

Viscosity of silicate network phase-separating from aqueous fluid

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Silicates and aqueous fluids are fundamental to multi-spatiotemporal-scale geodynamics on Earth. An important phenomenon of their mixtures is phase separation or phase demixing (Hack et al., 2007), creating different liquid phases; This is significant for magma differentiation (Kawamoto et al., 2012) as similar as crystallization, for magmatic dynamics from the crustal deep to the surface (Allabar and Nowak, 2018), and maybe, for crustal activities (Koyama, JpGU [2012, 2019]). To these time-consuming processes, viscous dynamics of the silicate phase slower than that of the aqueous one is crucial, but almost unknown. We have to consider the thermodynamic relations between the components in the non-equilibrium process, when using usual flow mechanism as the base.

This mechanism in melt silicate network, although still debated, can basically be interpreted as the conformation change through diffusion of the formers, Si, bridging O, Al, etc. (Mysen & Richet, 2005, Stebbins, 2016). In mixing hydrous melts, the increasing H₂O drastically lowers the viscosity; The modifier H with its non-bridging O makes the network variable through the dissociation and polymerization as the terminal OH and free H₂O, and through the inter-strand hopping as the OH (ibid.). However, these softening actions of H (OH & H₂O) through the reactive former-modifier collisions must be realized under the entropic mixing. Then, in coexisting melts of silicate network (formers) and aqueous fluid (modifiers) under SiO₂-H₂O demixing, are the actions of H the same as that in the mixing or not? Here, I expect to be not, or rather to be the opposite. This is because that the enthalpic demixing, overcoming the entropic mixing with the “both-like-both” one, leads to the “likes-like-likes” situation for both groups; The H₂O and OH, favoring themselves more than the network, must prevent the dissociation but promote the polymerization. Under this preferential polymerization the network shrinks with the strand tension instead of the fracturing to demix from the fluid. The tension must suppress the strand vibration creating the inter-strand collision for the OH hopping and for the network-former diffusion itself. Thus, these “inert” actions of H on the network in the demixing must be the hardening or the viscosity and solidity increasing, even in the highly hydrous situation, contrasted with the softening in the mixing. This state must be transient under the intense demixing in the early stage (transient gel: Tanaka, 2017; Koyama & Tanaka, 2018). Further, the hardening will be assisted by the actions of other modifiers, Na, K, Mg, etc., similar to that of H and by the enthalpy overwhelming the entropy in lower temperature and pressure.

Although having to be verified theoretically and experimentally, these qualitative results of the asymmetric behaviors of the silicate network and aqueous fluid in their demixing will connect to magmatic and seismic processes: the convection suppression due to the elastic resistance of the demixing network in the chamber and the explosive eruption to the surface through the brittle fracturing of the network under rapid volatile expansion; a hypothetical phase-separating continental lower crust of the shrinking-network and diffusing-fluid for compressive and fluid-concerned intracrustal seismicity (Koyama, JpGU [2012, 2019]).

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