

AFRICAN RARE-ELEMENT PEGMATITE PROVINCES: MINERALOGY AND GEOCHEMISTRY OF NB-TA MINERALS

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INTRODUCTION

Since about the year 2000, the African continent has developed into a major exporter of “coltan” – heterogeneous concentrates of niobium-tantalum ore mined from rare-element pegmatites and their eluvial and alluvial weathering products by mainly artisanal, small-scale mining operations. Probable links between raw material exports (i.e. tin, tantalum, tungsten, gold and diamonds) and continuing violent conflicts, especially in the Democratic Republic of the Congo (DRC), are widely discussed. On behalf of the German Federal Ministry for Economic Cooperation and Development (BMZ), the Federal Institute for Geosciences and Natural Resources (BGR) commenced a pilot project to test the general viability of “fingerprinting” the origin of coltan back to their source pegmatites.

METHODS

Polished sections were prepared from ore concentrates and investigated by quantitative mineralogical analysis using the Mineral Liberation Analysis software

on a Quanta 600 FEG scanning electron microscope. Ta-Nb-bearing minerals (mainly columbite-tantalite group minerals, CGM) were analyzed for major and trace elements by electron microprobe (CAMECA SX100). Trace elements were detected either in situ by LA-ICP-MS (193 nm laser coupled to a Thermo-Scientific Element II sector field ICP-MS, BGR; 266nm laser coupled to a Quadrupole-ICP-MS; H. Brätz, University of Erlangen) or by ICP-MS after sample dissolution using HF-HNO₃. U-Pb dating of columbite-tantalite grains was carried out both on crystal fragments using conventional thermal ion mass spectrometry (TIMS), and in-situ using an Element II ICP-MS coupled to a New Wave UP213 ultraviolet laser system with low-volume ablation cell (A. Gerdes, University of Frankfurt). The chemical procedures to separate U and Pb for TIMS analysis were adapted from Romer and Wright (1992).

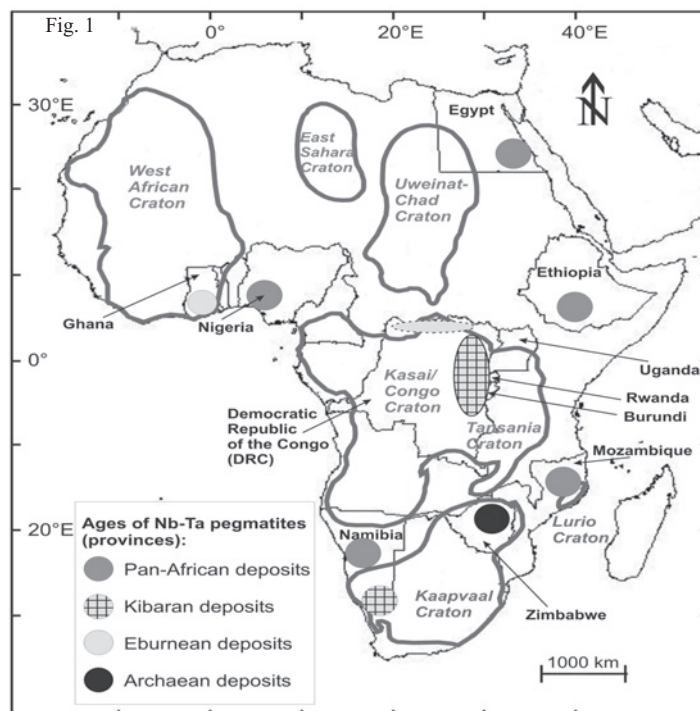
AFRICAN TANTALUM PROVINCES

Rare-element granites and pegmatites are distributed throughout the African continent (e.g., von Knorring &

Condcliffe 1987). Five “age provinces” have been established (Fig. 1): (1) Archean, e.g. Zimbabwe craton; (2) Paleoproterozoic, e.g. in the Birimian of West Africa; (3) early Neoproterozoic, e.g. Kibaran and Namaqualand Province; (4) late Neoproterozoic to Early Paleozoic, e.g. the Panafrican Belts of Eastern Africa, Namibia (Damara orogen) and Nigeria; (5) Mesozoic ring complexes of Nigeria.

Four areas were selected to document regional and local variations: the Kibaran belt of Ruanda and the DRC; the Alto Ligonha Province of Mozambique; the Adola belt of southern

Ethiopia; and the Tantalite Valley area in southern Namibia. Their characteristic features are summarized in Table 1. In addition, samples from central Namibia (Damara belt), Nigeria (Panafrican and Mesozoic examples), Ghana (Kokobin, Ghana; Birimian), Zimbabwe (Benson mine; Bikita; both Archean) and Egypt (Nuweibi) have been studied. In the following, we summarize mineralogical and geochemical data collected from the study of coltan ore concentrates and of single Ta-Nb mineral specimens of the pilot study.



KIBARAN BELT AND ADJACENT DRC-CAR CRATON

The NNE-SSW-oriented Kibaran Belt, extending from Uganda via Rwanda, Burundi and finally into the Katanga

region of the DRC (Pohl 1994), is Africa’s major producer of Ta-Nb, Sn and W. The largest single pegmatite, Manono-Kitotolo (Katanga Province) hosted total reserves of ~100 million tonnes of eluvial/alluvial and primary ore.

The Kibaran Belt is composed of Paleo- and Mesoproterozoic clastic sediments that have been intruded by various generations of granite. The oldest granites have been dated at 1.38 Ga, whereas the youngest are slightly younger than 1.0 Ga. Mineralization is exclusively connected with the youngest post-orogenic “tin granites”, also called G4 granites. It is developed in quartz veins, greisens and small pegmatitic bodies that are occasionally zoned, and which occur in some distance to the G4 granites (Varlamoff 1972). Most rare-metal pegmatites are of the lithium-caesium-tantalum type (LCT). They generally carry Sn, and many contain Ta-Nb, in addition to locally abundant Li, Be and phosphate mineralization. Cassiterite is also abundant in veins that are locally spatially related to pegmatites. However, cassiterite-bearing veins are usually devoid of tantalum mineralization. Quartz-wolframite veins are usually free of Sn and Ta.

In many Kibaran pegmatites, the tantalum mineralogy is dominated by columbite-(Fe,Mn) and tantalite-(Mn). Individual crystals are commonly complexly zoned, and tantalite-(Fe), tapiolite, wodginite and microlite are locally abundant. Rare phases such as stibiotantalite, bismutotantalite, rankamaite, rynersonite, cesplumtantite etc. have been reported from some occurrences. Most pegmatites carry abundant cassiterite. In general, two types of fractionation trends are observed: (1) columbite-(Fe) → tantalite-(Fe), with abundant tapiolite, and (2) columbite-(Fe) → columbite-(Mn) → tantalite-(Mn), commonly with wodginite and rare tapiolite. Minor element concentrations in CGM vary over wide ranges and display regional patterns: in the Kivu and Maniema provinces, CGM have elevated concentrations of W, Ti and Zr compared

to Katanga, Rwanda and Burundi. Concentrations of Sc, Li, Be, Bi, As and Sb are low, and uranium is highly variable. Rare earth element concentrations are generally low, and chondrite-normalised REE patterns show a variety of shapes, but lacking distinct Eu anomalies. U-Pb ages of columbite-tantalite and tapiolite range from 920 to 1010 Ma.

Tantalum-rich placer deposits have been intermittently mined in the Ituri Province (DRC), within the Archean “northern DRC-CAR Craton”. Pegmatitic veins carry columbite-(Fe), tantalite-(Fe), tapiolite, and some cassiterite and wolframite. U-Pb determinations yielded two populations attributed to 2480 Ma pegmatite intrusion and subsequent “rejuvenation” and intrusion during the Ubendian event close to 2000-2100 Ma.

ALTO LIGONHA PROVINCE, MOZAMBIQUE

More than 100 pegmatites are known from the Alto Ligonha Province, which forms part of the Zambesia Province in NE Mozambique (Lächelt 2004). The large Marropino pegmatite is a deeply weathered, kaolinitized, zoned LCT rare-metal pegmatite that intruded into mafic gneisses and schists of the Proterozoic Morruea Formation. The main ore body extends for 1 km in ENE-WSE direction and is up to 80 m thick. The pegmatites at Morruea, ca. 50 km north of Marropino, are less altered. Drilling has indicated six pegmatite bodies each >3 m thick, and extending up to 1 km along strike. At Mutala, 90 km north of Marropino, zoned pegmatites form up to 50 m thick bodies intruded into mica- and amphibole-schist.

Tantalum mineralization is dominated by primary microlite and tantalite-(Mn); columbite-(Fe), tantalite-(Fe) and tapiolite are virtually absent.

Bismutotantalite and REE-Th-bearing Ta oxides (e.g. samarskite, euxenite) that exhibit complex zoning patterns, are abundant at Marropino. Ore concentrates are characterized by common REE phases (monazite, xenotime) and zircon, and lack cassiterite. Concentrations of Sn, Zr, Hf, W in CGM are low, those of Sc and Ti are

high; U may be considerably enriched. REE concentrations are high and normalized REE patterns show pronounced negative Eu anomalies and fractionated MREE-HREE abundances ($Tb_n/Ho_n \sim 5-10$, $Gd_n/Lu_n \sim 10$). U-Pb ages of CGM range from 440 Ma (Moneia) to 473-481 Ma (Morrua, Marropino).

Table 1. Characteristics of major African coltan provinces

Province	Kibaran	DRC-CAR craton	Alto Ligonha	Adola Belt	Tantalite Valley
Countries	DRC, Rwanda, Burundi, Uganda	DRC (Ituri)	Mozambique	Ethiopia	Namibia
Localities (examples)	Manono, Pangi, Shabunda, Gatumba	Mambasa, Lubero	Marropino, Morrua, Mutala, Muiane	Kenticha, Bupo, Shuni Hill	Homestead, White City, Lepidolite
Host rocks	Metasediments, metabasite, granite	Gneiss, amphibolite, granite	Metabasite, metasediments	Ultrabasite, metasediments	Metabasite (gabbro), metasediments
Age of host rocks	Mesoproterozoic (Kibaran)	Archean, Paleoproterozoic	Mesoproterozoic	Neoproterozoic	Paleo- to Mesoproterozoic
Mineralization type	LCT pegmatites	unknown	LCT pegmatites	LCT pegmatites	LCT pegmatites
Age of intrusions	920 - 1010 Ma	1950 - 2490 Ma	440 - 480 Ma	530 Ma	985 Ma
Tantalum mineralogy	FeC, FeT, MnC, MnT, Tap, Wod, Mc	FeC, FeT, Tap, Mc	MnC, MnT, Mc, UMc, BiT	MnC, MnT, UMc, Mc	MnT, MnC
XMn	56 ± 18	25 ± 8	78 ± 17	76 ± 24	99 ± 1
XTa	37 ± 16	33 ± 7	57 ± 24	56 ± 17	84 ± 2
wt% TiO ₂	0.52 ± 0.39	1.71 ± 1.31	1.01 ± 0.77	0.37 ± 0.14	0.61 ± 0.16
wt% SnO ₂	0.28 ± 0.32	0.11 ± 0.15	0.07 ± 0.08	0.07 ± 0.02	<0.01
Production data	>600 t concentrates (2008), artisanal	artisanal	70 t Ta ₂ O ₅ (Marropino, 2007); artisanal mining	81 t Ta ₂ O ₅ (Kenticha, 2007)	dormant

Chemistry of columbite-tantalite group minerals: mean ± standard deviation; $XMn = 100 * Mn / (Mn + Fe)$; $XTa = 100 * Ta / (Ta + Nb)$, in mole%. Mineralogy: FeC, columbite-(Fe); FeT, tantalite-(Fe); MnC, columbite-(Mn); MnT, tantalite-(Mn), Tap, tapiolite, Wod, wodginite, Mc, microlite; Umc, uranmicrolite; BiT, bismutotantalite

Adola Belt, Ethiopia

The pegmatites in the Oromia Regional State intruded into a Precambrian terrane that is dominated by granitoids and ophiolites emplaced between 0.9 and 0.7 Ga. Collisional granitic magmatism occurred between 0.70 and 0.55 Ma. The pegmatites were classified as barren, beryl-columbite, complex spodumene and albite-spodumene. In the zoned Kenticha

LCT rare-metal granite pegmatite, the tantalum mineralization is hosted by post-tectonic alaskite and associated granitic pegmatite within fractured and partly sheared serpentinite along a regional-scale thrust fault separating the low-grade Kenticha greenstone belt from medium to high grade gneisses (Küster et al. 2009).

Tantalum mineralization at Kenticha includes zoned tantalite-(Mn), columbite-(Mn), as well as microlite, pyrochlore, uranmicrolite, and rare tapiolite, ixiolite/wodginite and Ta-bearing rutile. CGM compositions from Kenticha comprise a trend from columbite-(Fe-Mn) to tantalite-(Mn). CGM from the Bupo pegmatite are also Mn-rich ($XMn > 85$) at variable XTa. At the Shuni Hill pegmatite, CGM have columbite-(Fe) compositions with intermediate XMn (40) and XTa (30) numbers.

Concentrations of U in CGM may exceed 1000 ppm. Sn and Zr are both progressively enriched during Ta–Nb fractionation, whereas Ti and W decrease with XTa. REE concentrations of CGM show distinctive patterns for each of the three localities investigated. Most tantalites from the Kenticha orebody are characterized by low total REE and, if any, weak negative Eu anomalies. Columbite-(Fe), on the other hand, has higher total REE and small though distinct negative Eu anomalies. The REE patterns are generally less fractionated at Kenticha compared to Shuni Hill and Bupo, which both display high La_n/Lu_n ratios. U–Pb ages determined on tantalite-(Mn) from the Kenticha and Bupo pegmatites consistently yielded crystallization ages close to 530 Ma (Küster et al. 2009).

Tantalite Valley, Namibia

The Orange River Pegmatite Province extends from the northwestern Cape Province of South Africa into southern Namibia. The Tantalite Valley pegmatites are part of the Mesoproterozoic Tantalite Valley Basic complex, which is composed of olivine gabbro and gneisses. Up to 1 km long and > 10 m thick rare metal pegmatites of the LCT type intruded along a shear zone (Diehl 1992). Pegmatites commonly show symmetrical zonation. The major minerals are quartz, K-spar, albite and white mica, accompanied by spodumene, lepidolite, amblygonite, gahnite, beryllium and bismuth minerals, CGM, and phosphates.

Tantalum mineralization is characterized by a lack of cassiterite, and rather homogeneous, rarely zoned tantalite-(Mn) with $\text{XMn} = 95\text{--}100$ and $\text{XTa} = 40\text{--}90$ as the major oxide phase. W and U average to 3000 and 2000 ppm, respectively. Normalized REE concentration pattern

reveal distinct negative Eu anomalies, fractionated MREE/HREE with $\text{Tb}_n/\text{Ho}_n \sim 50\text{--}100$ and $\text{Gd}_n/\text{Lu}_n \sim 10$. U–Pb age determinations using LA-ICP-MS yielded 982 to 985 Ma.

SUMMARY

Determination of mineral formation ages using the U–Pb isotope system traces the origin of a coltan concentrate to one out of five “age provinces” known from African tantalum-bearing pegmatites. Mineralogical parameters such as modal mineralogy, grain size distribution and intergrowth relationships are different for different tantalum provinces in Africa. Coltan is a mixture of a large number of minerals, commonly dominated by columbite-tantalite group minerals. In some cases, their compositions and textural relations are considered as diagnostic proofs of origin. The concentrations of major and trace elements analysed in a representative number of columbite-tantalite grains from a given sample are also suitable for fingerprinting. Normalised REE concentration patterns can be used to differentiate between individual locations. So far, our data show that gross discrimination of “coltan provinces” is possible using age determination. Mineralogical and geochemical attributes of coltan concentrates and columbite-tantalite group minerals further allow distinction of single deposits within certain age provinces.

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