

REE partitioning between calcite and apatite - a tool for identifying truly magmatic vs. secondarily altered carbonatites

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Numerous studies about geochemistry, formation and emplacement of carbonatites *a priori* assume that these are magmatic and represent melt composition. Unfortunately, almost all carbonatite rocks are not true melt compositions, but either represent primary cumulates or, more often, are strongly affected by fluid alteration (from deuteric to hydrothermal to meteoric) causing e.g. dissolution of $(\text{Na,K,H})_2\text{CO}_3$ minerals and recrystallization of calcite or dolomite. Experimental studies [1, 2] and melt inclusions [3] indicate that primary carbonatite melts contain >5-20 wt% $\text{Na}_2\text{O}+\text{K}_2\text{O}$, yet, carbonatite rocks generally have <1-2 wt%.

Our aim is to develop a tool that allows to determine whether the calcite or dolomite of a given carbonatite is primary. For this purpose, we experimentally determined calcite/apatite REE partition coefficients D^{CC-Ap}_{REE} in carbonatite melts under subvolcanic conditions (1-2 kbar, 700-1100 °C). The experiments yield calcite/apatite REE partition coefficients in a narrow flat band with D^{La}/D^{Lu} of 0.8-0.9 and D^{CC-Ap}_{MREE} increasing from 0.1 to 1.4 with temperature. Natural calcite-apatite pairs that are clearly primary (e.g. from perfectly unaltered cc-bearing nephelinites or the Kaiserstuhl carbonatite comb layers) show congruent REE distribution patterns. Instead, several massive carbonatites yield calcite-apatite D^{La}/D^{Lu} of 10^{-2} - 10^{-3} with substantial LREE-depletion, i.e. a clear hydrothermal signature. A complete survey of the available calcite/apatite data from natural carbonatites reveals that in most cases calcite is secondary.

The D^{CC-Ap}_{REE} hence enables us to determine whether the carbonate mineral(s) of a carbonatite is primary or not, i.e. whether the rock sample is of magmatic origin or has been affected by chemical alteration. We posit that only the former are suited for deciphering magmatic processes; trace element and isotopic composition of the latter are rather meaningless in terms of magmatic processes.

A key role of apatite stems from (i) early precipitation in most carbonate magmas, (ii) its high REE contents and (iii) if hydrothermally altered, secondary minerals are formed instead of apatite. Calcite instead has relatively low REE contents and can be easily recrystallized hydrothermally or through meteoric waters.

[1] Lee + Wyllie (1998), *JPet.* [2] Brooker + Kjarsgaard (2011), *JPet.* [3] Guzmics et al. (2011), *CMP*.