The formation and differentiation of magmas

Kotelnikov A.R., Suk N.I., Korneeva A.A. **Distribution of Nb and Ta in the system of tantalum niobate – aluminosilicate melt.** *UDC 550.8.014*

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Abstract. Experiments on Nb and Ta distribution were carried out at 800°C and P = 3.5 kbar in a high gas pressure vessel. Specially synthesized Na-Si and Li-F glasses were used for loading. The fluid (NaF + KF + H_2O) was present in the supersaturation mode. Tantalum-niobates of manganese and iron were also synthetic, their composition corresponded to the formula: $(Fe_{0.5}Mn_{0.5})NbTaO_6$. Experiments showed that melts of Na-Si specificity are enriched in Nb, and Li-F specificity – in Ta. The obtained data explain the differentiation of rare elements depending on the compositions of residual melts enriched in incompatible elements.

***Keywords**: synthesis, feldspars, gallium, germanium, solid solutions*

In the works (Linnen R.L., 2005; Thomas R., 2006), it is shown that residual melts by their geochemical specificity can be both aluminous with enrichment in Li- and F-components (lithium-fluoride granites), and with Na-Si specificity (highly alkaline silicate). Based on the study of natural objects, R.L. Linnen (Linnen R.L., 2005) showed that lithium-fluoride granites are enriched in such elements as Be, Ta, Hf, Y, HREE, while sodium-silicate alkaline varieties are enriched in Nb, Zr, LREE. Below the data on the distribution of a number of elements (Nb, Ta, Zr, Hf, Mo, W) between these rocks are given (Fig. 1a, b, c).

It is shown that Nb and Ta behave quite contrastingly in the processes of differentiation of residual melts during crystallization of granite systems. Tantalum enriches plumasite melts enriched in lithium and fluorine, and niobium is redistributed into residual melts enriched in sodium and silica. Therefore, the aim of our work was to experimentally verify this observation.

Experimental method

Starting materials. Glasses of the following compositions were synthesized for the experiments (Table 1). The starting mixtures were stirred under a layer of alcohol, dried at 90°C and placed in a platinum crucible. Melting was carried out at 1100°C for 3 hours. The resulting glasses were analyzed by the microprobe method for homogeneity and compliance of the compositions with the initially specified ones. Lithium in the glasses was determined by the atomic absorption analysis method. Tantalum-niobates of Mn and Fe with the formula $(Fe_{0.5}Mn_{0.5})NbTaO_6$ was synthesized from mixtures of oxides, at 1250°C during 8 hours. To prevent iron oxidation, the "sand seal" method was used, using an oxygen buffer C-CO-CO_2. Analysis of the composition of synthetic tantalum-niobates (further – tnt) showed its compliance with the specified one.

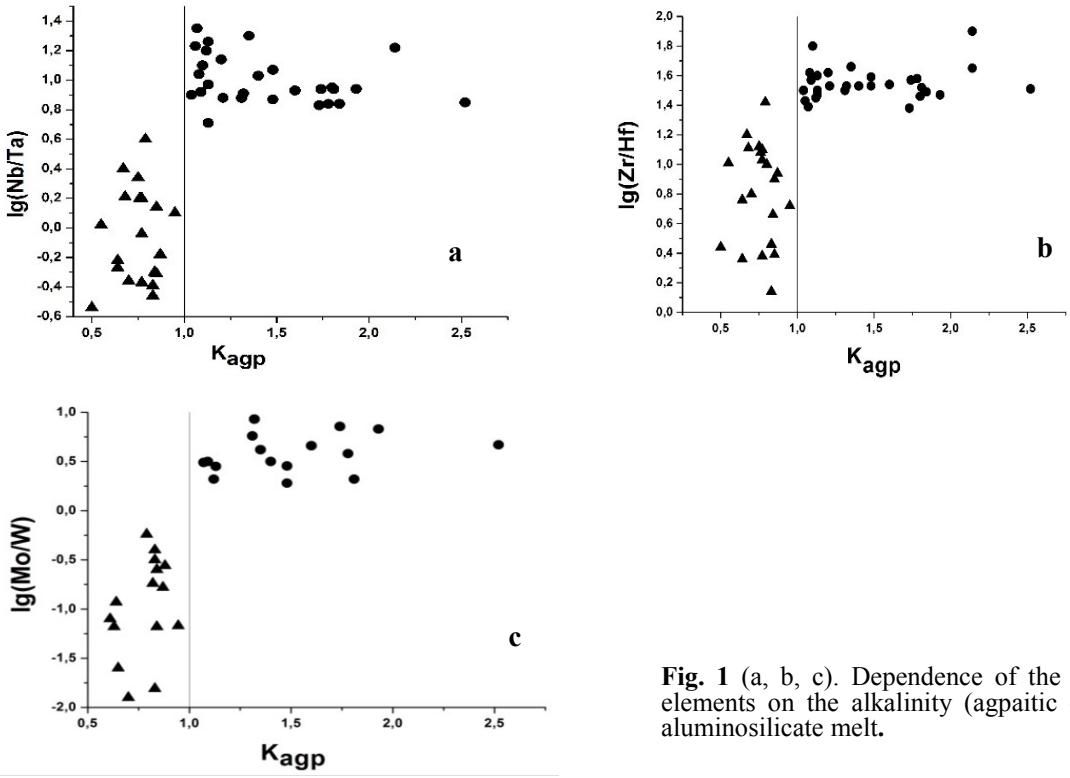

Fig. 1 (a, b, c). Dependence of the ratios of indicator elements on the alkalinity (agpaitic coefficient) of the aluminosilicate melt.

Table 1. Compositions of the original glasses (wt.%)

Specificity	Li_2O	Na_2O	K_2O	Al_2O_3	SiO_2	AlF_3	NaF	∑
Li-F	3.11	3.69	2.80	15.16	71.49	2.50	1.23	99.98
Na-Si	0.00	10.51	9.88	6.93	70.81	0.00	0.00	98.13

Equipment. The experiments were carried out on high gas pressure vessel (designed by IEM RAS). The accuracy of temperature regulation and control was ±2°C, pressure ±50 bar.

Experimental procedure. Gold ampoules with diameters of 4 and 7 mm were used. The initial sample (a mixture of glass and tnt in a ratio of 6:1) was loaded into the ampoule. 10-15 wt% of a 1M NaF + 1M KF solution was added to the ampoule, after which the ampoule was welded.

The duration of the experiments at 800℃ and P = 3.5 kbar was 14 days. After the experiments, the ampoule was weighed, opened and a microprobe analysis of the experimental products was performed.

Experimental results

The distribution of Nb and Ta between tantalum-niobate (tnt) and melt (liq) is described by the equation:

$(Fe,Mn)Nb_2O_6(tnt) + 2Ta(liq) = (Fe,Mn)Ta_2O_6(tnt) + 2Nb(liq)$.

To describe the distribution, it is convenient to use the mole fractions of tantalum and niobium $X_{Ta}^{liq(tnt)}$ and $X_{Nb}^{liq(tnt)}$, equal to the ratios: $X_{Ta}^{liq(tnt)}$ = Ta/(Nb+Ta) and $X_{Nb}^{liq(tnt)}$= Nb/(Nb+Ta).

The partition coefficients are written as follows: $K_D(Nb)=X_{Nb}^{liq}/ X_{Nb}^{tnt}$ and $K_D(Ta)=X_{Ta}^{liq}/ X_{Ta}^{tnt}$. The values of the partition coefficients are given in Table 2.

Table 2. Values of the partition coefficients of tantalum and niobium between the melt and $(Fe_{0.5}Mn_{0.5})NbTaO_6$ depending on the agpaitic coefficients: Kagp=(Na+K)/Al.

№ exp.	K_{agp}	$K_D(Nb)$	$K_D(Ta)$	Nb/Ta	Melt
7618	1.430	1.178	0.862	0.756	LiF-gr-gl
7588	2.000	1.890	0.670	1.000	NaSi-gl
7617	4.230	1.970	0.452	1.987	NaSi-gl
7590	5.300	2.050	0.548	1.571	NaSi-gl
7589	5.900	2.650	0.541	1.263	NaSi-gl

The dependences of the partition coefficients of Nb and Ta on the agpaitic coefficient are described by the following equations:

$K_D(Nb) = 1.092 + 0.227\times K_{agp}$; $S_x=0.24$
$K_D(Ta) = 0.855 – 0.64\times K_{agp}$; $S_x=0.14$.

Thus, it is shown that with increasing alkalinity the niobium content in the melt also increases, while for tantalum the inverse relationship is observed. Our experimental data quite well substantiate the empirical data based on the geochemistry of natural complexes (Linnen, 2005).

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References

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