



Review Paper

A perspective view of origins and occurrences of Niobium-Tantalum, Tin and Tungsten mineralisation in Rwanda

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Abstract

Rwanda is geologically situated in the Karagwe Ankole Belt, centre east Africa. Karagwe Ankole Belt (KAB) is separated from Kibara Belt (KIB) by a Paleoproterozoic Rusizian terrane and they both form mesoproterozoic orogenic belts of Central Africa. The KAB, which spans Rwanda, Burundi, Southwestern Uganda and Northwestern Tanzania, and the KIB together host a large metallogenic province that is composed of numerous rare -metal ore deposits mineralised in niobium-tantalum (Nb-Ta), tin (Sn) and tungsten (W). The part of the KAB located in Rwanda contains the bulk of cassiterite, columbite-tantalite, and wolframite. These ore minerals have been termed 3Ts and occur in Nb-Ta-Sn pegmatites, W-Sn hydrothermal quartz vein deposits and Sn greisens, which are components of one composite metallogenic system related to the granite generation (G4-granite or fertile granite) that occurred at 986 ± 10 Ma. The composition of hydrothermal fluid is $H_2O-CO_2-CH_4-N_2-NaCl$, and this fluid is characterized by the low to moderate salinity (2.7-14.2 eq. wt.% NaCl), high pressure (~100 MPa), and the mesothermal temperature (~300°C). The isotopic composition of the fluids indicated that the mineralised quartz veins are much more likely to be formed from the fluid mainly subjected to metamorphic processes, in equilibrium with magmatic rocks (G4 granite) from which the present precipitated metals were remobilised and got deposited in favoured preferential sites, within metasedimentary rocks, under structural and lithological controls. This paper owes the brief on the current status of views about the mineralisation of 3Ts in Rwanda.

Keywords: Karagwe-Ankole belt, Kibara belt, cassiterite, columbite-tantalite, wolframite, Rwanda.

Introduction

Background: Rwanda is a small mountainous landlocked country in the Great Lakes region of Africa. The country shares borders with the Democratic Republic of the Congo (DRC), Uganda, Tanzania and Burundi, and is located at $S2^{\circ}00.00'$ latitude and $E030^{\circ}00.00'$ longitude. The total land area is about 24,950 km², and inland lakes cover about 1390 km²¹. Rwanda is positioned in the south western part of the north-eastern Kibara belt (KIB), centre east Africa. The Kibara belt extends from Katanga DRC, through Burundi, western Tanzania, Rwanda, and northeast to southwest of Uganda. The Kibara belt consists of composite rocks from the paleo-mesoproterozoic age. The Kibara belt is well-known for its abundance of niobium-tantalum (Nb-Ta), tin (Sn), tungsten (W) and gold (Au) mineralization². In Rwanda, the Nb-Ta, Sn, W mineralisations mainly occur in pegmatite and quartz vein²⁻⁴. The metals exist in various forms of mineralisation considered to be granite related ore deposits⁵⁻⁷.

They occur as primary mineralisation in the form of pegmatite, quartz vein, and greisen, but also as secondary mineralization in alluvial or eluvial deposits⁷. There are two

major generations of granite which intruded the Kibaran rocks of KAB. The first generation is intrusion of granites G1-3 which occurred at 1380 ± 10 Ma. Within this geological period, at 1375 Ma, there has been prominent emplacement of bimodal magmatism (granitic and mafic) under extensional regime of Kibaran event in central Africa. Conversely, the second generation which is the intrusion of the G4 granite (or the so-called 'fertile granite') formed at 986 ± 10 Ma. The mineralized pegmatites and quartz veins are considered to be related to the G4-granite⁸.

Geological settings: Regional geology: The paleo-mesoproterozoic KAB and KIB emerged through a regional orogenic event between three pre-mesoproterozoic domains, the Archaean Tanzania craton to the east, the Bangweulublock to the south, and the archaean-palaeoproterozoic Congo craton to the west and north (Figure-1). In the northern part of the Kibara orogeny, which spans Rwanda, Burundi and Kivu, the granite subdivided into three types (G1-3), and intruded the paleo-mesoproterozoic rocks dated at 1380 ± 10 Ma.

The crystallization of these granites did not result in concentrations of economically valuable rare metal deposits.

Later, at 986 ± 10 Ma, the granite generation known as G4 granite (fertile granite, rare metal granite or tin granite) resulted in the emplacement of rare metals deposits⁹.

The Karagwe-Ankole belt and Kibaran belt comprise a fluctuate sequence of pelitic and carbonate sediments, minor volcanics and dolerite. The structural settings, stratigraphy, metamorphism of the Kibaran orogeny are well-documented by previous authors. The Kibaran belt is known for the emplacement of the rare metals niobium-tantalum (Nb-Ta), tin(Sn), tungsten(W), and gold(Au).

The primary emplacement of Sn was observed in hydroxyl bearing phases of granites and mineralisation hosted in the pegmatites and/or quartz veins, the W occurred in hydrothermal quartz veins was geochemically originated from the fertile granites, and a decisive fluid-rock interaction induced tungsten deposits^{10,11}. The Nb-Ta in pegmatite were mostly precipitated by driven hydrothermal fluid in the original pegmatites which were subsequently altered by hydrothermal processes¹².

These mesoproterozoic formations comprise three main lithological units: low-to-medium grade metavolcanic and metasedimentary sequences, large granite batholiths (with inliers of basic and metasedimentary rocks), and large complexes of high grade metasediments to amphibolites with granite, greisens and migmatites, the sediments within Rwanda have been subdivided into four stratigraphic groups, from oldest to the youngest, which are Gikoro, Pindura, Cyohoha and Rugezi groups^{16,17}.

The general pattern of meso-proterozoic domain in Rwanda comprises resistant cores (high-grade units) characterized by weak deformation separated by "Intensely Deformed Zones," noted as Shear Zones¹⁸.

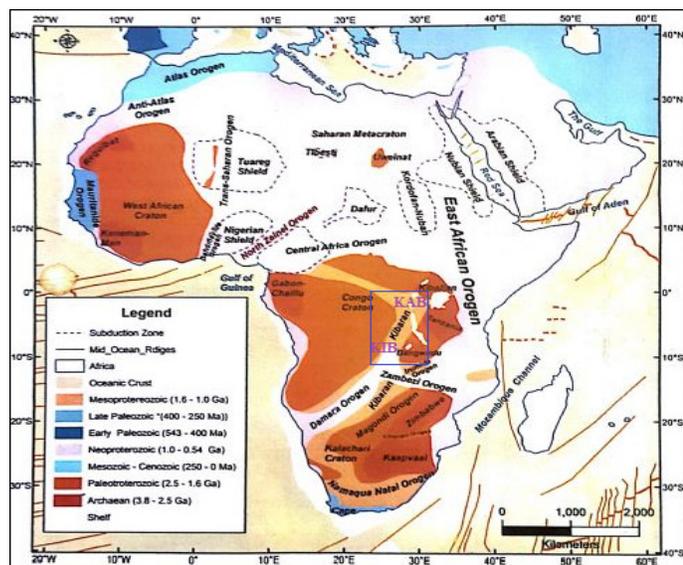


Figure-1: Geological map of Africa showing the Cratonic zones, KAB and KIB, with their respective ages of crustal formation^{13,14}.

Geology of Rwanda: Rwanda is underlain by the rocks of the Kibara Orogeny which consists predominantly of the basement and mesoproterozoic rocks (ca. 1.6-1.0Ga), and have been intruded by rocks of bimodal magmatic event (granitic and mafic rocks)^{13,14}. In Kibaran belt, including Rwanda, there is mineralisation of tin (Sn), niobium-tantalum (Nb-Ta), tungsten(W) and gold(Au) which mostly occur in the pegmatites, greisens and quartz veins in relation to the G4 granite¹⁵. The geology of Rwanda consists of middle (meso) proterozoic formations with tertiary age, East African rift valley, volcanic cover in South Kivu, Cyangugu and in the north-western Birunga mountains¹⁵.

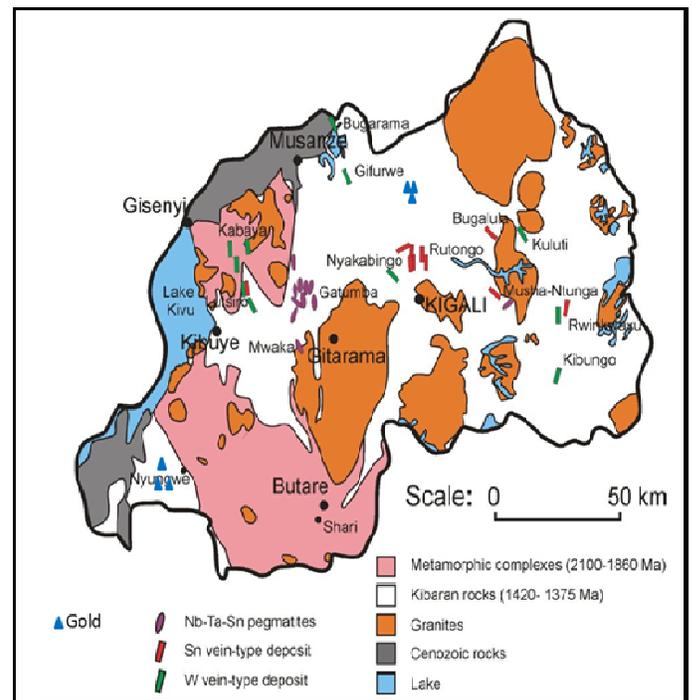


Figure-2: The geological map of Rwanda showing the ore deposits^{5,19,20}.

Generally, Rwanda hosts a large number of ore deposits located in various metallogenic places of Rwanda^{19,20}. the major commodities, economically mined now and in the past, being cassiterite (SnO_2), niobo-tantalite also called colombo-tantalite or coltan $(\text{Nb,Ta})_2\text{O}_5$ and Wolframite $(\text{Fe,Mn})\text{WO}_4$, the minor commodities (accessory minerals) found in association are beryllium $(\text{Be}_3\text{Al}_2\text{Si}_6\text{O}_8)$, spodumene $(\text{LiAlSi}_2\text{O}_6)$, amblygonite $(\text{Li,Na}) \text{AlPO}_4 (\text{F,OH})$, monazite $(\text{Ce,L a,Nd,Th}) \text{PO}_4$, tourmaline $(\text{Ca,K,Na}) (\text{Al,Fe,Li,Mg,Mn})_3 (\text{Al,Cr,Fe,V})_6 (\text{BO}_3)_2 (\text{Si,Al,B})_6\text{O}_{18}$ and gold (Au)^{21,22}.

They occur as primary mineralization in the pegmatites, greisens and quartz veins but also during the secondary mineralization in alluvial or elluvial deposits.

Columbite-tantalite found in association with cassiterite mineralisation in pegmatites, due to growing demand for the tantalum metal in current technological industrialisation, the coltan (tantalum ore) has only turned into economically

relevant in more recent times, later surpassing tin ore (cassiterite) and tantalum ore became the major export earner in 2012^{23,24}.

Table-1: The major mined ore deposits, representative mines and occurrences in Rwanda.

Ore mineral	Occurrence	Typical local mine
Columbite-tantalite	Mineralization occurs in pegmatites	Gatumba, Nyarusange, Ruli
Cassiterite	Mineralization occurs in quartz veins, greisen and pegmatite	Rutongo, Rwinkwavu, Musha-Ntunga, Gatumba
Tungsten	Mineralization occurs in quartz veins	Nyakabingo, Gifurwe and Bugarama

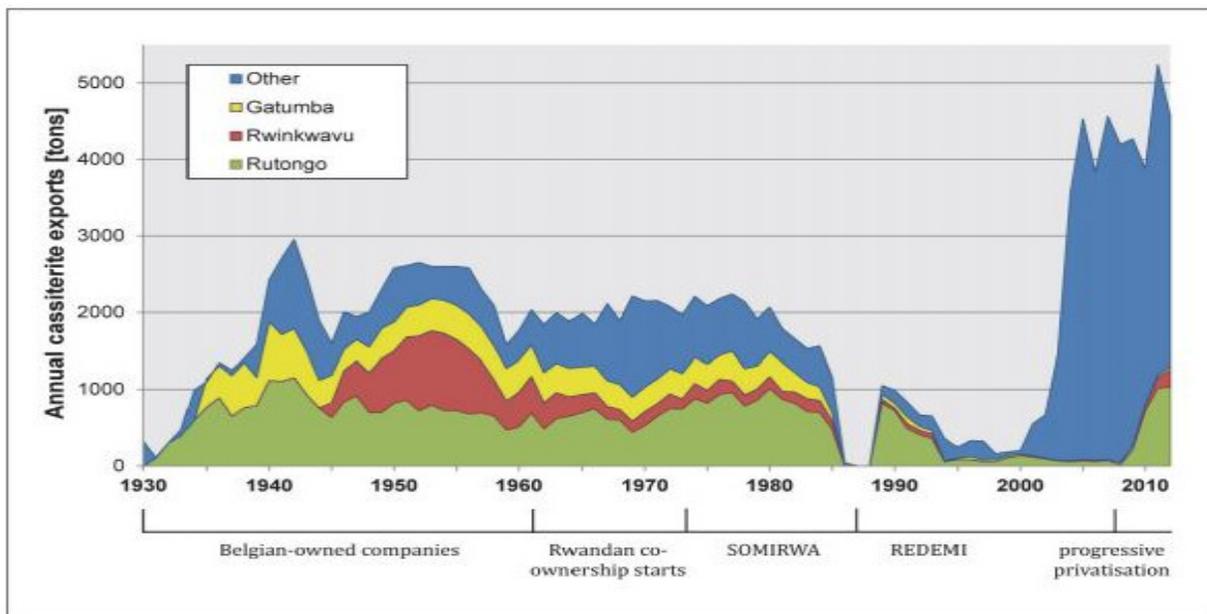


Figure-3: The exported production of cassiterite concentrates from Rwanda²³.



Figure-4: Historical production of 3Ts minerals in 5 past years, Rwanda²⁴.

Figure-4 indicates the annual production of 3Ts in the five past years in which 2014, Rwanda has reported the highest exporter of tin ore in the region. Subsequently, the production has dramatically decreased due to the prices which fell down on the international markets.

Contrary, the high demand of Coltan has been recently caused by the increasing demand from emerging markets and newly industrialised countries in Asia and South America, which are undergoing rapid economic growth²⁴.

Research Findings

Mineralised pegmatites and quartz veins: The Nb-Ta-Sn-bearing pegmatites are found at the middle and outer contact metamorphic aureole of fertile granite and are noticeably linear or lentoid or crosscut by quartz veins of centimetres in size (eg. Ntunga Nb-Ta-Sn pegmatite). Those pegmatites range up to the width of 10 meters and approximately length of 250 meters²⁵. For most parts of the area, these pegmatites are concordant with the regional foliation but some discordant and sub-horizontal bodies are also noticed.

Typically, the pegmatites are weakly bared and widely altered. Mine workings are limited to artisanal mining under the surface to some depths ranging less than 25 meters in altered materials. Mostly, the ore minerals such as columbite-tantalite, cassiterite and tungsten exist as coarse crystals or aggregate²⁶. Other non-silicates found in the heavy mineral concentrates are rutile, ilmenite and magnetite. Phosphate minerals, including amblygonite, wardite and augelite, are noticed in some occurrences. In large occurrences, the K-feldspar is partially to totally subjected to metasomatism and turned into pure kaolin. However, minor fresh K-feldspar is present in fresh granitoids¹⁰.

In the W/Sn-bearing quartz veins, the tungsten mineralisation is predominantly present in quartz veins, and tin is found either mixed with columbite tantalite in pegmatites or by itself in quartz veins.

Stable isotopes in ore deposits: The D/H and ¹⁸O/¹⁶O analyses are valuable applications to determine the history and origin of

H₂O in the hydrothermal fluids. The composition of hydrogen and oxygen isotopes was determined using the standard analytical techniques of Clayton and Mayeda to look into the origins of hydrothermal fluid of water in the sources of primary magmatic, metamorphic, meteoric water, organic and kaolinite lines^{27,28}.

Stable isotope studies therefore suggest a model in which a metal of interest (e.g. Nb-Ta-Sn-W) was moved from primary magmatic rocks during the metasomatism of metamorphic hydrothermal fluid systems that could be generated after crystallisation of the granites and pegmatites. The ore minerals were precipitated in structurally controlled emplacements, along with the alteration of the host-rocks.

Columbite-tantalite formed during the crystallisation of the pegmatites followed by intense alkali metasomatism which advanced the formation of albite (albitisation) and white mica²⁹.

The tungsten was postulated to have originated from a fluid mostly brought out by metamorphic processes, however, there is another model that suggests the fluids have a magmatic signature³⁰.

Seven quartz samples of the mineralised veins from Nyakabingo, Gifurwe and Rutongo were investigated by the stable isotope studies to determine the sources of mineralising fluids²⁹. The hydrogen isotopic composition of fluid inclusions inside the quartz crystals and the oxygen isotopic composition of quartz crystals were measured. Quartz samples were handpicked and O&H stable isotope analyses were carried out at SUERC. The samples were analysed using fluorination procedure and CO₂ laser as a heat provider. Reproducibility is better than 0.3% (= 1 σ). The reported results are ‰ deviation from the V-SMOW standard. The table 2 summarises the results which are presented in Figure-6. The samples analysed have almost identical δ¹⁸O values, but the sigma D values diverge considerably, i.e. between -30‰ to -115‰ V-SMOW. The δ¹⁸O composition of the ambient fluid from which the quartz crystals precipitated has been calculated using the equation of Matsuhisa et al³¹.



Figure-5: Field photographs taken during the field investigation. A. Intrusion of advanced argillic alteration of pegmatites with concentrates of coltan at Ntunga. B. Quartz vein mineralized in cassiterite and hosted in schist at Rwinkwavu. C. Quartz vein mineralized in tungsten and hosted in black shale at Nyakabingo.

Pohl and Günther³ and Pohl² estimated a formation temperature between 300°C and 500°C for the quartz crystals based on microthermometry. The $\delta^{18}\text{O}$ composition of the ambient fluid in equilibrium with quartz has been calculated for the temperatures 310°C and 510°C.

water at Rutongo, Nyakabingo indicated the source of metamorphic conditions, but Gifurwe was influenced by water-rock interaction of fluid mostly resulted from organic settings (Figure-6).

The hydrogen and oxygen isotope characteristics of water were reasonably diagnostic for its source, the hydrothermal fluid of

The movement of fluid was typically a response to temperature and/or pressure gradient which causes the both stress and strain to vary in rocks subjected to the deformation.

Table-2: Stable isotope data of mineralised quartz veins at Nyakabingo, Gifurwe and Rutongo^{3,4,30}.

Samples	δD fluid	$\delta^{18}\text{O}$ Quartz	$\delta^{18}\text{O}$ Quartz(310°C) (V-SMOW)	$\delta^{18}\text{O}$ Quartz(510°C) (V-SMOW)
NY06SD01	-53	15.2	8.2	13.1
NY06SD06	-45	14.9	7.1	13.0
NY06SD07	-32	14.4	7.0	12.9
GI06SD02	-90	1.33.64.9	-	-
GI06SD08	-100	0.32.13.5	-	-
GI06SD09	-112	2.03.95.2	-	-
RU06SD07	-30.1	14	7.2	12.5

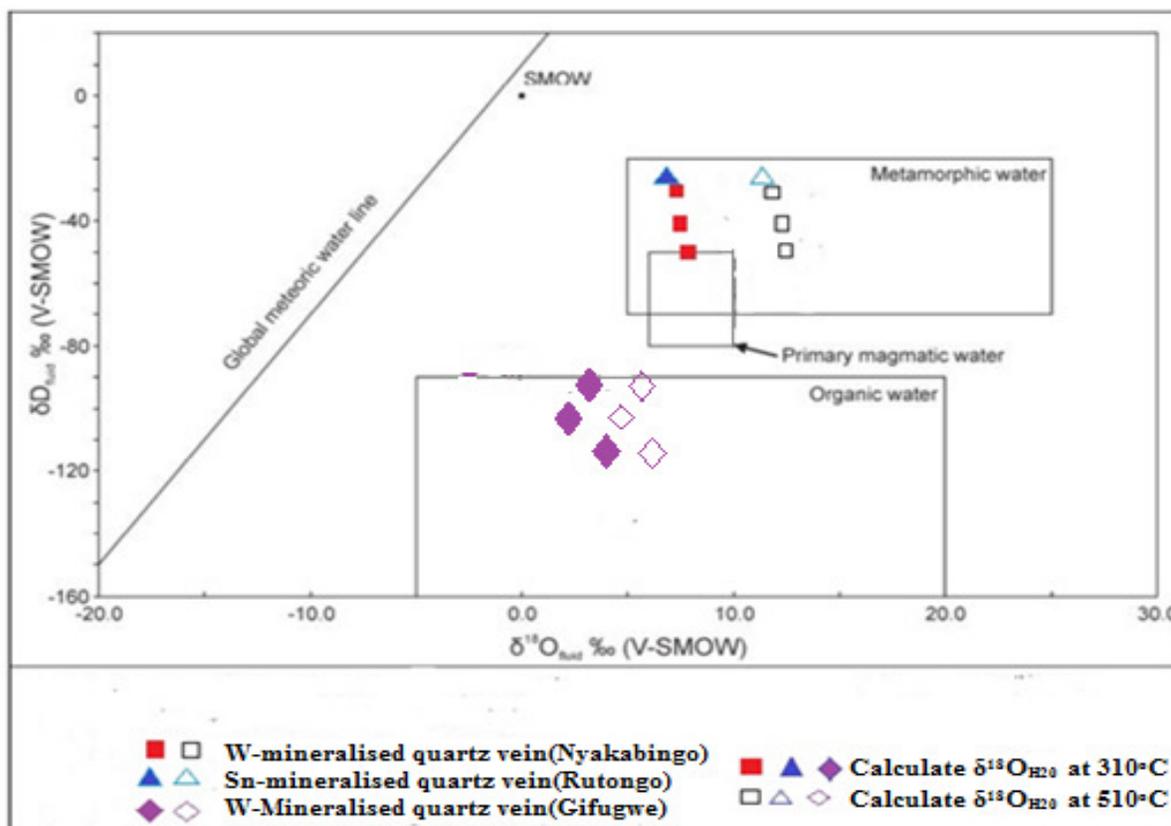


Figure-6: $\delta^{18}\text{O}$ - δD isotopic plot for the calculated fluid composition of the mineralised quartz veins of Nyakabingo, Gifurwe and Rutongo^{4,30}.

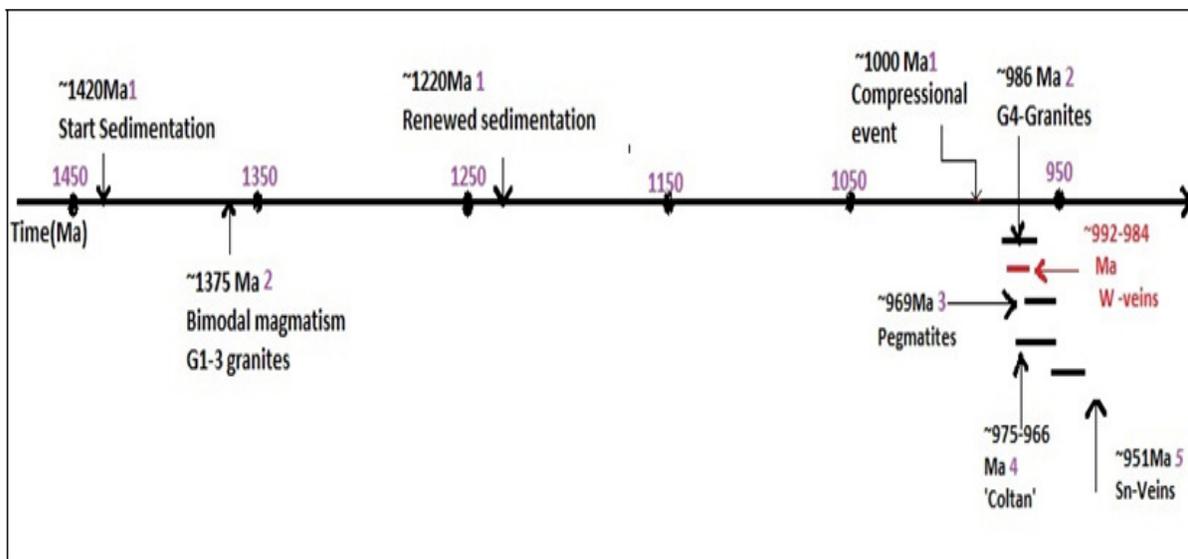


Figure-7: Radiogenic isotopic data in Rwanda^{6,8,17,35,36}.

Radiogenic isotope studies: We can largely define the principal applications of radiogenic isotope geochemistry by considering geochronology which makes use of the constancy of the rate of radioactive decay to measure time. As a radioactive nuclide decays to its daughter at a rate independent of everything, the time can be simply be deduced from the numbers of nuclide decayed. Based on earlier geochronological studies, the young Rb-Sr and ⁴⁰Ar-³⁹Ar ages for granites, pegmatites and veins have geochronological characterisation related to the crustal formation of Africa (≤ 960 Ma)³²⁻³⁴. These ages are most probably resulted in a reset of the radiogenic system due to recrystallisation or thermal re-equilibration during younger (Pan-african) tectono-thermal events. An integration of ages of mineralisation of 3Ts i.e Nb-Ta in the form of columbite tantalite, Sn in the form of cassiterite and W in the form of wolframite as well as ages of related parental granites in Rwanda with other occurred tectonic events are represented by the Figure-7.

The intracontinental Kibaran belt evolved in paleo-mesoproterozoic orogeny between 1400 to 900 Ma^{35,36}. later the kibaran belt was intruded by numerous granite bodies formed G4 granite (fertile granite) at 986 +10 Ma. The later granite was followed by the emplacement of granitic pegmatites and pegmatites at ~970 Ma. The granitic rare earth metals were precipitated and deposited into various occurrences of 3Ts mineralisation between 986 to 951 Ma (Figure-7).

Fluid inclusion microthermometry: By focusing on fluid inclusions captured within hydrothermal veins, we have a better picture than previous one about the nature of the processes and mineralizing fluids which formed mineral deposits. A new way to study minerals with large fluid inclusions, often coarse-grained was found in the techniques of microthermometry and bulk chemical analysis³⁷.

In this, idea was a way of analysing sample material to look into minerals with large fluid inclusions often coarse-grained, absolutely matched to the apprentice techniques of microthermometry and bulk chemical analysis according to the naissance father of fluid inclusion research, Henry Clifton Sorby³⁷.

In his usual paper Sorby³⁷ outlined samples from ore deposits having fluid inclusions and drew conclusions concerning ore formation that remained unpopular for many years.

Tin and tungsten mineralised quartz veins at Rutongo and Nyakabingo respectively formed from fluids that had the same primary magmatic origin and a similar evolution.

The gas composition of nitrogen and methane gases (N₂ and CH₄) and the temperature of the tungsten (W) mineralising fluid (~300°C) relatively indicate metamorphic conditions³⁸⁻⁴¹. Based on a similar geological setting and a comparable paragenesis and fluid inclusion composition, Dewaele et al.⁴ determined the oxygen and hydrogen isotopic composition of tin-mineralised quartz veins from Rutongo (Rwanda). The $\delta^{18}O$ - δD values of the tin mineralising fluids also fall in the field typical for metamorphic water (Figure-6).

Pohl and Günther³ and Pohl² postulated that the tungsten mineralisation at Nyakabingo formed from an H₂O-CO₂-CH₄-N₂-NaCl fluid characterized by the salinity (7.44-99. eq. wt.% NaCl), high pressure (~100MP), and the mesothermal temperatures (~300°C), similar to other Sn deposits predominantly composed of gaseous fluids of CO₂,N₂ and minor CH₄. According to these authors, the mineralising fluid was a primary magmatic (G4-granite) fluid. However, the stable isotope analysis of quartz crystals from Gifurwe and Nyakabingo tungsten belt indicates metamorphic conditions⁴².

In addition, the main cassiterite stage present in the greisens, based on petrographic observations was clearly not a primary magmatic liquid phase, but crystallized later during metasomatic alteration. These metasomatic fluids are generally interpreted as lower temperature fluids between 250°C and 450°C⁴³.

In Rutongo, the study of fluid inclusion microthermometry of quartz vein and cassiterite revealed the four types 1 to 4 (Table-3), and the total temperature of homogenisation was between 247°C and 341°C. So, the primary fluid inclusions in the quartz and cassiterite concentrated to H₂O-CO₂-CH₄-NaCl composition with the moderate salinity (2.9-14.2 % NaCl)⁴⁴.

Table-3: Total homogenisation temperatures and compositional data of fluid inclusions in quartz and cassiterite crystals from Sn mineralised quartz veins in the Rutongo area (after Günther). Both the calculated salinity given by Günther and the salinity recalculated by De clerq are given^{3,5}.

Type	Characteristics
Sn1A-C quartz and Cassiterite	<p>V_{gas phase}: 20-85%</p> <p>T_{hstot}: 247-341 °C</p> <p>Gas phase: Liquid CO₂+CH₄+</p> <p>Salinity: 6.6-17.6 ≠ 2.9-14.2 mass% NaCl</p> <p>Type: H₂O-CO₂-CH₄-(X)-NaCl-X</p>
Sn1D-E quartz and cassiterite	<p>V_{gas phase}: 15-30%</p> <p>T_{hstot}: 233-346 °C</p> <p>Gas phase: mainly gaseous CO₂+</p> <p>Salinity: 11.7-14.8 ≠ 4.6-12.1 mass% NaCl</p> <p>Type: H₂O-CO₂-(X)-CaCl₂-NaCl-X</p> <p>Solid inclusion: calcite,graphite</p>
Sn2 quartz and cassiterite	<p>V_{gas phase}: 5-15%</p> <p>T_{hstot}: 199-303 °C</p> <p>Salinity: 13.2-19.7 ≈ 11.8-18.9 mass% NaCl</p> <p>Type: H₂O-(CO₂-X)CaCl₂-NaCl</p> <p>Solid inclusion: calcite, muscovite?</p>
Sn3 quartz	<p>V_{gas phase}: 5%</p> <p>T_{hstot}: 80-257 °C</p> <p>Salinity: /</p> <p>Type: H₂O</p>

Conclusion

Niobium-Tantalum, Tin and Tungsten ore deposits are major mineralisations related to granite in Rwanda. G1-3 granites, which formed at 1380 ± 10 Ma, do not demonstrate any economically significant concentration of rare metals.

Conversely, the G4 granite generation, which formed at 986 ± 10 Ma, is related to rare metal ore deposits and hence called as fertile granite. Its emplacement was followed by pegmatite emplacement at 969 ± 8 Ma.

The so called fertile granites, or G4 granites are associated with the formation of rare metal mineralised pegmatites/quartz veins by hydrothermal movement. In Rwanda, niobium-tantalum (Nb-Ta), tin (Sn), and tungsten (W) mineralisations mainly occur in pegmatites and quartz veins.

The metals are present in various forms of mineralization; as primary mineralization, they are found in pegmatites, quartz veins, and greisens; but as secondary mineralization, they occur in alluvial or elluvial deposits. Ore mineral deposits are considered anomalies in the earth, and provide us with the clearest evidence of past events, such as the flow of solutions through fractures, faults, and porous rocks that dissolved, transported and concentrated elements of economic value.

The tin and tungsten-mineralised quartz veins at Rutongo and Nyakabingo both formed from fluids that had the same primary magmatic origin and a similar evolution. Dewaele et al.⁴⁴ determined the oxygen and hydrogen isotopic composition of tin-mineralised quartz veins from Rutongo (Rwanda) and interpreted their genesis based on related geological settings, comparable paragenesis, and fluid inclusion composition. The δ¹⁸O-δD values of the tin mineralising fluids also fall in the typical range for metamorphic water. As in the example of Gatumba (Rwanda), the Columbite-tantalite formed during the crystallization of pegmatite followed by intense alkali metasomatism or advanced argillic alteration i.e. a wide increase of albite to kaolin and white mica.

However, the major section of the cassiterite mineralisation is condensed in zones linked with phyllic alteration. Two models about origins of the mineralising fluids (magmatic hydrothermal and metamorphic fluids) in Rwanda were well documented by different authors, but the evolution of the mineralising fluid is still poorly known.

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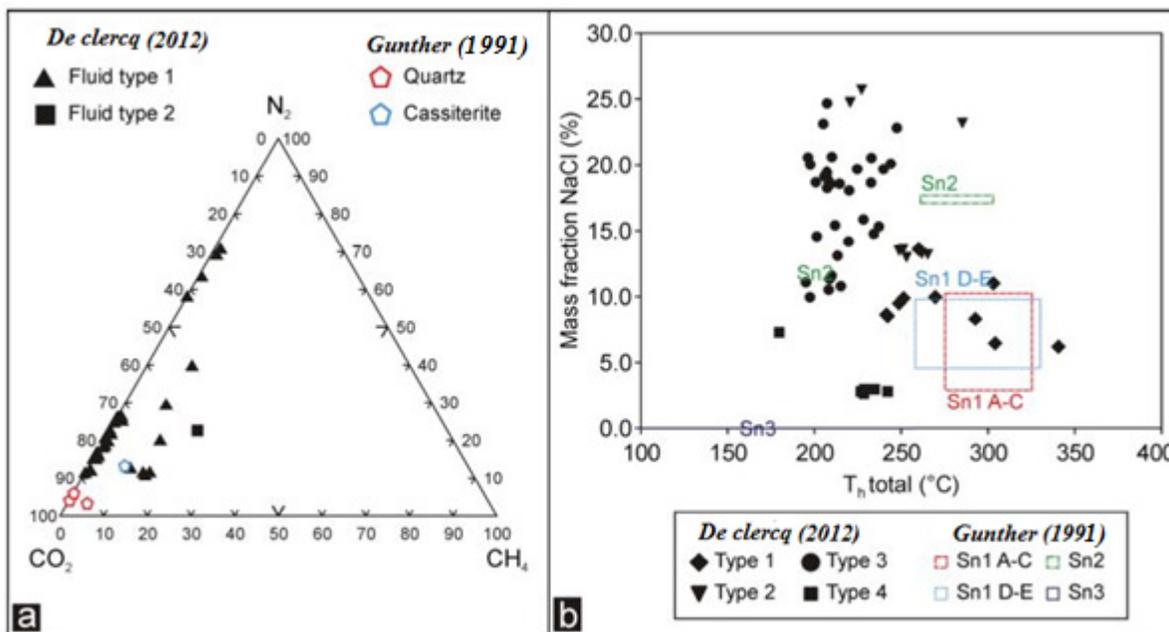


Figure-8: (a) CO₂-N₂-CH₄ ternary diagram showing the composition of the gas phase of fluid inclusions from quartz and cassiterite crystals in Sn mineralised veins from the Rutongo area by Günther and De clercq. (b) Scatter plot of T_{h,tot} versus Salinity of fluids from Sn-mineralised quartz veins from the Rutongo area in Rwanda, by Günther and De clercq^{3,5}.

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