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Oxidation-state dependence of rheology in peralkaline glasses of phonolitic composition

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The precise description of magmatic melts rheology at the glass transition is crucial in understanding dynamic processes in volcanology. The glass transition has been described to scale with the viscosity of the material according to Maxwell's relaxation theory for viscoelastic liquids (Dingwell and Webb, 1989). The temperature dependence of the viscosity of multi-component systems can adequately be calculated using empirical models such as Hess et al. (1996), Giordano et al. (2008) and Hui and Zhang (2008); yet, within these calculations, the influence of oxidation state has been so far considered minor and was consequently neglected. The rheological behavior of some iron-rich silicate melts has shown noteworthy oxidation state-dependent variations (Cukierman and Uhlmann 1974, Dingwell and Virgo 1987). The focus of our study is to improve the viscosity models by investigating the necessity of an additional redox-parameter.

Thirteen re-melted glass samples of natural phonolitic composition (peralkaline lavas with 8.5 wt. % FeO_{tot}) were produced under different oxygen fugacity (fO_2) conditions in a CO/CO_2 gas-mixing furnace. Their oxidation-state (Fe^{3+}/Fe_{tot}) ranges from 0.44 to 0.93 (±0.05). The viscosity above the liquidus was recorded via the concentric cylinder technique at a constant temperature of 1186 ˚C. Additionally, viscosities were measured in the interval of $10^7$to 10^{11}Pa swith temperatures up to 900 ˚C at ambient pressure via a BAEHR micro-penetration viscometer. Glass transition temperatures (T_g) have been determined with a constant heating/cooling rate of 10K/min on a SETARAM Sensys evo DSC using the peak of the specific heat capacity curve.

Under a constant temperature in the super-liquidus state, the viscosity increases strongly with increasing fO_2. In the sub-liquidus state, the measured calorimetric $T_g$is shifted to lower temperatures as the ratio of ferrous/total iron decreases from 638 ˚C to 610 ˚C. However, there is no equivalent measurable effect of the oxidation-state on super-cooled melt viscosity at T_g, within the precision of the micro-penetration experiment (all values are scattered around 10.10 ± 0.2 Pa s). Our results show a large discrepancy of 0.5 log units compared to the predicted viscosity at T_g using the kinetic model of Gottsmann et al. (2002). We further investigate additional relatively iron-rich compositions aiming for the development of an improved model for the viscosity prediction at the glass transition.

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