

## **The tantalite-tapiolite gap as an indicator of disequilibrium in granitic pegmatites : constraints from crystallization experiments**

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Crystallisation experiments of tapiolite [Fe(Ta>Nb)<sub>2</sub>O<sub>6</sub>] and tantalite [Mn(Ta>Nb)<sub>2</sub>O<sub>6</sub>] from water-saturated pegmatitic melts at 50 to 200 MPa and 800°C are used to investigate the role of disequilibrium crystallisation on the compositional gap limits of this common Nb-Ta oxide pair. The results show that isothermal disequilibrium crystallisation strongly impacts on Nb-Ta fractionation and this effect prevails over the solubility effect in evolved systems in which high degrees of supersaturation can be reached. Thus, even if global equilibrium in terms of the solubility of Nb-Ta-bearing minerals is attained, the Ta/(Nb+Ta) ratio in the crystals may differ significantly from equilibrium. It implies that Nb-Ta fractionation in Nb-Ta oxides is controlled by crystallisation kinetics rather than chemical fractionation driven by differential solubility. The experimental tantalite-tapiolite gap limits are compared with natural tantalite-tapiolite pairs from rare-element pegmatites and suggest that disequilibrium crystallisation may be responsible for the shift of the tapiolite and tantalite gap limits away from their stable domain of composition. We suggest that extreme Nb-Ta fractionation observed in highly evolved pegmatitic melts is primarily linked to supersaturation and disequilibrium crystallisation of Nb-Ta-Ti-oxides rather than F-complexing of Ta or fluid exsolution. Our results have important implications for the understanding of extreme fractionation and pegmatite-forming processes.