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Deciphering the rare earth elements enrichment in carbonatites: A fluid inclusions perspective.

MR. MOHSIN RAZA[1,2,3], MATTHEW STEELE-MACINNIS[3], DR. R. JOHANNES GIEBEL[4,5], JOCHEN KOLB[6] AND BENJAMIN F. WALTER[7]

[1]Karlsruhe Institute of Technology
[2]Bacha Khan University Charsadda
[3]University of Alberta
[4]University of the Free State
[5]Technische Universität Berlin
[6]Institute for Applied Geosciences, Karlsruhe Institute of Technology
[7]Universität Tübingen

Carbonatites are key hosts for critical metals, but the processes leading to metal enrichment are still under debate. Nevertheless, there is a broad consensus that fluids exsolved from carbonatitic melts play an important role. A systematic investigation of fluid inclusions trapped during the evolution of a carbonatite offers direct insights into the mineralizing fluids. The present contribution focuses on comparative microthermometry and Raman spectroscopy of fluid inclusions hosted in calcite, apatite, fluorite, and quartz from carbonatites of the Nooitgedacht, Tweerivier (both South Africa) and Amba Dongar (India) complexes, and quartz from fenites (Tweerivier complex). This includes various emplacement depths and whole rock REE budgets to decode the history and compositional diversity of involved fluids.

The results reveal various fluid types: (I) (H_2O-NaCl-CO_2-SO_4) with homogenization temperature (T_h) of 450->600°C, (II) (H_2O-NaCl-SO_4) with T_h=350-500°C, (III) (H_2O-NaCl-CO_2) with T_h=250-400°C, (IV) (H_2O-NaCl; 5-15 wt.% $NaCl_{eq}$) with T_h=120-200°C, and (V) CO_2-rich vapor inclusions (without NaCl) containing minor CH_4 and N_2. While types I, II, and IV occur in REE-rich samples, types III, IV, and V only appear in REE-poor samples. It is also noteworthy to mention that type III and V inclusions are mostly observed in fenite, while type IV is abundant in late-stage hydrothermal mineral assemblages. Sulfate daughter minerals (e.g., arcanite, anhydrite, glauberite) are also observed only in REE-rich samples. Sulfates like baryte or celestine are in textural equilibrium with REE minerals. This suggests a genetic link by the model that REEs are transported as sulfate complexes.

REE mineral precipitation occurs through mixing between Ba rich and SO_4 rich fluids, and sulfate-ligand breakdown (due to the precipitation of baryte). The presence of volatile rich (H_2O, CO_2) fluid inclusions in fenites suggests that fenitization is driven by fluids expelled from carbonatitic melt. Low-temperature, aqueous inclusions (H_2O-NaCl) in Amba Dongar indicate late-stage hydrothermal activity, which may have remobilized REEs into fractures and alteration zones. This study is the first comparative study of barren and mineralized carbonatites suggesting the role of sulfates in REE transportation and/or deposition in carbonatitic systems.