

## Oxidised primary arc melts trapped in sub-arc mantle xenoliths and the origin of calcalkaline or low-Fe rock suites

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Tholeiitic and calcalkaline rock suites respectively predominate at mid-ocean ridges and subduction zones. The low-Fe character of calcalkaline suites has mainly been attributed to the earlier phase saturation with Fe-Ti oxides relative to ferromagnesian silicates therein. However, experiments have shown that this requires either a high oxidation state or high H<sub>2</sub>O contents (or both) in the original magmas, while the former remains strongly debated in the absence of direct evidence from primary subduction zone melts yet.

Here we report measurements of the S oxidation state in primary picrite-boninite melt inclusions and the Fe oxidation state in coexisting Cr-rich spinel. The melt inclusions and their host spinel occur in primitive magmatic veins (hornblende-bearing websterite) cutting sub-arc mantle peridotites. Spinel analysis reveals a positive correlation between the atomic ratios  $Fe^{3+}/(Fe^{3+}+Fe^{2+})$  and  $Mg/(Mg+Fe^{2+})$  with primitive values of  $0.32\pm 0.02$  and  $0.69-0.72$ , respectively. These values, together with the presence of oxidised sulfur ( $S^{6+}$ ) in the melt inclusions, indicate a high oxygen fugacity ( $fO_2$ ) 1-2 log units above the synthetic fayalite-magnetite-quartz (FMQ) buffer for the primary picrite-boninite melts. Because the spectrum of melt inclusion major element compositions describes calcalkaline and low-Fe trends, our observations directly confirm a strong control of the early crystallization of  $Fe^{3+}$ -bearing spinel on the compositional evolution of oxidised primary melts. The onset of the formation of arc-related calcalkaline or low-Fe rocks in the Proterozoic eon may thus be directly related to the more oxidised nature of their mantle sources. Our observations also imply that the large impact of the formation of liquidus spinel on the  $Fe^{3+}/(Fe^{3+}+Fe^{2+})$  of evolving oxidised magmas can alter the mantle source redox state estimated from, e.g., basalts derived after spinel fractionation from more primitive and oxidised picrite-boninite parental melts.