

# Generation of Carbonatitic Melts via Mantle Melting and Melt Differentiation and Enrichment of Critical Metals

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Carbonatites are rare, carbonate-mineral-rich igneous rocks that are volumetrically minor on the Earth's surface. Yet, carbonatites are the primary source of niobium and Rare Earth Elements (REEs) critical for the targeted transition to clean energy technologies. Therefore, there is a renewed interest in understanding carbonatite petrogenesis and the enrichment processes in ore-forming elements. Here, we review the conditions of generation of carbonatitic melt in the mantle in various tectonic settings and present new experimental data on the differentiation of a primary, CO<sub>2</sub>-bearing alkali basalt, which may be a common mode of generation of carbonatitic melts in the shallow subsurface. We also discuss the role of magmatic fluorine in the REE budget of mantle-derived carbonatites.

Carbonatitic melts can form as the first melt from the ambient mantle or recycled lithologies at depths of  $\geq 300$  km in the oxidizing mantle, which may prevail beneath the ocean basins. However, such melts are not expected to surface as natural carbonatites as more volumetric basaltic melts prevail. Owing to cooler geotherms beneath continents and matured oceanic lithosphere, conditions may approach the carbonated peridotite solidus, and thus, most proximal, primary carbonatitic melts originate in these settings (e.g., Dasgupta, 2018). Yet, most natural carbonatites differ significantly both in major and trace element chemistry from mantle-generated carbonatitic melts.

To explore whether volatile-bearing basaltic melts can evolve to carbonatite, we conducted new phase equilibria experiments at 1.5-2.0 GPa and 1200-1500 °C. The phase assemblage of carbonated melt  $\pm$  olivine  $\pm$  spinel  $\pm$  clinopyroxene  $\pm$  calcite  $\pm$  vapor shows that the initial stages of crystallization result in alkalic residual melts, which subsequently transition to calcio-carbonatite compositions with  $>80$  wt.% crystallization. Our results indicated that crystallization at mid-lithospheric depths remains a viable mechanism of carbonatite petrogenesis without involving liquid immiscibility. Through a separate series of trace element doped experiments, we evaluated the effects of fluorine on mineral-carbonate melt (CM) partitioning of REEs and HFSEs. Fluorine in CM lowers the partition coefficients of olivine-CM, clinopyroxene-CM, and spinel-CM. Therefore, magmatic REE enrichment in carbonatite is partly helped by REE-F complexation in the incipient melt during mantle melting, melt metasomatism, and extreme fractional crystallization.