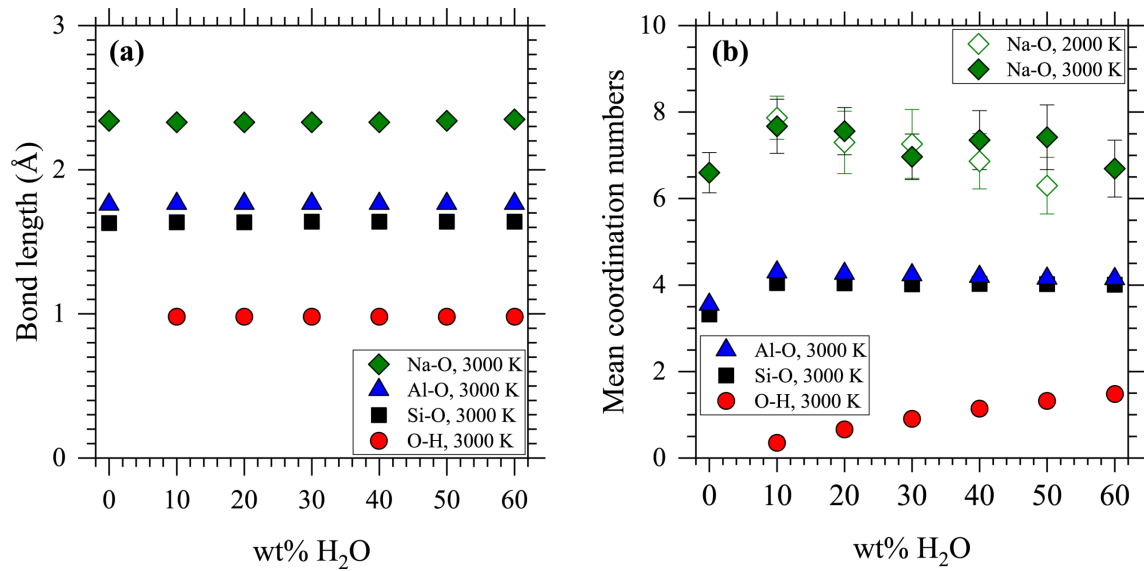


Figure S14. Na-O, Al-O, Si-O and O-H bond lengths and mean coordination numbers of the NaAlSi₃O₈-H₂O systems. Temperature has no effect on bond lengths and mean coordination numbers.

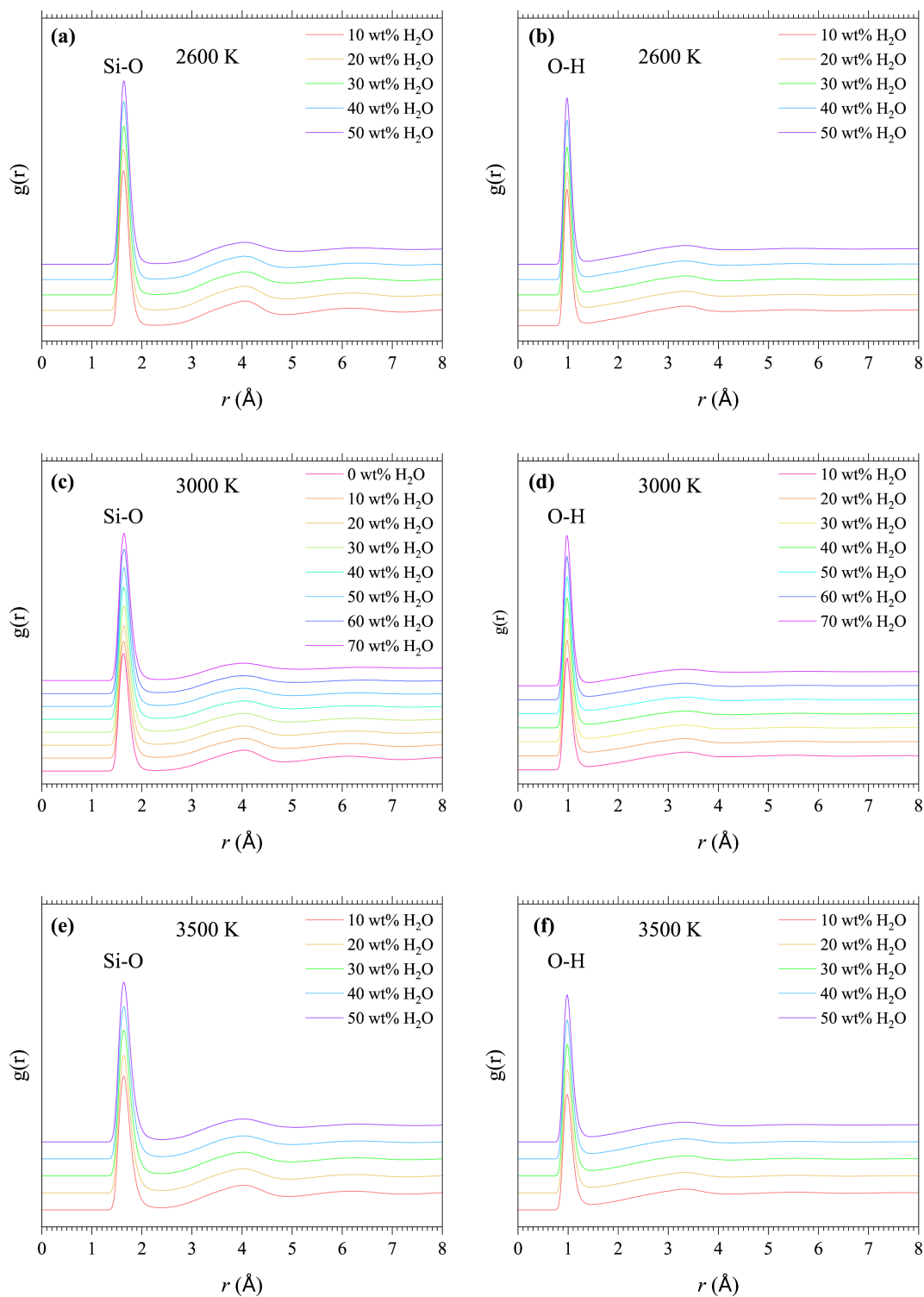


Figure S15. Radial distribution function of Si-O and O-H pairs in the SiO_2 - H_2O systems.

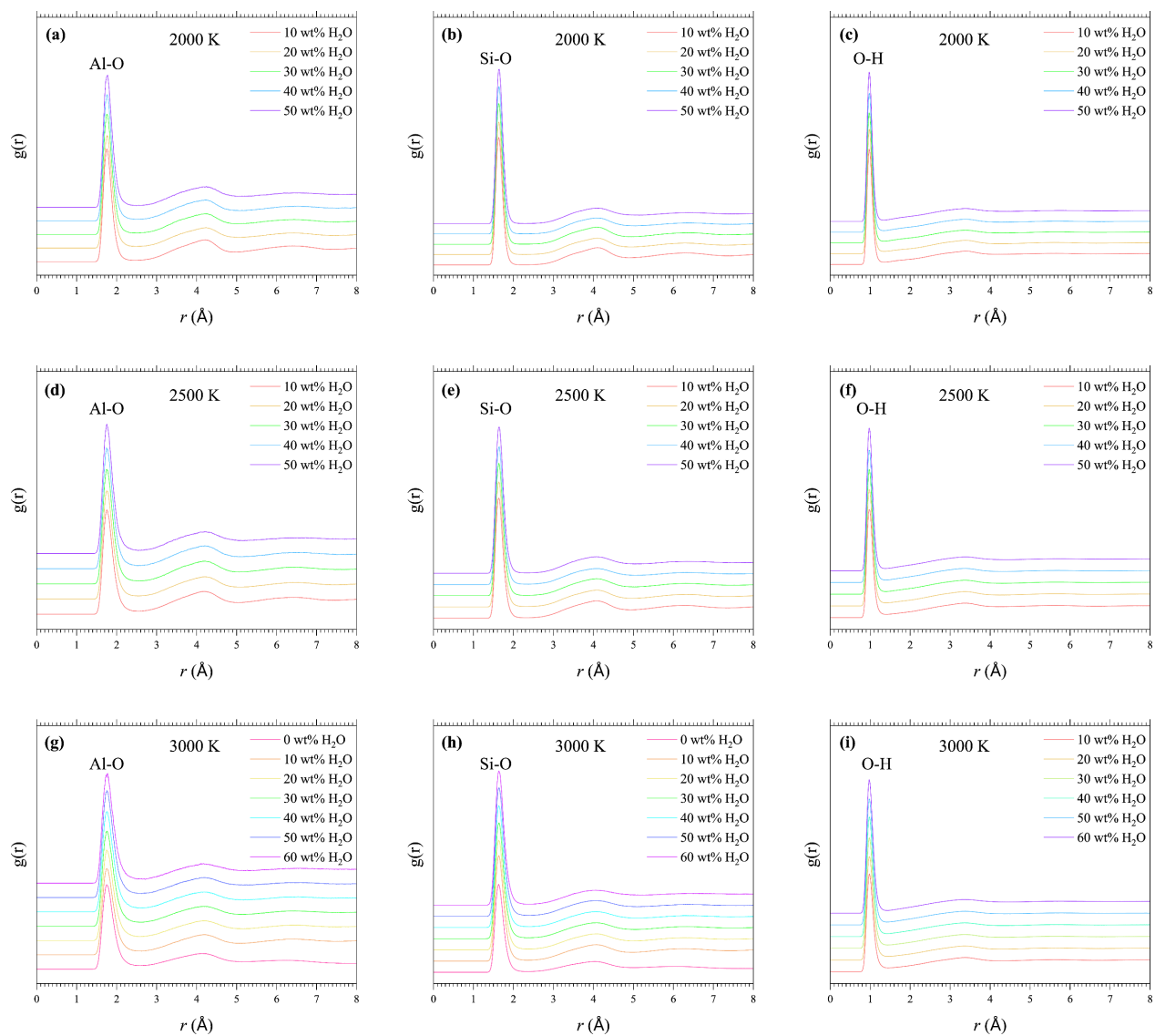


Figure S16. Radial distribution function of Al-O, Si-O and O-H pairs in the $\text{NaAlSi}_3\text{O}_8\text{-H}_2\text{O}$ systems.

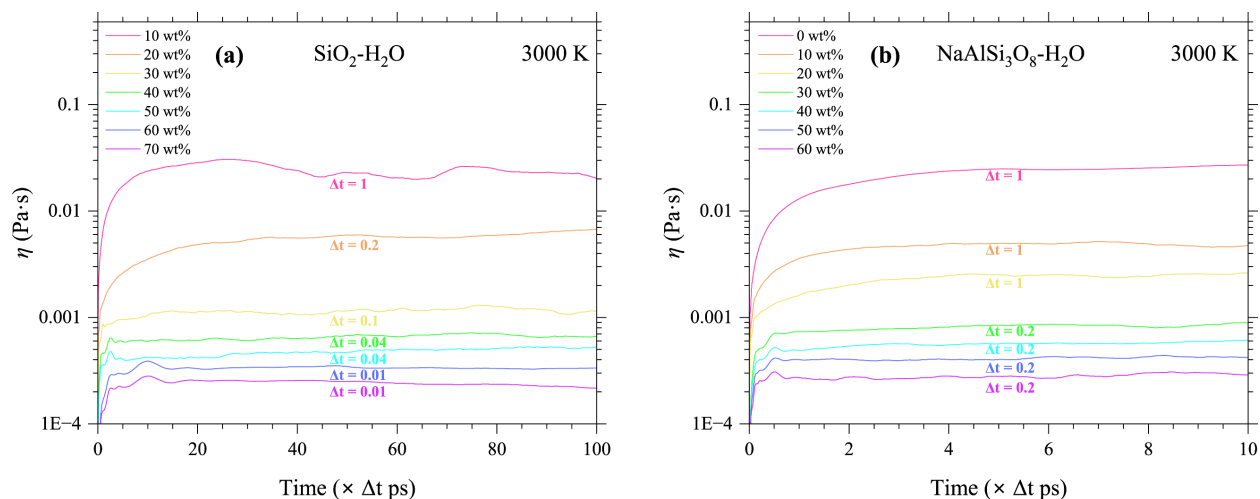


Figure S17. Viscosity-time relationships for the $\text{SiO}_2\text{-H}_2\text{O}$ and $\text{NaAlSi}_3\text{O}_8\text{-H}_2\text{O}$ systems.

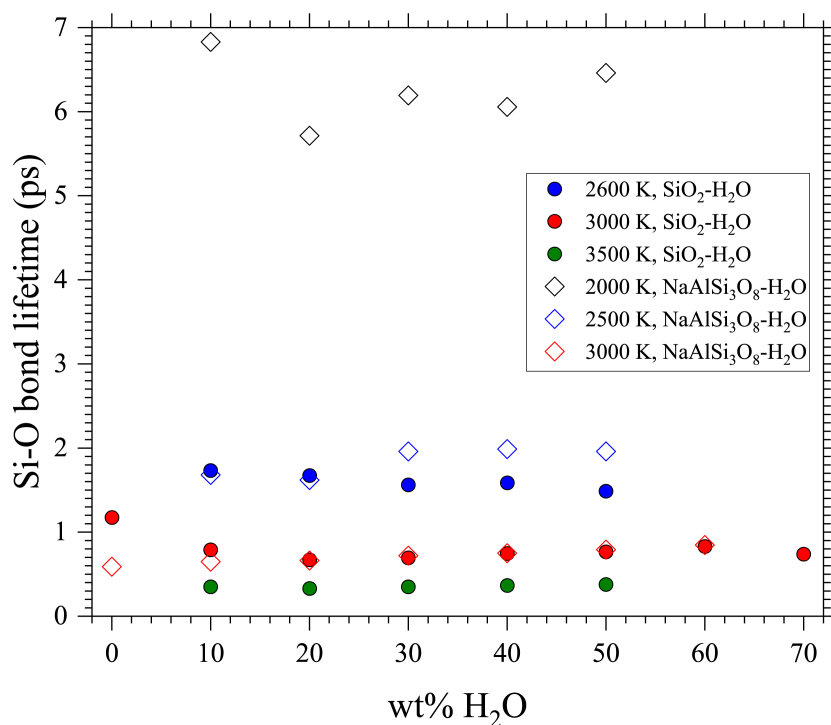


Figure S18. Average Si-O bond lifetime in the $\text{SiO}_2\text{-H}_2\text{O}$ and $\text{NaAlSi}_3\text{O}_8\text{-H}_2\text{O}$ systems.

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