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Origin of heavy rare earth elements in highly fractionated peraluminous granites

CHAOXI FAN AND CHENG XU

Peking University

Presenting Author: 13802926202@163.com

Rare earth elements (REEs+Y) play an important role in modern industry. Heavy REEs (HREEs) are particularly critical because of their relative scarcity in nature. Global HREE resources are predominantly present in granitoid weathering crusts in southern China. Although it is well known that REEs are generally enriched in alkaline rocks, in contrast, the parental granitoids in most Chinese HREE deposits are peraluminous. Here, we examined different REE mineral compositions, distribution patterns, and Nd isotope ratios of the granitoid complex in Zudong, South China, whose weathering crusts form the largest HREE deposits globally. The complex is composed of granodiorite, biotite-muscovite, and muscovite alkali-feldspar granites, whose REE patterns change from HREE depletion to enrichment relative to light REEs (LREEs) with increasing negative Eu anomalies. They are not fractional crystallization products from the same parental melts owing to the different zircon U-Pb ages . However, magmatic zircons from the three types of granitoids showed consistent initial Hf isotope compositions, indicating they evolved from the same sources. The granodiorite contains primary LREE minerals with low $\varepsilon Nd_{(t)}$ values , decoupled from high zircon Hf isotopes. This indicates a garnet-containing basement source resulted in Lu (HREE) remaining preferentially in the garnet phase, with Hf entering the melt. This contradicts the high HREE concentration recorded in the biotite-muscovite and muscovite granites, which contain abundant HREE minerals associated with muscovite, fluorite, and recrystallized quartz. The whole-rocks and their secondary REE minerals show variable and higher initial Nd isotope ratios than the early granodiorites and their primary REE minerals, indicating minimal ^{143}Nd was derived from radioactive decay of ^{147}Sm in the basement and wall rocks. This constrained the contribution of external REE-, volatile-rich liquids, which drove the segregation of highly evolved silicate melts from the magma chamber. This increased volatile saturation and fluid exsolution, which may have mobilized the HREEs and metasomatized the granites during their emplacement. Therefore, we infer that external fluid metasomatism appears to be crucial for the HREEenrichment in highly fractionated peraluminous granites.