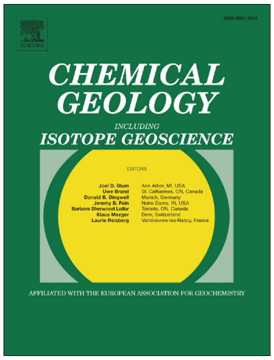

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BULK SOLUBILITY AND SPECIATION OF H_2O IN SILICATE MELTS

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ABSTRACT

The bulk solubility and speciation of H_2O in silicate melts of virtually any composition is predicted from first principles with a satisfactory precision. The solubility of molecular water is first predicted from the Scaling Particle Theory (SPT) coupled with an *ab initio* assessment of the electronic, dispersive and repulsive energy terms based on the Polarized Continuum Model (PCM). The Silver-Stolper ideal homogeneous speciation model is then applied to compute the fractional molar amount of neutral hydroxyl functionals [OH]° in the melt and the computed [OH]° amount is added to the molecular form [OH_2]. The Hydrogen Bonding (HB) electrostatic contributions to the stabilization of molecular water [OH_2] in solution are then resolved through an inverse non-linear minimization procedure on the basis of an extended dataset (970 samples) of experiments concerning the H_2O saturation hyper-surface carried out in the last half century. The Gibbs free energy of solution ΔG_S, the ΔG_{HB} contributions and the energy terms involved in the homogeneous speciation reaction are shown to be consistent with first principles. The procedure is fully predictive (i.e. no need of an initial hint about approximate bulk amounts of H_2O in solution) and sufficiently accurate to be proposed as an exploratory tool (mean absolute accuracy of ~ 2.1 kJ/mole in terms of energy and ~1.7% in terms of fractional molar amount X_{H2O} per unit mole of liquid).

KEYWORDS