

Pressure dependence of the solubility of Ar and Kr in melts of the system $\text{SiO}_2\text{-NaAlSi}_2\text{O}_6$

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ABSTRACT

The solubilities of Ar and Kr in supercooled melts in the system $\text{SiO}_2\text{-NaAlSi}_2\text{O}_6$ have been studied at temperatures of 1200 °C (SiO_2), 1000 and 750 °C ($\text{NaAlSi}_3\text{O}_8$), and 800 °C ($\text{NaAlSi}_2\text{O}_6$) at pressures from 200 to 6000 bar. Gas sorption experiments were performed in an internally heated pressure vessel, and noble gas concentrations in quenched samples were analyzed by absolute analytical techniques [gas chromatography (GC), and thermogravimetry (TG), gravimetry]. Comparison of the results from this study with other absolute techniques (Rutherford-backscattering, mass spectrometry) is possible for silica and exhibit excellent agreement. This agreement leads us to conclude that our silica glasses with an Ar concentration of 1.02 wt% and a Kr concentration of 0.54 wt% are suitable as standard materials. Solubility increases linearly with increasing pressure for all three compositions. In SiO_2 the solubility of Ar and Kr increases with pressure with nearly parallel slopes, whereas Ar and Kr exhibit diverging slopes in $\text{NaAlSi}_3\text{O}_8$ and $\text{NaAlSi}_2\text{O}_6$. The linear relationship between fugacity and dissolved gas below 2–3 kbar, indicates that Henry's law constants (in units of 10^6 bar) for Ar in SiO_2 , $\text{NaAlSi}_3\text{O}_8$, and $\text{NaAlSi}_2\text{O}_6$ are 0.94, 3.15, and 5.71, respectively, and for Kr are 1.38, 5.69, and 9.28, respectively. The experimental results can be modeled by two alternative thermodynamic approaches. First, assuming mixing of noble gases with the network of the melt, partial molar volumes (in cm^3/mol) can be calculated in SiO_2 , $\text{NaAlSi}_3\text{O}_8$, and $\text{NaAlSi}_2\text{O}_6$ for Ar as 25.9 ± 1.3 , 21.1 ± 1.6 , and 21.1 ± 0.7 , respectively and for Kr as 24.3 ± 1.8 , 27.6 ± 0.8 , and 24.7 ± 0.9 , respectively. Second, assuming the volume change upon solution to be zero, a Langmuir-Isotherm can be applied yielding saturation levels M (in units of 10^{20} sites/ cm^3 melt) in SiO_2 , $\text{NaAlSi}_3\text{O}_8$, and $\text{NaAlSi}_2\text{O}_6$ for Ar as 3.82 ± 0.6 , 1.33 ± 0.2 , and 0.72 ± 0.08 , respectively and for Kr as 3.49 ± 0.7 , 0.56 ± 0.05 , and 0.42 ± 0.06 , respectively. Highest solubility is observed in all three melts for Ar. Solubility decreases with increasing substitution of Si^{4+} by $\text{Na}^+ + \text{Al}^{3+}$. The dependence of solubility on composition supports the structural model for melts along the join $\text{SiO}_2\text{-NaAlSi}_2\text{O}_6$ based on a stuffed tridymite-like structure.