

EXPERIMENTAL STUDY OF PHOSPHORUS AND CHLORINE INFLUENCE ON THE SILICATE-CARBONATE IMMISCIBILITY AND DISTRIBUTION OF SOME RARE AND RARE EARTH ELEMENTS

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The possible role of liquid immiscibility in the genesis of carbonatites and associated rocks was discussed by previous authors [1-3]. Geochemical data estimate presence of apatite, sulphides, phlogopite and other minerals in carbonatites. More over, in massif Tomtor, for an example, there were observed the apatite-carbonate eruptive dykes or tubes and tuff-lavas of phosphate composition [4]. This fact confirms the supplementary fluid components (such as phosphorus and, probably, halogenides) enrichment in latter carbonatites. So, the experimental study of the effect of phosphorus and chlorine on silicate-carbonate immiscibility and behaviour of ore elements in such immiscible systems is an actual problem.

The results of the experimental study of liquid immiscibility in carbonate-silicate systems at $T=1250^{\circ}\text{C}$ and $P=2$ kbar and in phosphate-carbonate-silicate and chloride-carbonate-silicate systems are presented in this paper. The main attention has been paid to study of distribution of some ore elements (REE, Nb, Ta) between immiscible phases.

The experiments were carried out in a high gas-pressure vessel in sealed platinum capsules ($d=3$ mm) during 6 h and than quenched. Starting mixtures were prepared from the mixtures of rock-forming minerals (albite, diopside) and the compositions of carbonate model melts were determined by Na_2CO_3 or mixtures of Na_2CO_3 and CaCO_3 . Phosphorus and chlorine in some special experiments were added as NaPO_3 and NaCl respectively. Ore elements were added as La_2O_3 , CeO_2 , Y_2O_3 , Nb_2O_5 , Ta_2O_5 . The samples obtained were analyzed on a Camebax microprobe equipment with a Link H.E.D.A. system.

A wide region of layering of the initial melts into two liquids - silicate and carbonate ones was revealed. The carbonate drops in silicate matrix and the silicate drops in carbonate matrix were observed in the samples obtained. There are distinct phase boundaries between both phases.

The study of distribution of REE (La, Ce, Y), Nb, Ta between immiscible silicate and carbonate liquids shows their concentration by silicate melt. The partition coefficients for REE between layering phases ($K=C_{\text{carb}}^{\text{REE}}/C_{\text{sil}}^{\text{REE}}$) are presented in Fig. They are less than 1. The results obtained and our previous data [5, 6] reveal the dependence of the coefficient of distribution of rare earth elements on the temperature: the increase of temperature to 1250°C results in the redistribution of REE in favor of the silicate phase.

The phosphorus influence on the silicate-carbonate layering and distribution of REE between immiscible phases has been good illustrated by the results of our experiments carried out at $T=1250^{\circ}\text{C}$ and $P=2$ kbar with the addition of NaPO_3 (8 and 15 mas.%, which correspond to 5.6 and 10.4 mas.% P_2O_5 respectively). In studied region of compositions the phosphorus addition does not effect on the width of immiscible field in silicate-carbonate systems. But some increase of composition contrast of coexisting phases is likely to be estimated. Salt phase in studied systems has phosphate-carbonate composition. The increase of coefficient of distribution of REE between immiscible phases is found (fig. 1), and in alkaline systems they predominantly accumulate in salt melt ($K>1$). With increase of phosphorus content in initial melts the salt phase obtained was established to be inhomogeneous and contains the constituents of carbonate and phosphate compositions. The phosphate phase is more effective in concentration of REE than carbonate one, and enriched in these elements. So, our experiments show the positive effect of phosphorus addition on the extraction of rare earth elements by salt melts in studied systems.

Chlorine influence on the silicate-carbonate layering has been studied at $T=1100^{\circ}\text{C}$ and $P=2$ kbar with the addition of chloride (NaCl) to the system. In chloride-carbonate-silicate systems the separation of chloride-carbonate melts from the silicate ones is observed. Salt phase occurred as a layer in upper part of sample or as drops in silicate melt. Salt liquid is inhomogeneous phase and divided into chloride (NaCl with addition of Ca and Mg). Salt phase is depleted in rare earth elements, Nb and Ta, which concentrate in silicate melt. But it is observed that carbonate phase is richer in REE as compared with chloride phase. So, the presence of chlorine does not effect positively on the extraction of ore elements by salt melts. On the contrary, in alkaline systems at $T=1100^{\circ}\text{C}$ addition of chlorine promote the decrease of REE concentration in salt liquids, and REE accumulate in silicate melts. But in pure silicate-carbonate systems according to our previous investigations REE concentrate in carbonate liquids.

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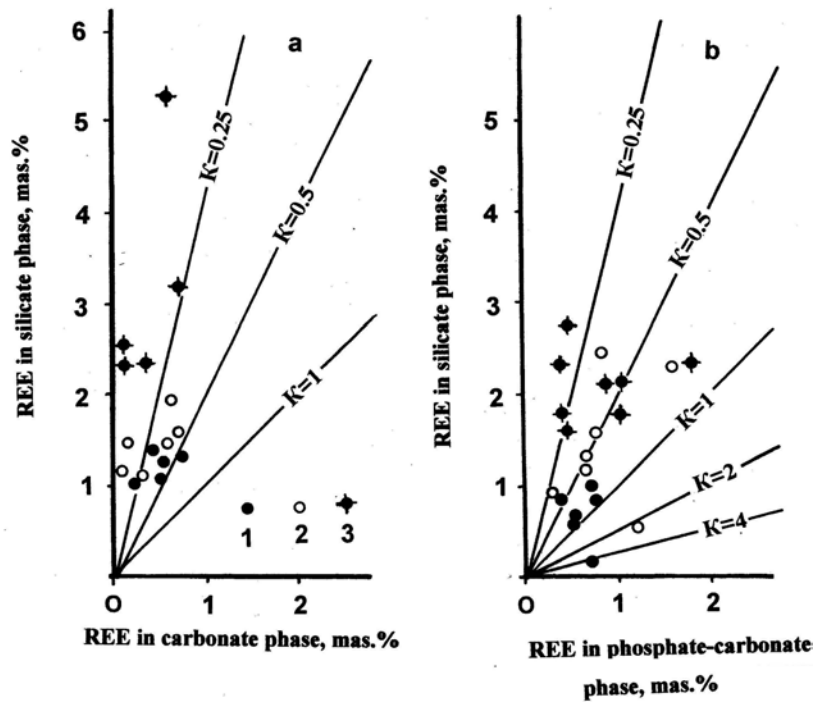


Fig. 1. Coefficients of distribution of rare earth elements between silicate and carbonate melts (a) and between silicate and salt (phosphate-carbonate) melts at $T=1250^\circ\text{C}$ and $P=2$ kbar. 1 - La_2O_3 ; 2 - Ce_2O_3 ; 3 - Y_2O_3 .