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Iron speciation and redox state of mantle eclogites: Implications for ancient volatile cycles during mantle melting and oceanic crust subduction

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Kimberlite-borne mantle eclogite xenoliths of Archaean and Palaeoproterozoic age are commonly interpreted as representing former oceanic crust. As such, they may retain a memory of the redox state of the ancient convecting mantle sources that gave rise to their magmatic protoliths and which controls the speciation of volatiles in planetary interiors. Mantle eclogite suites commonly include both cumulate and variably evolved extrusive varieties [1], which may be characterised by initial differences in Fe3+/Fetotal. Recent Fe-based oxybarometry shows mantle eclogites to have fO_2 relative to the fayalite-magnetite-quartz buffer (ΔFMQ) of -3 to 0, whereby low fO_2 relative to modern MORB may relate to subduction of more reducing Archaean oceanic crust or loss of ferric Fe during partial melt loss [2]. Indeed, using V/Sc as a redox proxy, it was recently shown that Archaean mantle eclogites are more reduced than modern MORB (ΔFMQ-1.3 vs. ΔFMQ -0.4) [3]. However, in the warmer ancient mantle, they were also subject to modification due to partial melt loss upon recycling and, after capture in the cratonic mantle lithosphere, may be overprinted by interaction with metasomatic melts and fluids.

In order to help further constrain the redox state of mantle eclogites and unravel the effect of primary and secondary processes, we measured Fe3+/Fetotal by Mössbauer in garnet from mantle eclogites from the Lace kimberlite (Kaapvaal craton), comprising samples with melt- and cumulate-like oceanic crustal protoliths as well as metasomatised samples. Fe3+/ΣFe in garnet shows a strong negative correlation with jadeite content and bulk-rock Li and Cu abundances, suggesting increased partitioning of Fe3+ into jadeite in the presence of monovalent cations with which it can form coupled substitutions. Broad negative correlation with whole-rock $Al2O_3/TiO_2$ and positive correlation with ΣREE are interpreted as incompatible behaviour of Fe3+ during olivine-plagioclase accumulation (exclusion of TiO_2 and REE). NMORB-normalised Nd/Yb, as a proxy of partial melt loss from subducting oceanic crust (<1) and metasomatism by typically LREE-enriched liquids (>1), shows no relationship with Fe3+/ΣFe. ΔFMQ, calculated using recently calibrated oxybarometers [2,4], broadly decreases with increasing pressure, which is ascribed to increasing garnet modes in metabasalts into which Fe3+ can be sequestered, similar to peridotite.

The very low Fe3+/ΣFe, like V/Sc, appears to be a relatively robust indicator of low-pressure igneous processes and, potentially, the redox state of the ambient convecting mantle source to the protoliths of mantle eclogites. In contrast, Fe-based fO_2 predominantly reflects pressure and bulk composition, and controls the speciation and mobility of volatiles in mafic heterogeneities during subduction and after emplacement in the cratonic mantle. The highly reduced nature of Archaean oceanic crust combined with further reduction upon pressure increase suggests that refractory graphite/diamond will be the stable carbon species. This may have prevented significant carbon output in Archaean subduction zones.

[1] Aulbach and Jacob (2016) Lithos 262: 586-605; [2] Stagno et al. (2015) Contrib Mineral Petrol 42: 207-219; [3] Aulbach and Stagno (2016) Geology 44: 751-754; [4] Vasilyev (2016) PhD Thesis, Australian Nat Univ