

## A review of origins and occurrences of Niobium-Tantalum, Tin and Tungsten mineralization in Rwanda

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**Abstract:** The mesoproterozoic Kibara belt in central Africa has the subdivision of the two mobile belts which are the Karagwe -Ankole belt (KAB) and the Kibara belt (KIB), separated by Rusizi terrane. 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 Karagwe-Ankole Belt 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  $986 \pm 10$  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 (~100MPa), 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, however, some authors postulated the hydrothermal fluid was the primary magmatic(G4 granite) fluid. The objective of this paper is necessarily brief owing to the limited work in the region.

**Keywords:** Karagwe-Ankole belt, Kibara belt, Cassiterite, Columbite-Tantalite, Wolframite, Rwanda

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### I. Introduction

#### 1.1 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  $S2^0 00.00'$  latitude and  $E030^0 00.00'$  longitude. The total land area is about 24,950 km<sup>2</sup>, and inland lakes cover about 1390 km<sup>2</sup> (Safari, 2010). Rwanda is positioned in the southwestern part of the northeastern Kibara belt(KIB), central 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) mineralization (Pohl, 1994).

In Rwanda, the Nb-Ta, Sn, W mineralisations mainly occur in pegmatite and quartz vein (Pohl, 1994; Pohl&Günther,1991; Dewaele et al.,2007). The metals exist in various forms of mineralisation. They occur as primary mineralisation in the form of pegmatite , quartz vein, and greisen, but also as secondary mineralisation in alluvial or alluvial deposits. The granites subdivided into types (G1 to G4) which intruded the Kibara belt. The U-Pb SHRIMP dating demonstrated that G1 to G3 granites were formed at  $1380 \pm 10$  Ma, (Cahen & Ledent, 1979; Cahen et al., 1984; Klerckx et al., 1984, 1987). Conversely, the G4 granite (or the so-called ‘fertile granite’) formed at  $986 \pm 10$  Ma (Tack et al., 2006). The mineralised pegmatites and quartz veins are considered to be related to the G4-granite.

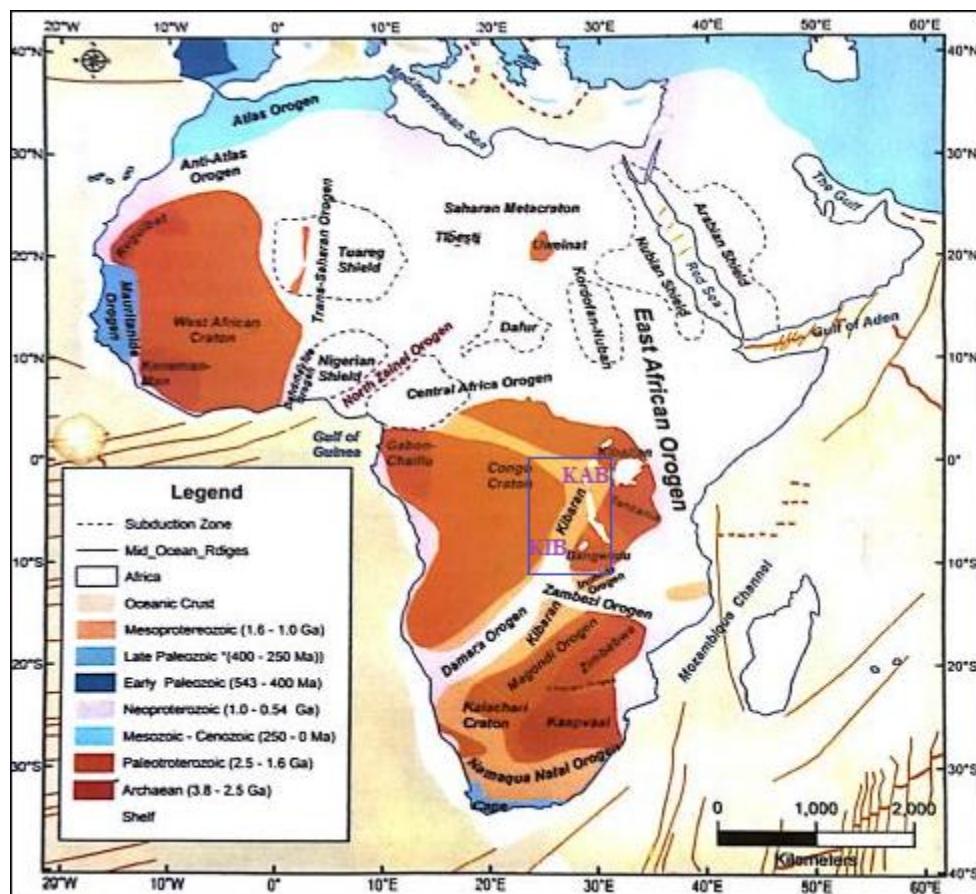
#### 1.2 Geological settings

##### 1.2.1 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 Bangweulu block to the south, and the archaean-palaeoproterozoic congo craton to the west and north (Tack et al., 2010). 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  $1380 \pm 10$  Ma. The crystallization of these granites did not result in concentrations of economically valuable rare metal deposits.

Later, at  $986 \pm 10$  Ma, the granite generation known as G4 granite (fertile granite, rare metal granite or tin granite) resulted in the emplacement of rare metals deposits. (Tack *et al.*, 2006, 2010; Dewaele *et al.*, 2011).

The Karagwe-Ankole belt and Kibara belt comprise a fluctuate sequence of pelitic and carbonate sediments, minor volcanics and dolerite. The geology, structural settings, stratigraphy, metamorphism of the Kibara orogeny are well-documented by previous authors (De Clercq, 2012; Dewaele *et al.*, 2011; Tack *et al.*, 2006; Muchez, Hulsbosch and Dewaele, 2014; Hulsbosch *et al.*, 2017; Pohl, 1987, 1994). The Kibara belt is known for the emplacement of the rare metals niobium-tantalum(Nb-Ta), tin(Sn), tungsten(W), and gold(Au) which occurred at  $970 \pm 10$  Ma. The primary emplacement of Sn was observed in hydroxyl bearing phases of granites and mineralization hosted in the pegmatites and/or quartz veins. (Bell, 1993; Dewaele *et al.*, 2011; Pohl, 1994; Romer and Lehmann, 1995).

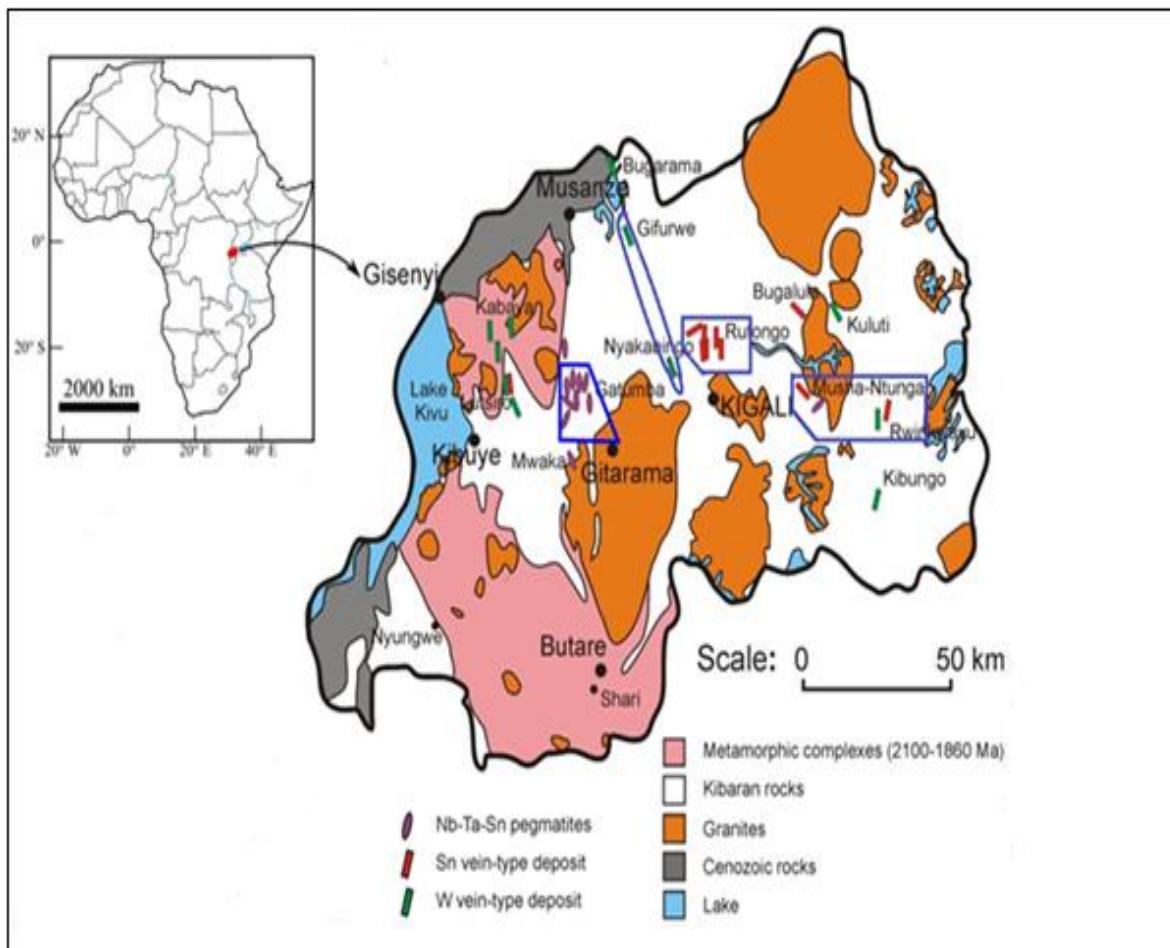


**Fig:1:** Simplified map of regional geological setting of Karagwe ankole belt and kibara belt (modified after Brinckmann *et al.* (2001)

### 1.2.2 Geology of Rwanda

Rwanda is underlain by the rocks of the Kibara Orogeny which consists predominantly of the basement and mezoproterozoic rocks (ca. 1.6-1.0 Ga), and have been intruded by rocks of bimodal magmatic event (granitic and mafic rocks) (Tack *et al.*, 2010). The Kibara belt is well-known for its mineralization 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 granite (BRGM., 1987&Dewaele *et al.*, 2010). 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 Northwestern Birunga mountains. These Meso-Proterozoic 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 (Dewaele *et al.*, 2010; Fernandez-Alonso *et al.*, 2012).

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 Zones (Fernandez-Alonso & Theunissen, 1998).

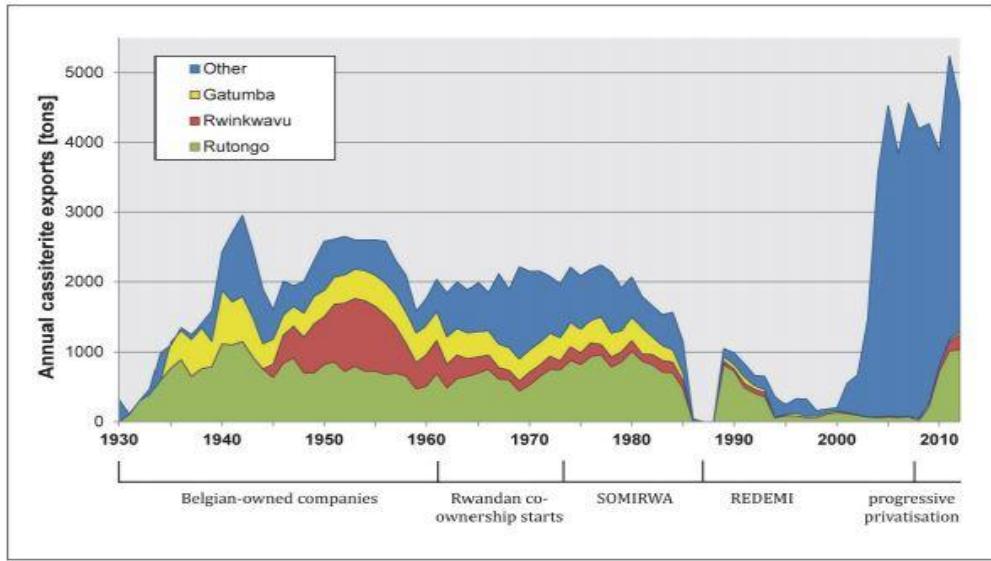


**Fig 2:** Simplified geological map of Rwanda with the indication of the most important granite-related ore deposits. Modified after Fernandez-Alonso et al.(2007)

Rwanda hosts a large number of minerals, the major commodities, economically mined now and in the past, being cassiterite ( $\text{SnO}_2$ ), niobio-tantalite also called colombo-tantalite or coltan ( $(\text{Nb}, \text{Ta})_2\text{O}_5$ ) and Wolframite ( $(\text{Fe}, \text{Mn})\text{WO}_4$ ). 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 ( $(\text{Li}, \text{Na})\text{AlPO}_4(\text{F}, \text{OH})$ ), monazite( $\text{Ce}, \text{La}, \text{Nd}, \text{Th}\text{PO}_4$ ), tourmaline ( $(\text{Ca}, \text{K}, \text{Na})(\text{Al}, \text{Fe}, \text{Li}, \text{Mg}, \text{Mn})_3 (\text{Al}, \text{Cr}, \text{Fe}, \text{V})_6 (\text{BO}_3)(\text{Si}, \text{Al}, \text{B})_6\text{O}_{18}$ ) and gold (Au) (Bertossa, 1965, 1967-1968, Minirena unpublished, 2012).

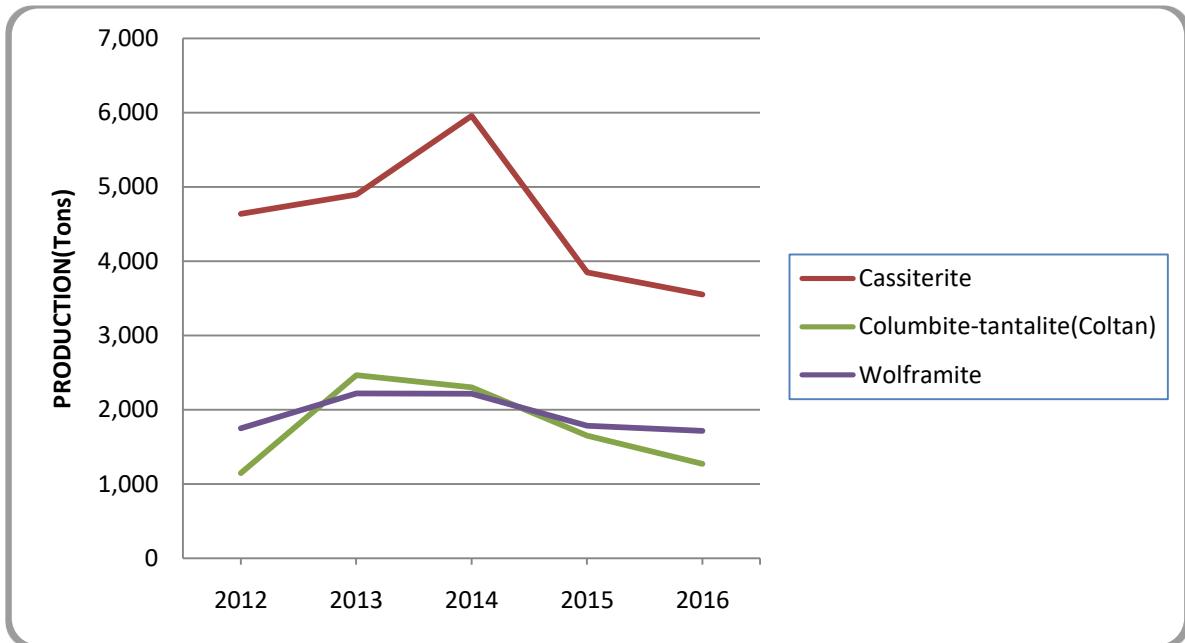
Ore mineral	Occurrence	Typical local Mine
Columbite-tantalite	mineralization occurs in pegmatite	Gatumba
Cassiterite	mineralization occurs in quartz vein, schists, greisen and pegmatite	Rutongo, Rwindwau, Musha-Ntunga, Gatumba
Tungsten	mineralization occurs in quartz vein	Nyakabingo, Gifurwe and Bugarama

**Table1 :** The representative mines for ore mineral deposits and occurrences in Rwanda (Minirena&BRGM., 1987) They occur as primary mineralization in the pegmatites, greisens and quartz veins but also during the secondary mineralization in alluvial or elluvial deposits(Dewaele et al., 2011).



**Fig:3:** Historic production/exports of cassiterite concentrate from Rwanda ( Schütte, 2014).

Coltan found associated with cassiterite mineralisation in pegmatite bodies, due to growing demand for the tantalum metal in current technological industrialisation, the coltan( tantalum ore) has only become economically relevant in more recent times, eventually surpassing cassiterite (tin ore) as the major export earner in 2012.



**Fig:4:** Historical production of 3Ts minerals in 5 past years, Rwanda(BNR.,2017)

Fig:4 indicated 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(BNR,2014). Subsequently, The production has dramatically decreased due to the prices 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 growth.

## II. Research findings

### 2.1. 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 lensoid 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 meters (Hulsbosch *et al.*, 2017). For most parts of the area, these pegmatites are concordant with the regional foliation but some discordant and sub-horizontal bodies are also noticed.

Most 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 aggregates (Ahmad, 1995). Other nonsilicates 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 granitoids (Ahmad, 1995; Dewaele, Hulsbosch, *et al.*, 2016b).

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.



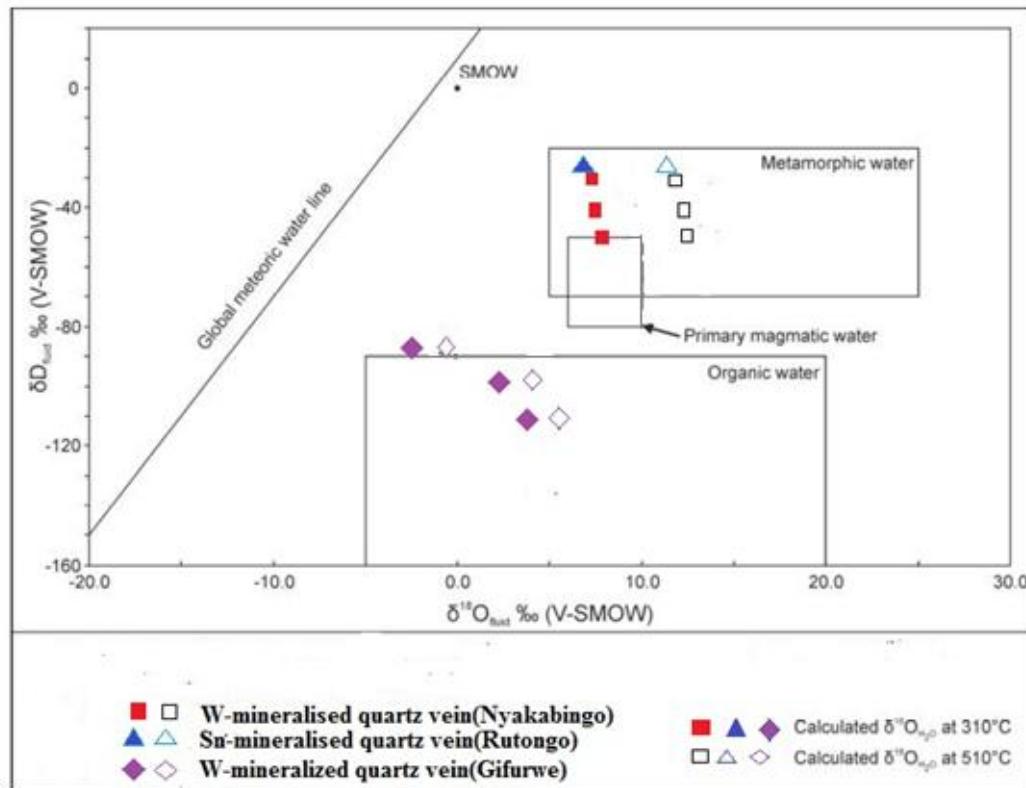
**Fig: 5:** A. Intrusion of advanced argillic alteration of pegmatites with concentrates of coltan at Ntunga. B. Quartz vein mineralized in cassiterite and hosted in schist at Rwinkwau. C. Quartz vein mineralized in tungsten and hosted in black shale at Nyakabingo

### 2.2. Stable isotopes in ore deposits

The D/H and  $^{18}\text{O}/^{16}\text{O}$  analyses are valuable applications to determine the history and origin of  $\text{H}_2\text{O}$  in the hydrothermal fluids. The composition of hydrogen and oxygen isotopes was determined using the standard analytical techniques of Clayton & Mayeda, (1963).

To look into the origins of hydrothermal fluid of water in the areas of primary magmatic, metamorphic , meteoric water, organic and kaolinite lines (Taylor, 1979). Stable isotope studies therefore suggest a model in which Sn 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 cassiterite was precipitated in structurally controlled emplacements, along with the alteration of the host-rocks.

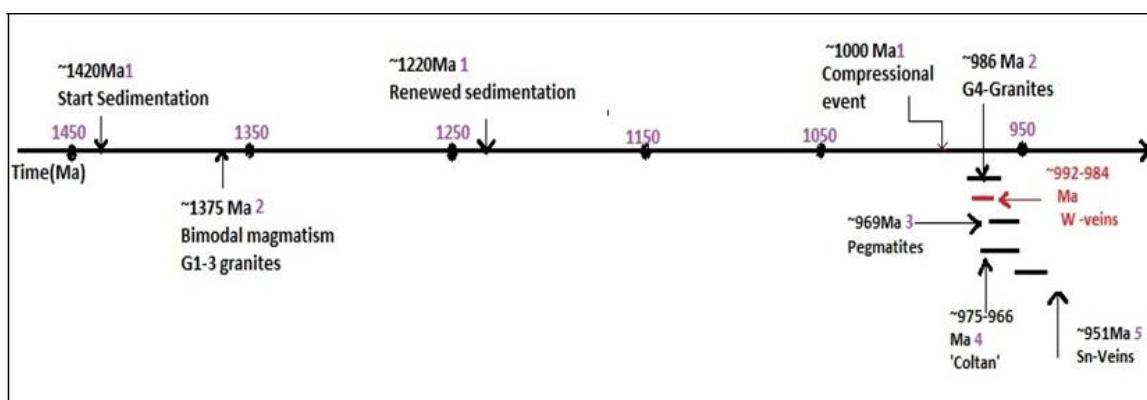
Columbite-tantalite formed during the crystallisation of the pegmatites followed by intense alkali metasomatism which advanced the formation of albite (albitisation) and white mica.(Stijn Dewaele *et al.*, 2009,2011). The tungsten was postulated to have originated from a fluid mostly brought out by metamorphic processes , however, there is another model that suggests the fluids have a magmatic signature (de Clercq *et al.*, 2008; Dewaele *et al.*, 2010).



**Fig:6:**  $\delta^{18}\text{O}$ - $\delta\text{D}$  isotopic plot for the calculated fluid composition of the mineralised quartz veins of Nyakabingo, Gifurwe and Rutongo ( modified after Dewaele et al., 2007, De Clercq et al., 2008).

### 2.3. 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, we can resolve a time simply by determining how much of the nuclide has decayed. The young Rb-Sr and  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  ages for granites, pegmatites and veins have geochronological characterisation related to the crustal formation of Africa ( $\leq 960\text{ Ma}$ ) (Cahen, 1982; Ikingura, J. R, 1992 ; Romer and Lehmann, 1995). 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 (Tack et al., 2010; Dewaele et al., 2011; Ngaruye, 2011; Dewaele, De Clercq, et al., 2016b)



**Fig:7:** Radiogenic isotopic data in Rwanda (integrated after Fernandez-Alonso et al.,2012,Tack et al.,2010, Brinckmann&Lehmann,1983,Dewaele et al.,2011,Brinckmann et al.,1994).

### 2.4. Fluid inclusion microthermometry

By focusing on fluid inclusions captured within hydrothermal veins, we have a better picture than was previous 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 new techniques of microthermometry and bulk chemical analysis. (Sorby,1858)

In this, idea was a way by adding news on 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 Sorby.

In his usual paper Sorby, (1858) outlined samples from ore deposits having fluid inclusions and drew conclusions concerning ore formation that remained unpopular for many years.

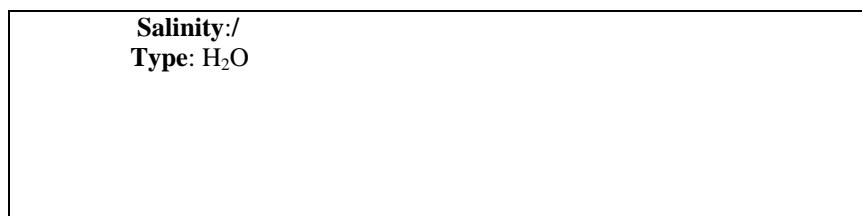
Tin and tungsten mineralised quartz veins at Rutongo and Nyakabingo respectively formed from fluids that had the same primary magmatic origin and a similar evolution (Pohl&Günther 1991). The gas composition of nitrogen and methane gases ( $N_2$  and  $CH_4$ ) and the temperature of the tungsten (W) mineralising fluid (~300°C) relatively indicate metamorphic conditions (Roedder , 1984; Kilius and Konnerup-Madsen, 1997; O'Reilly, Gallagher and Feely, 1997; Huff and Nabelek, 2007). Based on a similar geological setting and a comparable paragenesis and fluid inclusion composition. Dewaele et al. (2007) determined the oxygen and hydrogen isotopic composition of tin-mineralised quartz veins from Rutongo (Rwanda). The  $\delta^{18}O$ - $\delta D$  values of the tin mineralising fluids also fall in the field typical for metamorphic water(Dewaele, et al 2007, De clericq et al., 2008) .

Pohl & Günther (1991) and Pohl (1994) postulated that the tungsten mineralisation at Nyakabingo formed from an  $H_2O-CO_2-CH_4-N_2-NaCl$  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  $CO_2, N_2$  and minor  $CH_4$  . According to these authors, the mineralising fluid was a primary magmatic (G4-granite) fluid.

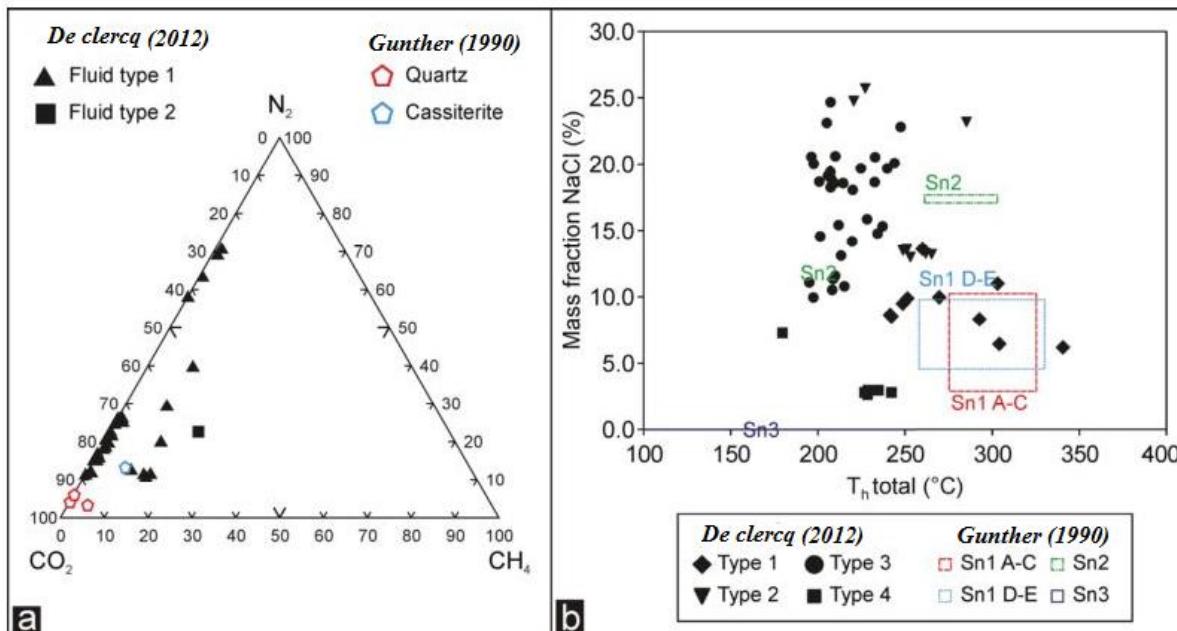
However, the stable isotope analysis of quartz crystals from Gifurwe and Nyakabingo tungsten belt indicates metamorphic conditions.(Dewaele, De Clercq, et al., 2016a). In addition, the main cassiterite stage present in the greisens based on petrographic observations was clearly not a primary magmatic liquid phase, but crystallized later during metasomatic alteration. These metasomatic fluids are generally interpreted as lower temperature fluids between 250°C and 450 °C. (Kontak and Kyser, 2009)

In Rutongo (Table1) , the study of fluid inclusion microthermometry of quartz vein and cassiterite revealed the four types(type 1 to type 4) , and the total temperature of homogenisation was between 247 and 341 °C. So, the primary fluid inclusions in the quartz and cassiterite concentrated to  $H_2O-CO_2-CH_4-NaCl$  composition with the moderate salinity(2.9-14.2 %Nacl) (De clericq 2012, W.Pohl and M.A Günther., 1990).

Type	Characteristics
Sn1A-C quartz and Cassiterite	<p><math>V_{gas\ phase}</math>: 20-85%</p> <p><math>T_{h\cdot tot}</math> : 247-341 °C</p> <p><b>Gas phase:</b> Liquid <math>CO_2+CH_4+?</math></p> <p><b>Salinity:</b> <b>6.6-17.6</b> ≠ 2.9-14.2 mass% NaCl</p> <p><b>Type:</b> <math>H_2O-CO_2-CH_4-(X)-NaCl-X</math></p>
Sn1D-E quartz and cassiterite	<p><math>V_{gas\ phase}</math>: 15-30%</p> <p><math>T_{h\cdot tot}</math> : 233-346 °C</p> <p><b>Gas phase:</b> mainly gaseous <math>CO_2+?</math></p> <p><b>Salinity:</b> <b>11.7-14.8</b> ≠ 4.6-12.1 mass% NaCl</p> <p><b>Type:</b> <math>H_2O-CO_2-(X)-CaCl_2-NaCl-X</math></p> <p><b>Solid inclusion:</b>calcite,graphite</p>
Sn2 quartz and cassiterite	<p><math>V_{gas\ phase}</math>: 5-15%</p> <p><math>T_{h\cdot tot}</math> : 199-303 °C</p> <p><b>Salinity:</b> <b>13.2-19.7</b> ≈ 11.8-18.9 mass% NaCl</p> <p><b>Type:</b> <math>H_2O-(CO_2-X)CaCl_2-NaCl</math></p> <p><b>Solid inclusion:</b> calcite, muscovite?</p>
Sn3	<p>quartz</p> <p><math>V_{gas\ phase}</math>: 5%</p> <p><math>T_{h\cdot tot}</math> : 80-257 °C</p>



**Table 2:** 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.,1991) . Both the calculated salinity given by Günther (1991) and the salinity recalculated by De clercq (2012) are given



**Fig:8:** (a)  $\text{CO}_2\text{-N}_2\text{-CH}_4$  Ternary diagram showing the composition of the gas phase of liquid inclusions from quartz and cassiterite crystals in Sn mineralised veins from the Rutongo area by Günther (1991) and De clercq (2012). (b) Scatter plot of  $T_{\text{h},\text{tot}}$  versus Salinity of fluids from Sn-mineralised quartz veins from the Rutongo area in Rwanda, by Günther (1991) and De clercq (2012).

### III. Conclusion

The granite generations (Gs) of the Kibara belt (KIB) and the Karagwe-Ankole belt (KAB) have subdivided into 3 types (G1-3) and emplaced into the Palaeo-Mesoproterozoic rocks at  $1380 \pm 10$  Ma. These granites do not demonstrate any economically significant concentration of rare metals. Conversely, the G4 granite generation, or fertile granites were emplaced at  $986 \pm 10$  Ma, and followed by pegmatites at  $969 \pm 8$  Ma.

The so called fertile granites, or G4 granites are associated with the formation of rare metal mineralised pegmatites by hydrothermal movement driven by the east African orogeny of pan-African mineralization. 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. In 2010, Dewaele et al. determined the oxygen and hydrogen isotopic composition of tin-mineralised quartz veins from Rutongo (Rwanda) based on related geological settings, comparable paragenesis, and fluid inclusion composition. The  $\delta^{18}\text{O}$ - $\delta\text{D}$  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 phyllitic alteration. Two

models about origins of the mineralising fluids(magmatic hydrothermal and metamorphic fluids) were well documented by different authors, but the evolution of the mineralising fluid is currently unknown.

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