

The role of multi-stage process on REE enrichment in Kangankunde carbonatites, Malawi

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Rare earth element (REE) are critical metals for several high-performance materials in modern industry, especially utilized for green technology. Carbonatites, volcanic rocks enriched in the REE, are focused on as REE sources especially during recent years since the security of supply is crucial in changing international relationships. Previous studies indicate that the REEs in carbonatites are re-distributed during post-magmatic processes, however, the nature of the respective processes to REE enrichment has not been well understood. Here, we performed geological, petrological and geochemical investigations of the Kangankunde carbonatite complex, which is the largest deposit of the Chilwa Alkaline province in Malawi. The complex is enriched in light rare earth elements (LREES) with up to 7.8 wt% REE content and preserve signatures of carbonatitic magma crystallization followed by subsequent post-magmatic alteration, hence providing a good platform for understanding REE enrichment process during the carbonatite evolution.

Main units of the Kangankunde carbonatite complex are Mn- and REE-rich carbonatites which are concentrated on the center part of the complex, whereas dolomite carbonatite, carbonatite agglomerate, quartz-rich and apatite-rich carbonatites irregularly occur as veins mostly in the outer zone. Field observation indicates that earlier carbonatites were notably cut through by later (carbothermal) fluids, suggesting multiple and complex post-magmatic process occurrence. Sampling was carried out in the different lithological units of the complex. X-ray diffraction analysis and petrographic observations indicate that the most abundant REE-bearing mineral is monazite, whereas bastnaesite and synchysite were observed as minor components. Scanning electron microscope observation indicates that some monazite shows well developed $\sim 100 \mu\text{m}$ crystalline structure with porous and non-porous structures inside and outside crystals respectively, commonly occurring along dolomite edges, whereas others are observed as polycrystalline monazite. Back scatter images of $\sim 100 \mu\text{m}$ monazite show elemental zoning between core and rim. The rim shows Nd-rich and La-poor compositions compared to core, suggesting that monazite growth would occur under at least two alteration stages. Bulk isotopic analysis of carbon and oxygen indicates that the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ ranged from -4.79‰ to -0.13‰ and 14.21‰ to 29.48‰ respectively. Part of REE-poor samples show similar values to primary carbonatites, while most of REE-rich samples show heavier $\delta^{18}\text{O}$, which is consistent with magmatic fluid-related values in a previous study (Guarino et al., 2016).

Isotopic results suggest that minor primary magmatic signatures may have been preserved in the C isotopes, while the wide variation of isotopic O point towards post-magmatic processes at low temperatures, which could be responsible for the widespread alteration features throughout the complex. The interpretation is consistent with two distinct microscopic occurrences and elemental zoning of monazite. Our studies reveal two major stages of carbonatite evolution with the early phase having minimal REE enrichment while the post-magmatic processes may have played a significant role in the enrichment of REEs in the carbonatites.

Keywords: Kangankunde, Malawi, carbonatite, Rare Earth Element, carbon isotope, oxygen isotope

