The dissolution mechanism of water in alkaline earth silicate and aluminosilicate melts: One view from ^{1}H MAS NMR

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Despite its relevance to mafic to ultramafic magmas, studies on the dissolution mechanism of water in alkaline earth silicate melts have been limited. Here we report ^{1}H MAS NMR results on hydrous $CaMgSi_2O_6$(Di)-SiO_2(Qz) and Di-$CaAl_2Si_2O_8$(An) join glasses quenched from melts, to address this issue. 0.78Di·0.22Qz glasses (NBO/T=1.75) with 0.93 and 2.0wt% H_2O, and 0.62Di·0.38An glasses (NBO/T=0.90) with 0.75 and 1.7wt% H_2O were synthesized at 2kbar and 1400~1500˚C for 2~4 hours in an IHPV. No paramagnetic impurities were doped.

For the Di-Qz glasses, the ^{1}H MAS NMR spectra resemble those of a similar sample containing 0.2wt% Gd_2O_3(Kanzaki and Xue, 2002), although peaks are sharper and the intensity decay with delay τ in the Hahn-echo spectra much slower than for the latter. There are two peaks near 4.7 and 1.3ppm of nearly equal height, and a high-frequency shoulder extending to about 17ppm. The 4.7-ppm peak intensity decreases with decreasing water content and increasing τ. As discussed previously, the 4.7-ppm and its high-frequency shoulder, common to hydrous silicate glasses, can be ascribed to molecular H_2O and H-bonded SiOH. The 1.3-ppm peak is due to non-H-bonded OH: Similar peaks have not been observed for hydrous silica or other Al-free silicate glasses, but are typical of CaOH/MgOH in crystalline silicates and phosphates, and thus can be ascribed to (Ca,Mg)OH.

The ^{1}H MAS NMR spectra for the hydrous Di-An glasses contain an asymmetric peak with a maximum near 2.4ppm, and a high-frequency shoulder near 4ppm extending to about 17ppm. The intensity of the high-frequency shoulder decreases with decreasing water content and increasing τ: the Hahn-echo spectra with τ= 50ms contain an asymmetric peak with a maximum near 1.6ppm. These spectra thus resemble those of the hydrous Di-Qz glasses. The component near 1.6ppm is possibly due to (Ca,Mg)OH, although contribution from AlOH cannot be excluded. The high-frequency shoulder is likely from molecular H_2O and H-bonded SiOH and/or bridging OH.

In conclusion, our study revealed that free OH ((Ca,Mg)OH) is an important water species in hydrous Di-Qz melts, and possibly also in Di-An melts, and thus must be taken into account in modelling hydrous silicate melts.

References

Kanzaki M. and Xue, X. (2002) *Geochim. Cosmochim. Acta*, **66**, A382 (abstr.)