The structure of highly polymerised silicate melts near the 'charge-balanced' join: Insights from shear viscosity measurements.

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Few concepts about the structure of silicate melts are as generally accepted as the notion that, at one atmosphere, a subaluminous melt ($M^{n+}/n\text{Al} = 1$; M is a metal cation, e.g. Na^+, Ca^{2+}) is fully polymerised (i.e. contains no non-bridging oxygens). In this case, at constant silica content, melt viscosity may be expected to show a maximum at the subaluminous join, where no non-bridging oxygen occur. As a test of this hypothesis, we have measured the shear viscosity of over 100 compositions close to the subaluminous join ($M^{n+}/(M^{n+}+n\text{Al})$ in the range 0.60 to 0.40) along isopleths of SiO_2 (generally 75, 67 and 50 mol% SiO_2). The systems Na_2O-Al_2O_3-SiO_2 (NAS), CaO-Al_2O_3-SiO_2 (CAS) and MgO-Al_2O_3-SiO_2 (MAS) have been studied. In the system NAS, a single viscosity maximum is observed near the subaluminous join at all the investigated silica isopleths. However, its exact position is not generally at Na/Al =1, but deviates toward the peraluminous side. In the system MAS a broad maximum in field of excess magnesium is clearly identified at 1600°C at all silica contents. The behaviour of melts in the system CAS is more complex, typically showing a broad maximum in viscosity in the peraluminous field at high temperature. These results clearly indicate that currently employed models for the structure of 'highly polymerised' liquids are inadequate and require revision. The detailed implications of our results for the structural role of aluminium and oxygen in melts containing different cations will be discussed.