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THE ASSOCIATION OF COLUMBITE, TANTALITE AND TAPIOLITE IN THE SUZHOU GRANITE, CHINA

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Abstract

We present the results of an investigation of the chemical composition of Nb–Ta oxide minerals from the Suzhou granitic complex, China. This complex is composed of three map-units, granite II being the most important. In the upper part of granite II, three well-evolved facies are distinguished; they show signs of disseminated mineralization in rare metals (Nb, Ta, Zr, Hf, *REE*, Th). From the top down, these are (1) the albite-dominant facies, (2) the topaz-bearing facies, and (3) the biotite-bearing facies. Optical microscopy studies and electron-microprobe analyses have shown that ferrocolumbite occurs in the biotite-bearing facies, and ferrocolumbite to manganocolumbite is found in the topaz-bearing facies. Ferrotantalite and tapiolite are concentrated in the albite-dominant facies, these two latter minerals being identified for the first time in the Suzhou granite. The tapiolite has an unusual composition, beyond the known field of stability of tapiolite, suggestive of disequilibrium crystallization. The chemical evolution of the Nb–Ta minerals from the biotite-bearing granite to the albite-dominant facies reveals an important fractionation of Nb and Ta, accompanied by a more moderate evolution of the Fe:Mn ratio. Tapiolite in the albite-dominant granite indicates an increase in the activity of Fe at the end stage of differentiation of the residual magma. By comparison with the Beauvoir granite, in France, it is likely that this pattern of Fe and Mn fractionation is due to the reduced activity of fluorine.

Keywords: columbite, tantalite, tapiolite, electron-microprobe data, granite, Suzhou, China.

Sommaire

Cette étude porte sur la composition chimique des minéraux de Nb et Ta du granite de Suzhou (Chine). Ce complexe est composé de trois unités granitiques, parmi lesquelles le granite II est le plus important; on y distingue trois faciès évolués minéralisés en métaux rares (Nb, Ta, Zr, Hf, terres rares, Th) qui sont définis, du haut en bas, comme: faciès à albite dominant (1), faciès à topaze (2), et faciès à biotite (3). Les études microscopiques et les analyses à la microsonde électronique ont permis d'identifier la ferrocolumbite dans le faciès à biotite, et la séquence ferrocolumbite à manganocolumbite dans celui à topaze, tandis que ferrotantalite et tapiolite se concentrent dans le faciès à albite, ces deux derniers minéraux étant signalés pour la première fois dans le granite de Suzhou. Dans un échantillon, la tapiolite présente une composition inhabituelle, en dehors du domaine stable connu, ce qui indique des conditions de déséquilibre lors de sa cristallisation. L'évolution chimique des niobo-tantalates, du granite à biotite au granite à albite, met en évidence un important fractionnement de Nb et Ta, accompagné de celui, plus modéré mais net, de Fe et Mn. La présence de tapiolite dans le granite à albite marquerait une augmentation de l'activité de Fe à la fin de la différenciation du magma résiduel. Par comparaison avec le granite de Beauvoir (France), ce comportement du Fe et du Mn dans le faciès à albite est probablement lié à sa faible concentration en fluor.

Mots-clés: columbite, tantalite, tapiolite, données de microsonde électronique, granite, Suzhou, Chine

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Introduction

Over the last twenty years or so, the Nb–Ta minerals of granitic pegmatites have been the focus of systematic studies (*cf.* Černý & Ercit 1985, 1989). The investigations have addressed, on one hand, compositional and structural variations of these minerals, and on the other, their evolution during the crystallization history of the pegmatites (Černý & Ercit 1989, Spilde & Shearer 1992, Ercit 1994). However, similar studies on Nb–Ta minerals in granites are scarcer. Granitic suites mineralized in Nb–Ta have been divided in two subtypes, one richer in phosphorus than the other (Taylor 1992). The topaz-bearing sodic granite at Beauvoir, in France, corresponds to the P-rich subtype, and represents one of the rare examples of rare-element-enriched granites that has been well studied from the point of view of mineralogy of the Nb–Ta phases columbite – tantalite, pyrochlore – microlite and ixiolite (Kosakevitch 1976, Ohnenstetter & Piantone 1988, 1992, Wang *et al.* 1987, Wang 1988). The Suzhou granite, in China, also mineralized in Nb–Ta, differs mineralogically and geochemically from the Beauvoir example. Among the minerals of Nb–Ta, we have established the presence of columbite, tantalite and tapiolite, the latter never having been noted as a constituent of a granite, to our knowledge. The associated accessory minerals are: hafnian zircon (Wang *et al.* 1996), thorite, xenotime-(Y) and monazite-(Ce). All these minerals have been characterized by optical microscopy, electron-microprobe analysis, and X-ray diffraction, the latter with the Debye–Scherrer method owing to the small size of the grains. This investigation has allowed us to characterize the compositional peculiarities and geochemical evolution of the Nb–Ta minerals. In light of these findings, it is possible to compare the Suzhou granite with the Beauvoir granite and granitic pegmatites.

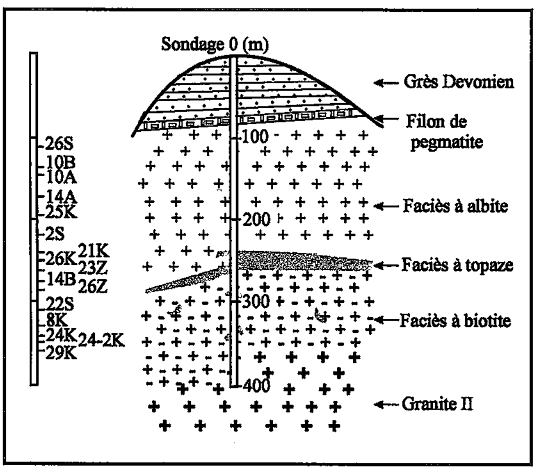

FIG. 1. Cross-section of Granite II at Suzhou, made as a result of observations made on surface exposures and from the drill core. To the left is the position of samples used in this project.

The Suzhou Granite

The principal characteristics of the Suzhou granite, as well as its magmatic origin, have been discussed in an earlier publication (Wang *et al.* 1996). We give here a cross-section of the granitic pluton and the location of the drill hole (Fig. 1). We recall that three units can be recognized in this granite body, but that Granite II is the only one to be mineralized (Nb, Ta, Zr, Hf, Y, the rare earths). Granite II itself can be subdivided into three facies, albite-dominant at the top, with topaz-bearing and biotite-bearing facies beneath it (Wang *et al.* 1996).

Characterization of the Nb–Ta Minerals

Analytical method

The conditions of chemical analysis and the correction procedures are identical to those described by Wang *et al.* (1996). The following standards were employed: $LiNbO_3$ (Nb), SnO_2 (Sn), titanite (Ti), $LiTaO_3$ (Ta), W metal (W), Fe_2O_3 (Fe), and graftonite (Mn). Structural formulae have been calculated on

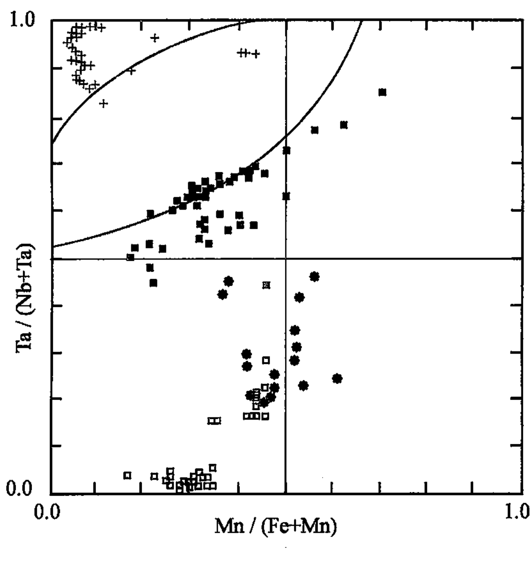

FIG. 2. Composition of the columbite–tantalite in the biotite-rich facies (□), the topaz-rich facies (•), and the albite-dominant facies (■), as well as of the tapiolite in the albite-dominant facies (+), expressed in terms of the Mn/(Fe + Mn) *versus* Ta/(Nb + Ta) quadrilateral.

the basis of six atoms of oxygen, in the case of both columbite–tantalite and tapiolite. In the text and figures, we utilize the symbol $\mathbf{R_{Ta}}$ to represent the ratio Ta/(Ta + Nb), and the symbol $\mathbf{R_{Mn}}$ to represent the ratio Mn/(Mn + Fe).

Columbite

Columbite is found mostly in the biotite- and topaz-bearing facies. In the first, the crystals are euhedral, of variable dimensions (5 to 10 μm by 5 to 8 μm), and included in biotite, in which it has caused pleochroic haloes to form. In the topaz-bearing facies, it forms intergranular crystals that are somewhat coarser, 10–30 μm by 5–10 μm. In both facies, the grains are zoned and mostly associated with zircon.

We have analyzed many crystals; compositions are presented in Table 1 and plotted in the columbite–tantalite quadrilateral (Fig. 2). The biotite-bearing facies contains ferrocolumbite having an $\mathbf{R_{Ta}}$ approximately equal to 0.13, which corresponds to the lowest concentrations of Ta in columbite-group minerals in the Suzhou granite. In contrast, in the topaz-bearing facies, $\mathbf{R_{Ta}}$ and $\mathbf{R_{Mn}}$ are higher, with compositions in the manganocolumbite field in three cases.

The crystals of columbite (*sensu lato*) in the biotite-bearing facies are zoned, with a relatively large central part and a narrow periphery (<5 μm). Images of the distribution of Nb and Ta in such a zoned crystal (Fig. 3) reveal a core relatively Nb-enriched compared to the rim. Five crystals of columbite, each showing the same pattern of zonation, were selected for detailed study. In each case, the crystals are significantly enriched in Ta near the rim, and show a more moderate, yet clear depletion there in Mn (Table 2, Fig. 4). Furthermore, the edge of the grains is richer in tungsten than the central part. In the normal pattern of zoning in columbite-group minerals, according to Lahti (1987), one expects $\mathbf{R_{Ta}}$ and $\mathbf{R_{Mn}}$ both to increase rimward. In the case of the Suzhou granite, $\mathbf{R_{Ta}}$ conforms to the expected pattern, but $\mathbf{R_{Mn}}$ systematically shows the opposite trend.

Tantalite

The presence of tantalite is documented for the first time at Suzhou, mostly in the albite-dominant facies. The crystals are tabular, 15–50 μm by 5–10 μm, and in

TABLE 1. REPRESENTATIVE CHEMICAL COMPOSITION OF COLUMBITE–TANTALITE FROM THE BIOTITE-RICH AND TOPAZ-RICH FACIES OF THE SUZHOU GRANITE

	1 14B3 -5	2 26Z4 -3	3 22S5 -1	4 8K10 -1	5 24K4 -2	6 21K2 -2	7 21K3 -1	8 23Z5 -2	9 23Z2 -5
Nb_2O_5 wt.%	47.02	58.43	70.40	73.02	75.35	49.33	47.52	48.51	40.74
Ta_2O_5	32.12	17.93	4.87	4.52	3.09	26.28	27.67	26.58	36.14
FeO	9.88	12.63	17.42	13.93	15.21	8.11	9.16	7.16	8.40
MnO	8.08	6.65	3.47	6.75	5.81	9.26	8.24	10.88	9.02
SnO_2	0.14	0.08	0.14	0.06	0.00	0.07	0.53	0.19	0.25
TiO_2	0.78	1.71	1.55	0.15	0.38	2.99	2.82	2.93	2.82
WO_3	0.93	1.95	1.61	0.88	0.66	4.25	3.94	2.72	1.62
Total	98.95	99.38	99.46	99.31	100.50	100.29	99.88	98.97	98.99
Nb^{5+}	1.385	1.608	1.835	1.906	1.928	1.392	1.358	1.388	1.214
Ta^{5+}	0.569	0.297	0.076	0.071	0.048	0.446	0.476	0.458	0.648
Fe^{2+}	0.539	0.643	0.840	0.673	0.720	0.423	0.484	0.379	0.464
Mn^{2+}	0.446	0.343	0.170	0.330	0.279	0.490	0.441	0.584	0.504
Sn^{4+}	0.004	0.002	0.003	0.001	0.000	0.002	0.013	0.005	0.007
Ti^{4+}	0.038	0.078	0.067	0.007	0.016	0.140	0.134	0.139	0.140
W^{6+}	0.016	0.031	0.024	0.013	0.010	0.069	0.065	0.045	0.028
Total	2.996	3.002	3.015	3.001	3.001	2.962	2.971	2.998	3.005
R_{Ta}	0.29	0.16	0.04	0.04	0.02	0.24	0.26	0.25	0.35
R_{Mn}	0.45	0.35	0.17	0.33	0.28	0.54	0.48	0.61	0.52

Columns: 1–5: biotite-bearing facies, 6–9: topaz-bearing facies. Compositions are expressed in wt.% oxides.

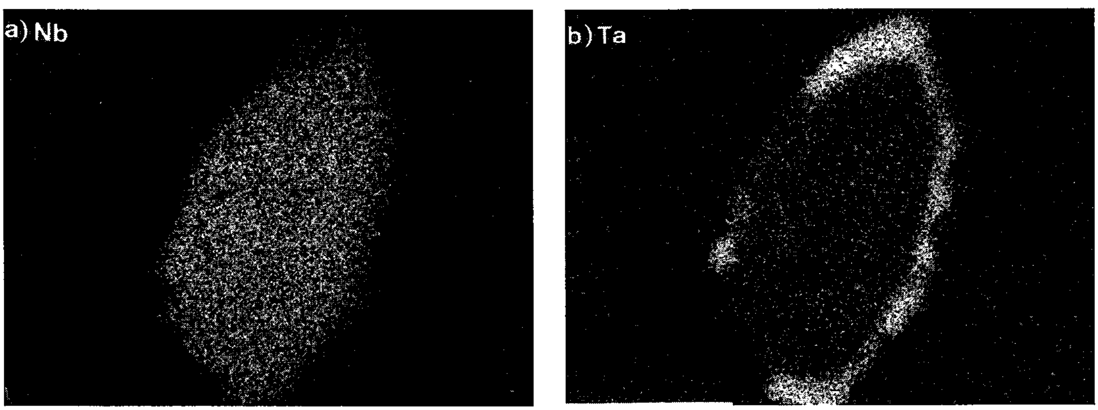

FIG. 3. Images showing the distribution of Nb (*Lα* radiation, a) and Ta (*Mα* radiation, b) in a zoned crystal of columbite, and revealing a pattern of Ta enrichment toward the margin. The width of the crystal is roughly 20 μm.

an intergranular position with respect to the main rock-forming minerals. On the basis of the $\mathbf{R_{Mn}}$ values (Table 3, Fig. 2), the nine crystals examined define a range of compositions from ferrotantalite ($0.2 < \mathbf{R_{Mn}} < 0.3$), to an intermediate member of the series ($\mathbf{R_{Mn}} = 0.5$), and to manganotantalite ($0.5 < \mathbf{R_{Mn}} < 0.7$). Intracrystalline variations are negligible; in contrast, the tantalite grains associated with tapiolite are strikingly heterogeneous.

Tapiolite

In the granitic pegmatites mineralized in Ta, the presence of tapiolite is generally linked to a low activity of fluorine and a limited degree of evolution in terms of Fe/Mn (Černý & Ercit 1989). The presence of tapiolite in a rock having a granitic texture does not seem to have been noted before. In a recent study, Wise & Černý (1996) mentioned the occurrence of tapiolite in granitic host-rocks of pegmatitic texture only. In the Suzhou granite, the tapiolite is well represented, but is restricted to the albite-dominant facies, and especially to its apical parts, where it is present in subhedral grains, 30–50 μm by 15–30 μm. In polished sections, tapiolite can be recognized by its good polish, light gray color, distinct pleochroism, low reflectivity, strong anisotropy, and intense brownish red internal reflections. It usually is simply twinned, but in some cases where it is polysynthetically twinned, the twin lamellae are relatively fewer and narrower than in the case described in the Anti-Atlas, in Morocco (Permingeat 1955). One can see, in a single crystal, from two to four orientations of twin lamellae (Fig. 5). The tapiolite is associated with grains of tantalite, hafnian zircon, and thorite. The $\mathbf{R_{Ta}}$ value varies from 0.87 to 0.97, and $\mathbf{R_{Mn}}$, from 0.05 to 0.42 (Table 4); these values correspond to the field of stability of tapiolite (Fig. 2). However, one grain of tapiolite (10A1) shows an unusual composition, beyond this field, possibly an indication of particularly rapid crystallization.

TABLE 2. REPRESENTATIVE CHEMICAL COMPOSITION OF ZONED COLUMBITE FROM THE BIOTITE-RICH FACIES OF THE SUZHOU GRANITE

	8K6		24–2K1		24–2K2		24–2K3		24–2K4	
	C*	R*	C	R	C	R	C	R	C	R
n	1	1	2	4	1	1	4	1	1	1
Nb_2O_5	76.47	42.90	74.77	43.15	72.05	41.07	75.90	43.65	74.02	42.27
Ta_2O_5	2.04	36.25	3.74	36.61	5.72	37.80	2.63	35.68	4.53	36.39
FeO	14.67	13.51	14.81	13.75	14.11	13.13	14.19	13.59	14.93	13.40
MnO	6.70	3.98	5.95	3.80	6.22	4.17	6.74	3.69	5.96	4.07
SnO_2	0.00	0.01	–	–	0.01	0.10	0.01	0.00	–	–
TiO_2	0.10	0.43	0.33	0.40	0.70	0.17	0.27	0.75	0.34	0.31
WO_3	0.18	2.84	0.50	1.97	0.88	2.88	0.48	2.09	0.66	3.00
Total	100.16	99.92	100.10	99.68	99.68	99.32	100.22	99.45	100.43	99.44
Nb^{5+}	1.956	1.284	1.925	1.293	1.877	1.250	1.943	1.303	1.905	1.275
Ta^{5+}	0.031	0.653	0.058	0.660	0.090	0.692	0.041	0.641	0.072	0.660
Fe^{2+}	0.694	0.748	0.706	0.763	0.680	0.740	0.672	0.751	0.712	0.748
Mn^{2+}	0.321	0.223	0.287	0.213	0.304	0.238	0.323	0.206	0.287	0.230
Sn^{4+}	0.000	0.000	–	–	0.000	0.003	0.000	0.000	–	–
Ti^{4+}	0.004	0.021	0.014	0.020	0.030	0.008	0.011	0.037	0.015	0.015
W^{6+}	0.003	0.049	0.007	0.034	0.013	0.050	0.007	0.036	0.010	0.052
Total	3.010	2.978	2.997	2.983	2.994	2.981	2.999	2.974	3.001	2.980
R_{Ta}	0.02	0.34	0.03	0.34	0.05	0.36	0.02	0.33	0.04	0.34
R_{Mn}	0.32	0.23	0.29	0.22	0.31	0.24	0.32	0.22	0.29	0.24

* C: core, R: rim; *n*: number of analyses; – not analyzed. Compositions are expressed in wt.% oxides.

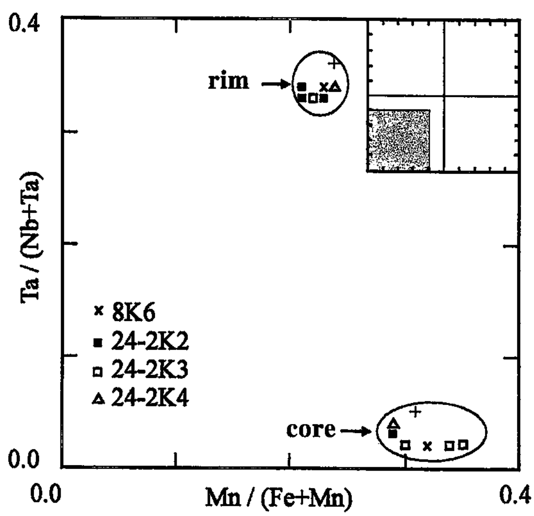

FIG. 4. Compositions recorded in zoned columbite, expressed in terms of the Mn/(Fe + Mn) *versus* Ta/(Nb + Ta) quadrilateral.

TABLE 3. REPRESENTATIVE CHEMICAL COMPOSITIONS OF TANTALITE FROM THE ALBITE-DOMINANT FACIES OF THE SUZHOU GRANITE

	26S -3	14A2 -2	25K4 -1	2S2 -1	26K2 -5	10A1 R(1)	10A1 P(6)	10A3 R(2)	10A3 P(8)
Nb_2O_5 wt.%	19.05	27.97	27.44	20.26	22.18	7.25	19.45	12.51	19.13
Ta_2O_5	61.17	53.10	51.86	60.67	56.62	72.47	60.75	67.18	60.76
FeO	10.17	13.67	12.73	7.50	11.32	4.09	10.43	7.09	9.99
MnO	5.08	2.94	3.94	7.31	3.91	9.51	4.99	7.89	5.05
SnO_2	0.11	0.02	0.11	0.08	0.16	0.71	0.12	0.36	0.16
TiO_2	2.44	1.13	0.88	1.70	1.80	4.67	2.22	3.32	2.36
WO_3	1.09	0.94	2.71	0.89	4.95	0.97	1.08	0.60	1.32
Total	99.11	99.77	99.67	98.41	100.94	99.67	99.04	98.95	98.77
Nb^{5+}	0.642	0.902	0.888	0.688	0.825	0.254	0.655	0.434	0.646
Ta^{5+}	1.239	1.030	1.010	1.239	1.113	1.530	1.231	1.402	1.235
Fe^{2+}	0.634	0.816	0.763	0.471	0.684	0.266	0.650	0.455	0.625
Mn^{2+}	0.321	0.178	0.239	0.465	0.239	0.626	0.315	0.513	0.320
Sn^{4+}	0.003	0.001	0.003	0.002	0.005	0.022	0.004	0.011	0.005
Ti^{4+}	0.137	0.061	0.047	0.096	0.098	0.273	0.124	0.192	0.133
W^{6+}	0.021	0.017	0.050	0.017	0.093	0.020	0.021	0.012	0.026
Total	2.997	3.005	3.000	2.978	2.957	2.990	3.001	3.019	2.989
R_{Ta}	0.66	0.53	0.53	0.64	0.61	0.86	0.65	0.76	0.66
R_{Mn}	0.34	0.18	0.24	0.50	0.26	0.70	0.33	0.53	0.34

10A1, 10A3: the grains of tantalite are heterogeneous; R: portion rich in Mn, P: portion poor in Mn. The number of analyses is shown in parentheses. The compositions are expressed in wt.% oxides.

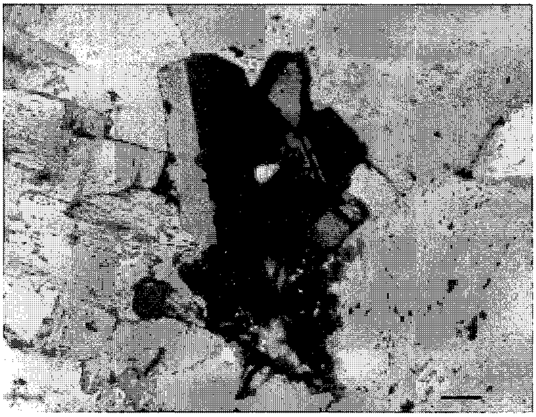

FIG. 5. Twinned crystal of tapiolite (scale bar: 150 μm).

Tapiolite–tantalite association

The association tapiolite–tantalite is developed in the apical part of the albite-dominant facies. Here, tapiolite grains are distinguished from tantalite by a strong anisotropy and by the presence of twin lamellae. Furthermore, it is usually located at the margin of tantalite grains. Images of the distribution of elements show that these minerals are heterogeneous, in particular in the distribution of Mn. Zones characterized by different levels of Mn have smooth boundaries in both minerals.

Quantitative analyses were done on associated grains of tapiolite and tantalite (Tables 3, 4). The value of $\mathbf{R_{Mn}}$ ranges from 0.05 to 0.42 in tapiolite, and from 0.18 to 0.70 in tantalite. Images showing the distribution of elements reveal the coexistence of tapiolite and ferrotantalite (in some cases, manganotantalite). It is important to emphasize that the Mn-rich part of the tapiolite grains has an $\mathbf{R_{Ta}}$ of 0.94 and an $\mathbf{R_{Mn}}$ of 0.42, coordinates that lie outside its presumed field of stability (Fig. 2). The only example of a similar composition, in fact part of a tapiolite crystal, was documented by Lahti *et al.* (1983). In the opinion of Černý *et al.* (1992), such a composition of tapiolite probably is metastable. Chemical re-equilibration could lead to an intergrowth of exsolution-induced domains of ferrotantalite and ferrotapiolite, as noted in the Nyanga No. 2 granitic pegmatite, Uganda (Černý *et al.* 1989). In the case of the Suzhou granite, a wide variety of compositions has been found, either between different crystals or between zones of a single crystal of tapiolite or tantalite, which must reflect disequilibrium crystallization (London 1991). Furthermore, we have not been able to find exsolution-induced domains in tapiolite, such that the extent of compositional re-equilibration must have been rather limited.

TABLE 4. CHEMICAL COMPOSITION OF TAPIOLITE IN THE ALBITE-DOMINANT FACIES OF THE SUZHOU GRANITE

	26S5 -1	26S3 -1	10B5 -1	10B7 -2	10A1 R(3)	10A1 P(3)	10A3 R(1)	10A3 P(4)
Nb_2O_5	3.81	4.33	5.33	4.28	3.09	1.65	4.91	6.75
Ta_2O_5	79.89	78.77	76.58	78.57	79.45	80.46	77.46	75.61
FeO	13.07	13.67	13.32	13.35	8.00	12.57	12.23	13.32
MnO	0.90	0.68	0.93	0.86	5.79	1.25	2.67	1.18
SnO_2	0.70	0.49	0.37	0.37	0.53	1.34	0.12	0.19
TiO_2	1.61	1.42	3.36	1.48	2.15	2.00	1.76	1.86
WO_3	0.00	0.41	0.37	0.09	0.32	0.17	0.31	0.33
Total	99.98	99.76	100.26	99.00	99.33	99.44	99.46	99.24
Nb^{5+}	0.141	0.160	0.191	0.160	0.115	0.062	0.181	0.246
Ta^{5+}	1.778	1.753	1.651	1.762	1.776	1.815	1.715	1.659
Fe^{2+}	0.895	0.936	0.884	0.921	0.550	0.872	0.833	0.899
Mn^{2+}	0.062	0.047	0.062	0.060	0.403	0.088	0.184	0.081
Sn^{4+}	0.023	0.016	0.012	0.012	0.017	0.044	0.004	0.006
Ti^{4+}	0.099	0.087	0.200	0.092	0.133	0.125	0.108	0.113
W^{6+}	0.000	0.009	0.008	0.002	0.007	0.004	0.007	0.007
Total	2.998	3.008	3.008	3.009	3.001	3.009	3.032	3.010
$\mathbf{R_{Ta}}$	0.93	0.92	0.90	0.92	0.94	0.97	0.90	0.87
$\mathbf{R_{Mn}}$	0.07	0.05	0.07	0.06	0.42	0.09	0.18	0.08

26S et 10B: grains of tapiolite are homogeneous; 10A: grains of tapiolite are heterogeneous. R: portion of grains rich in Mn; P: portions of grains poor in Mn. The number of analyses carried out is shown in parentheses. The compositions are expressed in wt.% oxides.

The minor elements present in tapiolite are mainly Sn, Ti and W. The levels in Sn vary from 0.002 to 0.067 atoms per formula unit (*apfu*), whereas those of Ti vary between 0.087 and 0.200 *apfu*; W is present in mere traces (<0.016 *apfu*). In a comparison of the compositions of tantalite and coexisting tapiolite, we note the affinity of Sn for the tetragonal structure of tapiolite, in agreement with the observation of Černý *et al.* (1989), whereas tantalite, with its orthorhombic structure, accepts W more readily.

Chemical evolution of the Nb–Ta minerals

In the Suzhou granitic complex, the Nb–Ta minerals are found only in the most evolved facies of granite II, in which they illustrate the generally observed paragenetic sequence: ferrocolumbite (biotite-bearing facies) → ferrocolumbite to manganocolumbite (topaz-bearing facies) → ferrotantalite to manganotantalite + ferrotapiolite (albite-dominant facies). Figure 6 shows evidence of an important late magmatic buildup in Ta, whereas the enrichment in Mn is less clear and seems more complex, with $\mathbf{R_{Mn}} \approx 0.32$ in the biotite-bearing facies, grading to $\mathbf{R_{Mn}} \approx 0.54$ in the topaz-bearing facies, and then to $\mathbf{R_{Mn}} \approx 0.30$ in the case of homogeneous tantalite, and 0.48 in the case of heterogeneous grains typical of the albite-dominant facies. The experimental studies of Keppler (1993) have emphasized the role of fluorine in determining the solubility of columbite-tantalite in the melt. More recently, Linnen & Keppler (1997) have demonstrated that this solubility is a function of the composition of the felsic magma, the solubility of tantalite being twice as high as that of columbite in a peraluminous magma, as monitored by the aluminum saturation index Al/(Na + K + Mn) (Linnen & Keppler 1997), in the range 1.0–1.2. In the Suzhou granite, this index, calculated on the basis of whole-rock compositions, is 1.13 in the biotite-bearing facies, and 1.05 in the albite-dominant facies. The crystallization of columbite can lead to an increase of Ta/Nb.

The pattern of relative enrichment of Nb and Ta during the crystallization of granitic pegmatites is well known (Černý *et al.* 1986, Mulja *et al.* 1996). The same tendency for Ta to become relatively enriched has been found in the Beauvoir granite (Fig. 4; Wang *et al.* 1987, Wang 1988, Ohnenstetter & Piantone 1988). The evolved facies of granite II at Suzhou provide yet another example of progressive fractionation on the basis of Nb/Ta; the biotite-bearing facies was the first to crystallize, with ferrocolumbite having an $\mathbf{R}_{Ta}$ approximately equal to 0.09, followed by the topaz-bearing facies, with an $\mathbf{R}_{Ta}$ of approximately 0.32, and finally by the albite-dominant facies, with an $\mathbf{R}_{Ta}$ of 0.59 and 0.73, for the homogeneous and heterogeneous tantalites, respectively, whereas all tapiolite grains show an $\mathbf{R}_{Ta}$ between 0.87 and 0.92. In general, we can expect that in granitic pegmatite systems, the pattern of behavior of the pair Fe and Mn parallels that of Nb and Ta; furthermore, in the Beauvoir granite, $\mathbf{R}_{Mn}$ increases with advancing magmatic evolution (Ohnenstetter & Piantone 1988, Wang *et al.* 1987). However, the pattern of fractionation displayed by Fe and Mn observed in the Nb–Ta minerals at Suzhou is clearly different, and characterized by weak and early enrichment in Mn. Černý *et al.* (1986) have proposed a correlation between strong increase in $\mathbf{R}_{Mn}$ and the activity of fluorine. Among the evolved facies of granite II at Suzhou, the biotite-bearing facies contains 0.33% F, whereas the topaz-bearing facies contains 3.78% F, and the albite-dominant facies, where topaz-absent, contains only 0.05% (Wang *et al.* 1992). This pattern of distribution of F coincides fairly well with the variation of $\mathbf{R}_{Mn}$ of the Nb–Ta minerals. Furthermore, the patterns of enrichment or depletion of Fe, Mn and Nb, Ta resemble closely those described in the beryl–columbite granitic pegmatite of Cap de Creus, Spain, poor in fluorine (Alfonso Abella *et al.* 1995), and in a pegmatite body near Yellowknife, Northwest Territories, which shows a major, and locally extreme, enrichment in Nb and Ta over a rather limited interval in which $0.05 < \mathbf{R}_{Mn} < 0.45$ (Černý *et al.* 1986). That pegmatite, also of the beryl–columbite type, is equally depleted in F; by analogy, the unusual pattern of enrichment of Fe, Mn in the evolved facies at Suzhou could be due to the restricted activity of fluorine in the magma. The presence of tapiolite is known to be limited to granitic pegmatites having a low activity of fluorine and, consequently, limited fractionation of Fe and Mn (Černý & Ercit 1989).

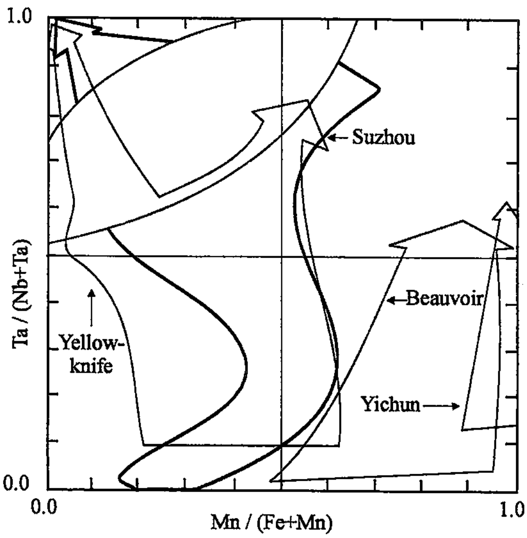

FIG. 6. Pattern of relative enrichment in columbite–tantalite of the Suzhou granite, compared with that in the Beauvoir (Wang 1988) and Yichun (Belkasmi *et al.* 1992) granites, and the granitic pegmatite from the Yellowknife area (Černý *et al.* 1986).

CONCLUSIONS

In the granitic complex at Suzhou, the Nb–Ta minerals are present in the three facies of granite II, enriched in biotite, topaz and albite, respectively. We note that columbite is most prominent in the earliest, biotite-bearing facies, whereas tantalite and tapiolite are concentrated in the latest facies, enriched in albite. The extreme fractionation of the Nb and Ta during the crystallization of the felsic magma provokes an important increase in $\mathbf{R}_{Ta}$ of the Nb–Ta minerals, from 0.09 (biotite-bearing facies) to 0.92 (albite-dominant facies); at the same time, the limited variation in $\mathbf{R}_{Mn}$ is manifested in an increase from the biotite-bearing facies to the topaz-bearing facies, and then a decrease in the albite-dominant facies, the activity of fluorine being considered the main factor that affects this fractionation. The presence of ferrotapiolite could be the sign of the relative activity of Fe in the residual magma. We also note that the tapiolite that is associated with tantalite has an unusual composition, beyond the field of stable compositions of tapiolite, suggesting disequilibrium conditions of crystallization.

In the evolved facies of the Suzhou and Beauvoir granites, the Nb–Ta minerals reveal important distinctions in the behavior of Fe and Mn. The Suzhou granite is notable also for its elevated concentrations of Hf, Th, Y, and the rare earths, manifested in zircon, thorite, xenotime and monazite (Wang *et al.* 1996). These distinct characteristics allow us to classify the Suzhou granite as an example of the "low-phosphorus subtype" (P_2O_5 < 0.1%), according to the criteria of Taylor (1992), with certain peculiarities (F, Hf, Al) compared to other evolved granites, for example the Pleasant Ridge (Taylor 1992), Beauvoir and Yichun (Belkasmi *et al.* 1992) granites, which are examples of the phosphorus-rich subtype.

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