EXPERIMENTAL STUDY OF CRYSTAL COLUMBITE SOLUBILITY IN GRANITIC MELTS OF PERALUMINOUS (A/NK ~1.7), NORMAL (A/NK ~1.1) AND PERALKALINE (A/NK ~0.64) COMPOSITIONS AT T = 650-850°C AND P = 0.3-4 KBAR: THE FIRST RESULTS

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The gel mixtures of Na_2O-K_2O-Al_2O_3-SiO_2 composition with varying values of mole Al_2O_3 / (Na_2O+K_2O) ratio (for short A/NK) from ~0.64 up to 1.7 (0.64, 1.1 and 1.7) were prepared for realization of experiments. The starting water-saturated Li- and F-enriched homogeneous glasses were obtained by two-times melting of these gel mixtures. Initially these mixtures were melted at 1 atm in air using slow stepwise heating up to 1500°C, and then the glasses were crushed and they were melted again after addition of FeO, MnO, LiF and 0.2 N HF solution at P_{H2O} =1 (0.3, 4) kbar, T =900°C in the presence of Ni-NiO buffer. The crystals of natural columbite $(Mn,Fe)(Nb,Ta)_2O_6$ were used in our experiments. The columbite crystal was placed inside glass powder and 0.2 N HF solution (2-4 wt. %) was added thereto. Experiments were carried out using gold capsules in internally heated pressure vessels. Columbite was dissolved by diffusion in aluminosilicate melt during the experiments.

After the runs Ta, Nb, Fe, and Mn contents in aluminosilicate quenched glasses along perpendicular profiles to columbite crystal boundary were measured by electron microprobe analyses using (1) energy-dispersive x-ray ***(EDX)*** microprobe analyzer with solid-state Si-(Li) detector ***INCA Energy*** to electron microscope CamScan MV-2300 (in IEM RAS) and (2) crystal-diffraction (wave dispersion) spectrometers of microprobe analyzer Cameca MS-46 (in IGEM RAS). The obtained diffusion profiles were approximated using exponential equations. The maximal concentrations of Ta, Nb, Fe and Mn in quenched glass on columbite / glass boundary are calculated. They correspond to the solubilities of these metals in the melt. The dependence of columbite solubility from three major factors: melt composition, temperature and pressure, is investigated.

The melt composition has the greatest effect on the columbite solubility (Fig. 1). The solubilities of Ta and Nb are maximal in alkaline melt (A/NK ~0.64), they decrease almost in order of magnitude in the melt of normal composition (A/NK ~1.1) and they continue to decrease appreciably with increase of alumina content in the melt composition (A/NK ~1.7).

The temperature has weaker effect on the columbite solubility in comparison with effect of the melt composition. This effect is appreciable only for normal melt composition with A/NK ~1.1 (Fig. 1). For this melt composition at P =1 kbar the Ta content in the glass decreases ~3.5 times, and the Nb content in the glass decreases down to ~10-15 times with reduction of temperature from 850 up to 650°C. At P =1 kbar the Ta content is higher than Nb content (Nb/Ta ~0.5) for alumina-rich melt composition (A/NK ~1.7) at the all studied range of temperature. For alkaline-rich melt composition the Nb/Ta ratio is opposite (Nb/Ta ~1.25-2.0), and for normal melt composition with A/NK ~1.1 the tantalum dominate at 650°C, and the niobium dominate at 750-850°C. As a whole the Nb solubility changes with temperature more strongly in comparison with the Ta solubility.

The pressure decrease from 4 up to 0.3 kbar has also weaker effect on the columbite solubility in comparison with effect of the melt composition. For alkaline-rich melt composition the Nb content and, probably, the Ta content increase as pressure decrease. It is possible at P =4 kbar and for melt composition with A/NK ~1.1 the Ta content in the glass starts to dominate above the Nb content at higher temperature (750°C). The first obtained results with respect to the form change of diffusion profiles show the essentially higher diffusion rates of Nb and Ta at P = 4 kbar in comparison with P = 1 kbar and, accordingly, the reduction of diffusion rate at P =0.3 kbar. That is at the increased pressure the mobilities of Nb and Ta in the melt are appreciably elevated. This growth is primarily connected to the reduction of melt viscosity with increase of pressure.

Conclusions:

1). The composition variation of granitic melt has the greatest effect on columbite solubility in comparison with influence of temperature and pressure. The solubilities of Ta and Nb are maximal in alkaline melt, they decrease almost in order of magnitude in the melt of normal composition and they continue to decrease as alumina content increase.

2). The solubility of Nb in granitic melt changes with temperature more appreciably as compared with the solubility of Ta, that is the Nb/Ta ratio in the melt decreases usually with reduction of

temperature. The positive temperature dependence of columbite solubility is more strongly expressed for granitic melt with A/NK ~1.1 in comparison with alkaline-rich and alumina-rich melt compositions.

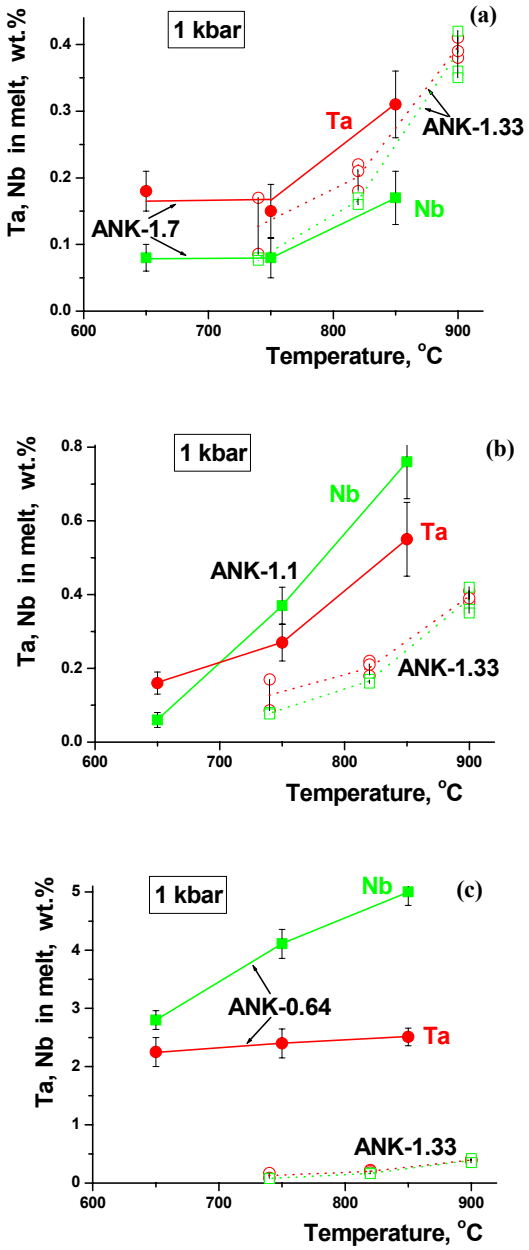

Fig.1. Effect of melt composition (A/NK ~1.7 (a), 1.33, 1.1 (b) and 0.64 (c)) on solubilities of Ta and Nb in granitic melt at columbite dissolution. Our prior results for the melt of natural Li-F granite (A/NK ~1.33) are added for comparison as fixed mark.

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