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Kaersutitic and Ti-pargasitic amphiboles and oxygen fugacity

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Since long time kaersutite and Ti-pargasite were studied because they are retained to be useful for the estimation of fO_2 and fH_2 conditions of their hosting rocks (especially for mantle derived rocks and nodules). These amphiboles have currently low hydrogen (even close to 0) content than the classical 2 a.p.f.u.. This feature is normally attained (separately or together) by substitution vectors: 1) ${}^{VI}Fe^{3+}_xO^{2-}_x \leftrightarrow {}^{VI}Fe^{2+}_{1-x}(OH)^{1-}_{1-x}$ and 2) ${}^{VI}Ti^{4+}_xO^{2-}_x \leftrightarrow {}^{VI}Al(\text{or } {}^{VI}Fe)^{3+}_{1-x}(OH)^{1-}_{1-x}$. However, albeit it is now experimentally demonstrated that these amphiboles can grow in laboratory under high fO_2 and low fH_2 conditions, it is still an open question if the high oxidation state (high Fe^{3+}/Fe^{TOT} ratio) of these natural amphiboles reflects a primary feature of crystallization, or is acquired during the transport from their source region up to higher crustal levels. The latter condition, particularly, may represent a disequilibrium signature, driven by kinetic factors, mainly magma cooling and/or decompression rates. In order to elucidate these aspects, we have analysed about 200 kaersutites chemical analyses from literature. The accuracy of these data was re-evaluated on crystal-chemical grounds. Only high-quality data (sum of C-site cations ranges between 4.75 and 5.25 a.p.f.u and T-site = 8 a.p.f.u.) were retained for further analysis. All these calcic amphiboles show a good positive correlation between temperature and the amount of ${}^{C}Ti$, ${}^{C}Mg$ and alkaline cation at A-site. Iron shows a negative trend, whereas the Al content at both C- and T-sites is mainly controlled by the pressure of crystallization. The Mg and Fe^{TOT} amounts are inversely related between them for all amphiboles ($R^2 > 0.8$). This correlation is mainly due to Mg *vs* Fe^{2+} for intrusive and synthetic amphiboles; conversely, for volcanic kaersutite and

Ti-pargasite amphiboles bivalent iron is slightly correlated with Mg; in comparison, ferric iron *vs* Mg do not shown any clear relation. Therefore, we interpret that the oxidation of these volcanic samples occurred mainly after their growth. An inverse and linear relation, as expected, between Ti+Fe^{3+} *vs* H+F is high for all the retained analysed amphiboles. This relation is mainly imposed by the trend of volcanic samples, and is markedly due to the relation between Fe^{3+} and H+F (F is low for these entire sample). The same relation for intrusive and synthetic amphiboles is in comparison low. Conversely, the relation between H+F *vs* Ti is low for volcanic amphiboles and relatively high for intrusive and synthetic samples. The different behaviours shown by kaersutites and/or Ti-pargasites in volcanic and intrusive/synthetic rock are significant. The major difference invariably involves the oxidation state of Fe. Ti amount seems to play a minor role with respect to the final amount of H for volcanic amphiboles, i.e. H can be low or relatively high for both low and high amount of Ti. In other words, the oxidation trends of these volcanic amphiboles is principally acquired during the magmas rising up from their source regions across the Earth crust. Therefore, growth of these volcanic amphibole phases in an oxidized upper mantle is not supported. H-loss is likely to be due to diffusive processes, mainly occurring at high T, with the consequent oxidation of Fe^{2+}. The presence of high amount of Fe^{3+} at M1- and M3-site can be considered as an evidence of this acquired oxidation trend. Indeed, such a redox exchange could reflect the crystal attempt to maintain equilibrium with the surrounding degassed magma, under large chemical water/hydrogen gradients. In this view, oxidizing rates may be related to the $\Delta P/\Delta T/\Delta t$ gradients, such that the observed dehydrogenation may represent a valuable tool to infer the ascent paths of alkali basalts.