１気圧・高温条件での不混和珪酸塩メルトースフェーンージルコン間の微量元素と希土類元素の分配実験

佐藤桂 [1, 2]、David J. Ellis[2]、Andrew G. Christy[3]
[1] *東京工業大学 理工学研究科 地球惑星科学専攻*
[2]*Research School of Earth Sciences, The Australian National University*
[3]*Centre for Advanced Microscopy, The Australian National University*

Experimental study of partitioning of trace elements and rare earth elements between immiscible silicate liquids–titanite–zircon at the atmospheric pressure

Kei Sato[1, 2], David J. Ellis[2] and Andrew G. Christy[3]
[1]*Department of Earth and Planetary Sciences, Graduate School of Science and Engineering, Tokyo Institute of Technology*
[2]*Research School of Earth Sciences, The Australian National University*
[3]*Centre for Advanced Microscopy, The Australian National University*

The immiscibility between silicate liquids has been discussed in much study since it was discovered by Greig (1927a; 1927b), because it is well-known fact that many substances may separate into the plural number of liquids upon cooling or heating (e.g. Barth, 1962). Since the 1950s, further discoveries of coexisting immiscible silicate liquids have been reported from natural occurrences of terrestrial volcanic rocks (e.g. Roedder and Coombs, 1967; De, 1974; Philpotts, 1982) and lunar magmas (e.g. Roeder and Weiblen, 1970; Snyder et al., 1993; Shearer et al., 2001), and from experimental studies (e.g. Roedder, 1951; Rutherford et al., 1974; Watson, 1976; Ellison and Hess, 1989; Vicenzi et al., 1994; Veksler et al., 2006). The distribution of elements between such immiscible liquids could be useful for an understanding of the evolution of lunar crust, and/or of the process of fluid inclusions in terrestrial rocks because such liquids might correlate with the transportation of elements from upper mantle to crust, and crystallization differentiation.

It is known that zircon and titanite (sphene) can acquire large amounts of HREEs and LREEs, respectively, as well as Th and U. Thus, the present study was aimed at demonstrating the distribution of trace elements and REEs between two immiscible silicate liquids coexisting with liquidus minerals (i.e. zircon and titanite) in the high zirconium quartzo-feldspathic system to which natural zircon and monazite were added. Th/U ratio in bulk composition of this starting mixture is about 8.4. The experiments were performed at the temperatures of 1500-1200°C, at one atmosphere experiments, by using an electric furnace housed at the Research School of Earth Sciences, The Australian National University. The starting mixture was ground under acetone in agate mortar and pressed into a tablet in die. This tablet was put into the Pt basket, hung in the electric furnace and heated at 1400°C under controlled low oxygen fugacity. The tablet was melted, and then quenched. This run product was then re-ground, and again melted at 1500°C under the same fO_2 condition. Both of the 1400°C and 1500°C materials were quenched to a single quartzo-feldspathic liquid (glass) together with a small amount of euhedral zircon. The 1500°C material was re-ground, and this powdered sample was used as starting material for other experiments at 1350°C, 1300°C, 1250°C and 1200°C under the same oxygen fugacity. All run products were mounted in epoxy resin and polished for examinations utilizing electron microprobe and LA-ICP-MS.

As an experimental result, a single silicate liquid was obtained coexisting with liquidus zircon at the temperatures of 1500-1350°C. The single liquid split into two immiscible silicate liquids (Si-rich liquid and Si-poor one) coexisting with liquidus zircon at 1300°C. The Si-rich liquid was obtained as groundmass liquid, and the immiscible Si-poor liquids were found as small size of droplets in this run product. Since titanite and more zircons crystallized at 1250°C, Si-poor liquid decreased sharply in amount. Such immiscible Si-poor liquid has not been found in run product at 1200°C. This suggests the closure of the liquid immiscible loop at this temperature.

The following thirty-nine elements were measured by the LA-ICP-MS study: ^{29}Si, ^{31}P, ^{39}K, ^{43}Ca, ^{45}Sc, ^{49}Ti, ^{51}V, ^{59}Co, ^{71}Ga, ^{72}Ge, ^{75}As, ^{85}Rb, ^{88}Sr, ^{89}Y, ^{91}Zr, ^{93}Nb, ^{95}Mo, ^{118}Sn, ^{133}Cs, ^{137}Ba, ^{139}La, ^{140}Ce, ^{141}Pr, ^{146}Nd, ^{147}Sm, ^{153}Eu, ^{157}Gd, ^{159}Tb, ^{163}Dy, ^{165}Ho, ^{166}Er, ^{169}Tm, ^{172}Yb, ^{175}Lu, ^{177}Hf, ^{181}Ta, ^{182}W, ^{232}Th and ^{238}U. The distribution of Al between the produced phases was determined by the electron microprobe. At 1300°C, K^{1+}, Rb^{1+}, Cs^{1+}, Al^{3+} and Si^{4+} were enriched in the groundmass Si-rich liquid, as compared to the second Si-poor liquid. At 1250°C, in addition to these elements, Ba^{2+}, Hf^{4+} and Th^{4+} were also enriched in the groundmass Si-rich liquid. Other elements were enriched in the Si-poor liquid at both of 1300°C and 1250°C. The droplets of Si-poor liquids obtained at 1300°C yielded the Th/U ratio of about 11.0. As had been mentioned above, Si-poor liquid decreased sharply in amount at 1250°C. As results, the U content in this residual Si-poor liquid decreased sharply, and thus Th/U ratio in the Si-poor liquid increased to 56.5. Such change of chemical characteristics in the Si-poor liquid may be correlated to the formations of monazite and/or allanite which often yield very high Th/U ratio and high LREEs concentrations.

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