



Li, Cs, Ta and Rb enrichment in metasedimentary intrusive body: a marker of rare metal potential mineralisation of pegmatite in origin, Musha-Ntunga area, Eastern Province, Rwanda

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Abstract

The study aims to geochemically characterize the pegmatite and elucidate the current processes occurred after the primary emplacement of ores in Musha-Ntunga area, Rwanda. The rare metal pegmatites intruded the metasedimentary rocks dominated by schists in Musha formation. The laboratory analyses of petrography and geochemistry were used. The petrographic analysis investigated mineral assemblages, and whole rock analysis for major elements by ICP AES and trace elements by ICP MS. Petrographic studies revealed the mineral assemblages include plagioclase, mica, tourmaline and muscovite. The results of geochemical analysis revealed medium to high silica content (in wt%) ranges from 67.08-70.66, the Al_2O_3 ranges 26.6-28.7 (in wt%), Na_2O ~0.06 (in wt%) and CaO varies between (0.06-0.07) (in wt%) for the intrusive body of pegmatite in origin intruded metasedimentary rocks. These altered intrusion of pegmatite in origin are enriched in trace elements of Rb (~715.18ppm), Cs (~34.43ppm), Li (~148.33ppm), W (~492.ppm), and the rare metals Nb (1154.16ppm~), Ta (>2500 ppm), and Sn (>10.000 ppm), the enrichment of those elements has commonly indicated a marker of a magmatic-hydrothermal alteration of pegmatite intrusion. The molar $Na_2O-Al_2O_3-K_2O$ plot revealed that the pegmatites evolved from the peraluminous granite (or S-type). REE abundance in whole rock pegmatitic intrusive body was ΣREE (~120.65ppm) signifying the moderate to high form of enrichment. The different plots such as K/Rb vs. Rb, Ta vs. Ga and K_2O vs. Rb were plotted to appraise the mineralisation of pegmatites, with the K/Rb values (~ 20.0) less than 100 which are commonly accepted for mineralisation.

Keywords: Trace elements, mineralisation, altered pegmatitic intrusion, Musha-Ntunga.

Introduction

Pegmatitic rocks are defined as coarse-grained and crystalline rocks that contain giant crystals of feldspar, quartz or mica that compositionally make these rocks contrast to similar felsic lithology of granites that normally lie in their nearby vicinity¹. They are formed from felsic magma in which the residual melts can be supplemented in trace elements and heavy metals. Pegmatites often contain some of the least abundant elements and may contain semi-precious gemstones such as beryl, topaz and tourmaline. The pegmatites evolved from crustal granites in orogenic belts are characterized by high enrichment in incompatible elements such as Rb, Cs, Li, Be and Sn. They are found associated with mineralisation of Nb-Ta, Sn, W elements. The petrogenesis of pegmatites can be studied from their composition that indicates magmatic and post magmatic events responsible for pegmatite evolution².

Various studies have been carried out on the pegmatites in Rwanda by different authors²⁻⁷. The pegmatites are mineralised in rare metals from the parental magmas fractionated and

evolved with high incompatible trace elements Sn, Li, Rb and F and low Ba, Sr and Zr⁸.

In the eastern part of Rwanda, pegmatites are hosted within varieties of rocks of metasedimentary and Igneous origin where they intruded the older lithologies discordantly⁹ but there is rareness of information on pegmatites in Musha-Ntunga area. This work aims to geochemically characterize the pegmatite in relation to Tin and Tantalum mineralisation, and elucidate the processes occurred after primary emplacement of ore deposits by using the whole rock geochemical analysis and petrography of pegmatite and associated rocks from Musha-Ntunga area, eastern province, Rwanda.

Regional geological settings: The centre east Africa has been recognised for the Kibaran belt and Karagwe Ankole Belt formed between the pre-mesoproterozoic domains such as the archaean Tanzania craton to the east, palaeoproterozoic Congo craton to north and west, and block of Bangweulu to the south (Figure-1). The Karagwe Ankole Belt (KAB) goes across Burundi, Rwanda, North west Tanzania, south west Uganda up

to the Kivu-Maniema region in Democratic Republic of Congo (DRC). The Kibaran Belt (KIB) extends to south west in the Katanga region of DRC, which includes the kibaran mountainous area near Mitwaba town. KAB and KIB which were separated by paleoproterozoic Rusizian-Ubendian belt host two types of granites operated at 1375 ± 10 Ma and youngest granite dated at 986 ± 10 Ma which was related to ore deposits of typical Nb-Ta-Sn-W metal association¹⁸. The metals are mainly occurred in pegmatites or quartz veins. Pegmatites can be found largely mineralized in Nb-Ta/Sn minerals namely Columbite-Tantalite/Cassiterite while mineralized quartz veins found mineralised in Sn/W minerals of Cassiterite/wolframite. The primary mineralisation occurs in quartz veins and pegmatites, but also the secondary mineralisation in alluvial/eluvial deposits.

Geological settings of study area: The Musha-Ntunga (MN) area is within the Mesoproterozoic KAB, MN is positioned in eastern province of Rwanda. There were four lithostratigraphic formations found in the area (to younging order): Nyabugogo, Musha, Nduba, and Bulimbi formations.

The Nyabugogo, Musha and Nduba are formations of Gikoro Group in the Akanyaru Supergroup, while Bulimbi formation is found in Pindura Group¹². These formations of Pindura and Gikoro groups which are mainly composed of the quartzites and schists with sandstones near the surface, are deposited between 1375 and 1420 Ma^{13,14}. The Nyabugogo Formation (Ng) is schist and metasandstones with a minimum thickness of 700m. The formation of Musha (Mh) consists mainly of siltstones coloured pale-grey and sandstones covering clay drapes. The

MN area shows the intercalations of alternative metapelite, metasandstone and metasilstone.

The 300 m is the estimated thickness of the Musha Formation. The thickness scale between 150-200 m is estimated for Nduba Formation (Nd) dominantly has metasandstones and quartzites. Musha Formation is main outcrop of the study area. The Bulimbi Formation is found rich in pyrite, graphite black shales intercalating metasandstones and metapelites⁶.

The parental granite has been found close to the pegmatites and quartz veins in the study area. The Tin mineralised quartz veins are in continuous series with the pegmatites zoned to quartz rich pegmatites¹⁵. Nduba quartzites contain the quartz veins, but more often schists in Musha Formation¹⁵. After Slatkine A.¹⁶ remarked that the mines of Musha-Ntunga are rich in quartz-muscovite cassiterite.

The young granites were related to the types of ore deposits. The pegmatites strongly enriched in metals and associated with parental granites were classified as lithium-cesium-tantalum (LCT) pegmatites of the class of rare elements after Černý P.¹⁷ and can be mineralized in Nb-Ta and Sn^{7,18}.

Hydrothermal quartz veins, in some cases spatially and temporally associated with the granitic pegmatites can hold Cassiterite or W-bearing minerals^{7,18}. The origin of the W-bearing minerals that occurs in quartz veins sets in hydrothermal fluids that exsolved early from granitic-pegmatitic melts and hosted in rocks of high degree of carboniferous compounds like black shale¹⁹.

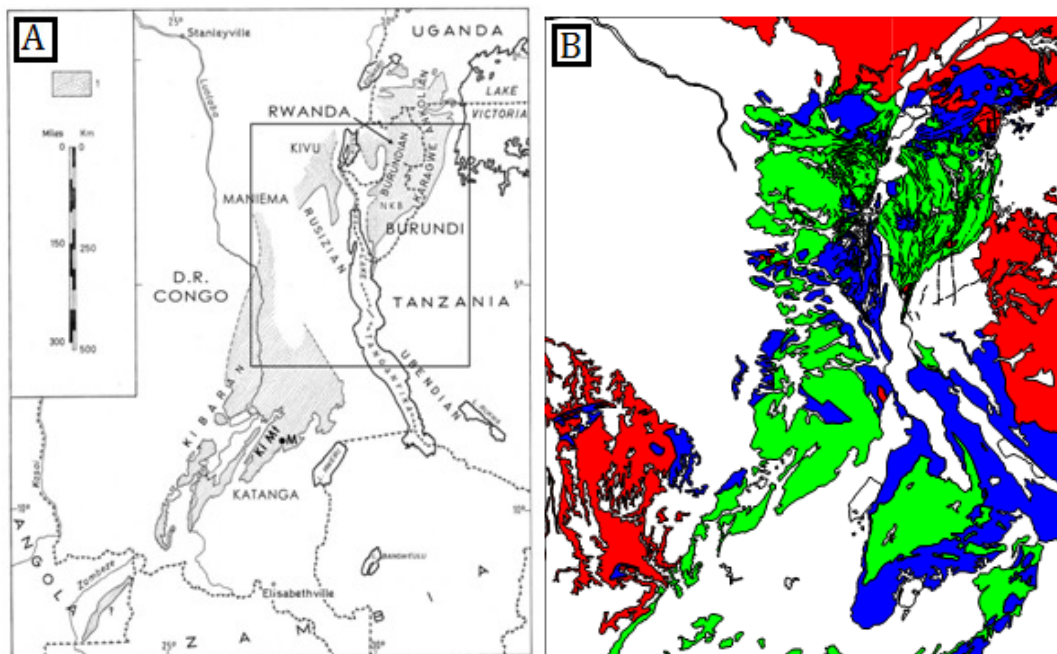


Figure-1: Regional geological maps showing the 2 segments separated by Palaeoproterozoic Rusizian-Ubendian basement: A¹⁰, and B¹¹: Green: Mesoproterozoic, Blue: Paleoproterozoic, Red: Archaen.

The pegmatites found in the Musha Ntungga (MN) area consist predominantly of kaolinized feldspar with quartz and muscovite. Muscovite typically has a transparent white to greenish colour and can be found dispersed across the pegmatite or at the margins of host rock-Pegmatite. The morphologies of hydrothermal quartz observed in Musha-Ntungga are transparent white quartz, blocky quartz and saccharoidal quartz. Muscovite mainly occurs at the edge and the centre of the vein. Black tourmaline observed at the border of the quartz veins or spread to the zone of quartz veins adjacent to host rock. In the MN area, the quartz veins were observed to be barren or mineralized in Sn. W-mineralization is not present. The pegmatites and quartz veins were emplaced, at least partially, contemporaneously as is evidenced by cross-cutting relationships²⁰. The mining activities in the Musha-Ntungga area initiated during colonial time in the 1930s. The Musha-Ntungga area has potentially produced coltan and Cassiterite concentrates around 1934 and 1985. The Niobium-Tantalum and Tin rich area of MN is nowadays licenced mined by PIRAN Resources Ltd.

Materials and methods

Geological mapping, field observation and rock sampling in Musha-Ntungga area has been carried out to describe the petrography and whole rock pegmatites to understand the origin of mineralisation and processes occurred after the primary emplacement of ore deposits. The procedure for making transparent thin sections is as follows: The rock samples were cut into small chips, thin down and mounted on glass plate, this was placed on a hot plate to dry. Araldite was used to glue the dry chip on a glass and left overnight to dry, further reduction in size was achieved using silicon carbide to produce a very thin

layer that was covered with Canada basalm and placed on hot plate to dry. Excess Canada basalm was washed off with acetone to obtain a clean thin section. 6 thin sections were prepared and chosen for pegmatite bodies and associated rocks (granites, schists) for petrographic investigation at University of Ibadan, Ibadan, Nigeria. The thin sections from the samples were observed with a transmitted polarized light microscope equipped with a Nikon D3400 camera which allowed taking pictures of the samples. The images were obtained using total magnification equals to 100x.

Over 9 rock samples of pegmatites and some associated rocks have been set for 50g of pulverised powers at 85% passing 75 microns for whole rock geochemical analysis to determine major and trace elements. The Inductively coupled Plasma-Mass Spectrometry (ICP-MS) and Atomic Emission Spectrometry (ICP-AES) were used to analyse the sample with Lithium metaborate (LiBO₂) fusion and base metals by 4 acid digestion for base metal elements that includes Li element as one of the elements of interests in our study. The Lab was the ALS, Johannesburg, S.A. A prepared sample of 0.100gr added to LiBO₂ flux, mixed well and put at 1000⁰C in furnace. Then the melts was cooled and dissolved in 100mL of 4% nitric acid and 2% Hydrochloric acid solution. The extracted ions by pinhole-sized orifice were focused with an ions lens into a mass spectrometer. The 7700x Agilent ICP-MS analysed samples for 30 trace elements include rare earth elements (Table-2 and 3). Then, the ICP-AES analysed same solution for major oxides. The house standard and references were used for calibration. Some plots of geochemical data were analysed by using the excel office and GCD kit tool.

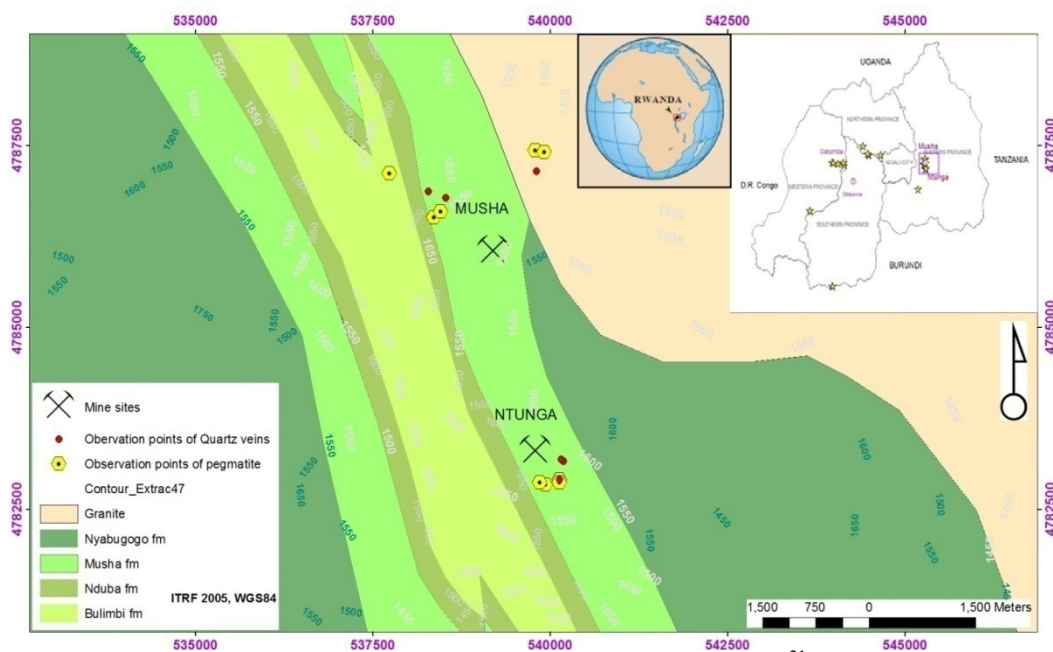


Figure-2: Geological map of Musha Ntungga area indicates the four formations after²¹ with the integration of the observed Pegmatites and Quartz veins.

Results and discussion

Field observation: The field work and observation were carried out and revealed that the outcrops with exposures of white intrusion of pegmatite in origin in metasedimentary rocks which can be observed in Musha-Ntungwa area. The granite in Musha-Ntungwa (MN) has been stated as the parental granite for the mineralisation of pegmatites in the MN area as documented in other parts of Rwanda such as Gitarama granite. The observation revealed the white intrusion in the Musha-Ntungwa (MN) area which consist predominantly of kaolinized feldspar with muscovite and minor quartz. Muscovite typically has a transparent white to greenish colour and disseminated in pegmatite or condensed to the margins between metas and stone-siltstone host rock and Pegmatite. The main hydrothermal quartz morphologies observed were pale white and fine crystalline quartz, saccharoidal quartz, clear and blocky quartz. Muscovite occurs in different positions to the quartz veins include margins and the centre of the vein. The accessory mineral found was Black tourmaline often occurs, mostly at the border of quartz veins or disseminated in the host rock adjacent to the veins. In the MN area, the quartz veins were observed to

be barren or mineralized in Sn. W-mineralization is not present. The altered intrusion of pegmatite (transformed) observed to have eye spots of mineralisation in Nb-Ta, Sn (Figure-3C). The quartz veins were emplaced, at least partially, contemporaneously as is evidenced by cross-cutting relationships²⁰.

Petrography: The thin sections from the samples were observed with a transmitted polarized light microscope equipped with a Nikon D3400 camera which allowed taking pictures of the samples. The images were obtained using total magnification between 40x and 100x. The mineral assemblage identified were quartz, Feldspar, Muscovite, Tourmaline (Figure-5).

The pegmatitic intrusion in metasedimentary rocks in MN were petrographically composed of Muscovite, Albite and Kaolin which indicate the processes of muscovitization, albitisation and kaolinisation that occurred. These are ones of the paragenetic sequences of Niobo-Tantalum and Tin mineralization in the Musha-Ntungwa area⁶.

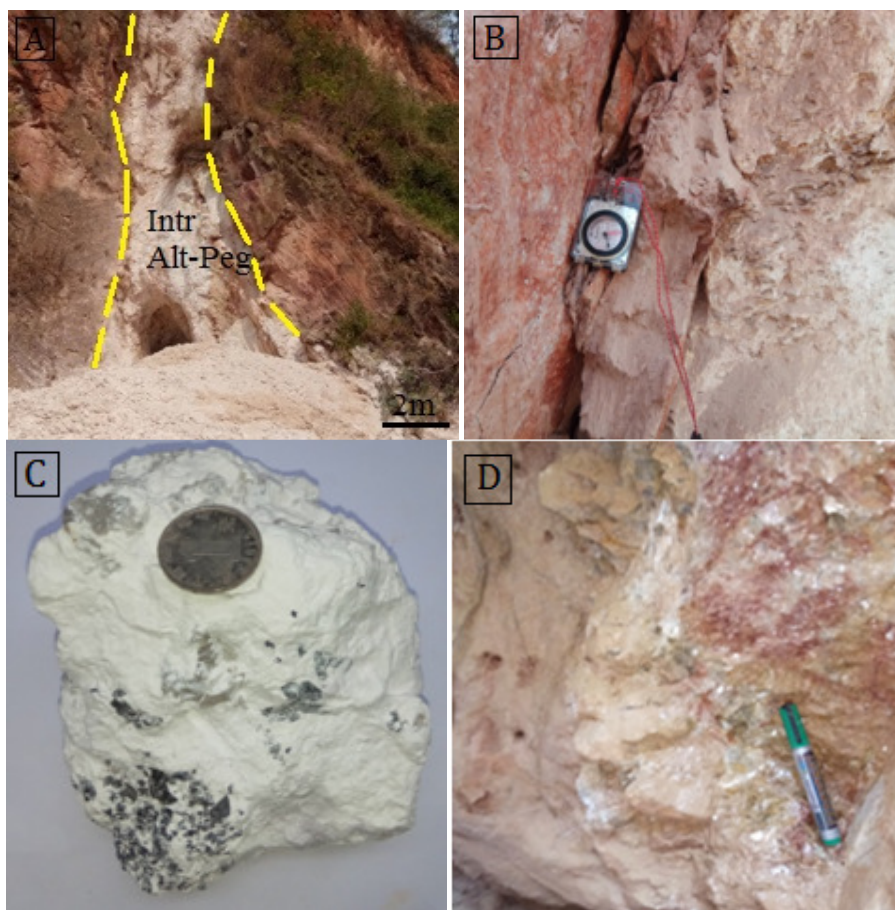


Figure-3: A: The Intrusion of altered pegmatitic intrusion (Int Alt-Peg) in metasedimentary rock, B: The contact zone between kaolinised pegmatitic intrusion and metasedimentary host rock(compass for scale), C: Hand specimen of transformed pegmatite with kaolin observed to have eye spots of Nb-Ta andSn potential mineralisation(coin for scale). D: The quartz vein with white to green muscovites observed to be found mineralised in Sn (marker for scale).

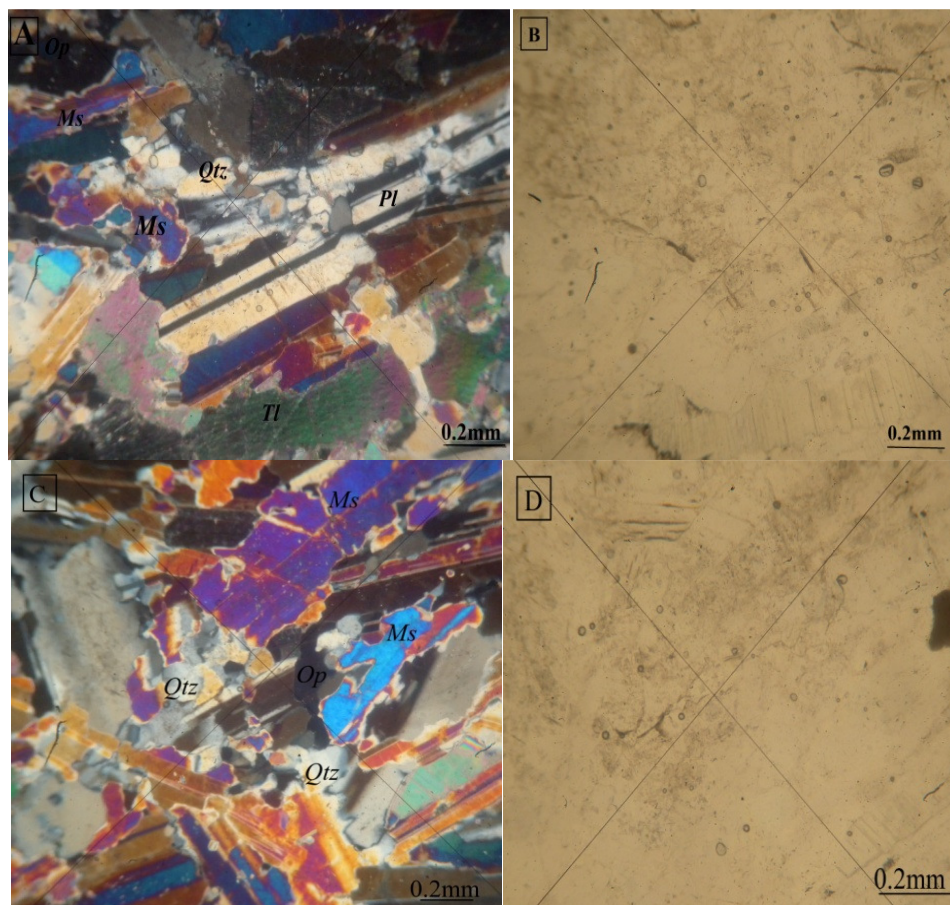


Figure-4: Thin section photomicrographs. A: XPL, Presence of plagioclase, Quartz, Tourmaline, Muscovite and opaque mineral. B: PPL C: XPL, Quartz, Muscovite, and opaque mineral. D: PPL. *Pl*: Plagioclase, *Qtz*: Quartz, *Ms*: Muscovite, *Op*: Opaque mineral, *Tl*:Tourmaline XPL: crossed polarised light, PPL: Plane polarised light.

Whole rock composition: Major elements: The whole rock composition of the altered pegmatite from Musha-Ntungwa area shows the moderate concentrations (in wt%) of SiO₂ (~69), medium to high concentration of Al₂O₃ (26.6-28.7), low concentration of Na₂O(0.06) and CaO(0.06-0.07) (Table-1). The Musha-Ntungwa pegmatites have low concentration in wt% K₂O (0.99-1.03), with the average of 2.02 wt%, high concentration of silica linked to granite differentiation and low-K values indicate the origin from a tholeiitic magma. The molar Na₂O-Al₂O₃-K₂O plot revealed that the pegmatites evolved from the peraluminous granites (or S-type).

Trace elements: The K/Rb versus Rb plot indicates the pegmatitic trend for the samples (Figure-6). This indicates highly differentiated pegmatites with Rb(690-751) ppm averaged at 715.18ppm of enrichment and depletion of Ba (91.6ppm-102.0ppm) averaged at 95.61ppm and Sr (22.0ppm-25.5ppm) with the average equals to 23.33ppm (Table-2).

The K/Rb values range approximated at the average of 11.8 (Table-4), thus shows the high content of Rubidium for Tin pegmatites. The K/Rb ratios less than 100ppm usually indicate

mineralization²². Further economic mineralization of these intrusions from Musha-Ntungwa area was evaluated using various plots like K/Rb versus Rb (Figure-6), Ta versus Ga (Figure-7), and K₂O versus Rb (Figure-8).

As any rock falling below the defining line of mineralization²³ is considered barren, while those plotting above the lines are considered mineralized (Figure-7). However, for the plot of K/Rb versus Rb with defining line²² for mineralisation, the pegmatite rocks samples fall below the line in mineralised field (Figure-6).

Similarly, the plot K₂O versus Rb (Figure-8) of pegmatitic intrusion of study area indicates the high Rubidium content revealed the (Tin) rare metal pegmatites²⁴.

Rare earth elements: The rare-earth element (REE) abundances of the pegmatitic intrusion are listed in Table-3. The plot of RRE chondrites after Boynton 1984 (Figure-10) shows anomalous Tb element that is mostly unreported in geochemistry but there is a remarked distribution of REE depleting that probably reveals alteration and weathering processes occurred in Musha Ntungwa area.

Table-1: The major oxides of pegmatitic intrusion in metasedimentary rock in Musha-Ntunga area (all values are in Wt%).

Rock	Pegmatites						Quartz vein	Cassiterite ore	
Sample	MN23	MN24	MN25	MN26	MN27	MN28	MN Qtz vein	MN cass32	MN cass33
SiO ₂	69.57	70.66	69.19	67.08	69.23	69.75	95.2	73.0	71.1
Al ₂ O ₃	28.7	26.7	27.4	28.3	27.1	26.8	1.56	9.2	9.76
Fe ₂ O ₃	0.37	0.34	0.32	0.32	0.31	0.33	1.17	5.21	5.54
CaO	0.07	0.06	0.06	0.06	0.07	0.06	0.19	4.53	4.81
MgO	0.04	0.03	0.03	0.03	0.04	0.03	0.05	3.1	3.26
Na ₂ O	0.06	0.06	0.06	0.06	0.06	0.06	0.22	1.27	1.37
K ₂ O	1.03	0.99	1.00	1.03	1.02	1.00	0.43	1.36	1.42
Cr ₂ O ₃	0.005	0.006	0.005	0.004	0.005	0.006	0.009	0.017	0.017
TiO ₂	0.01	0.01	0.01	0.01	0.01	0.01	0.04	0.5	0.55
MnO	0.07	0.08	0.08	0.08	0.07	0.08	0.01	0.08	0.08
P ₂ O ₅	0.08	0.06	0.07	0.05	0.07	0.07	0.08	0.09	0.09
SrO	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
BaO	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.03	0.03
LOI	0.98	0.99	0.66	0.77	0.88	0.75	0.33	1.01	0.96
Total	101.0	100.0	99.1	98.2	98.8	99.5	99.3	100.6	99

Table-2: The Trace elements of pegmatite intruded metasedimentary rock from Musha-Ntunga area (all values are in ppm).

Rock	Pegmatites						Qtz vein	Cassiterite ore	
Sample	MN23	MN24	MN25	MN26	MN27	MN28	MN Qtz vein	MN cass32	MN cass33
Ta	>2500	>2500	>2500	>2500	>2500	>2500	>2500	>2500	3
Nb	1070	1095	1245	1165	1100	1250	1025	1010	4
Sn	>10000	>10000	>10000	>10000	>10000	>10000	10000	10000	41
Cr	<10	<10	<10	<10	<10	<10	90	80	20
Ga	40.5	37.7	38.3	42.0	41.2	39.2	14.7	14.3	2.1
Hf	65.9	77.0	75.7	76.0	68.1	76.5	13.1	11.7	0.4
Zr	232	270	259	271	265	260	131	131	18
Sr	25.5	22.0	23.1	23.7	23.2	22.5	73.5	73	17.4
Rb	741	695	696	751	710.1	698	88.9	87.2	32.3
Cs	35.6	33.0	33.7	36.6	34.6	33.1	12	12.45	3.98
Ba	102.0	91.6	94.1	95.8	98.2	92.0	278	270	88.2
Th	5.72	5.67	5.62	5.96	5.65	5.81	4.82	5.29	3.56
U	5.72	6.36	5.50	6.81	5.65	6.25	2.17	2.01	0.71
V	<5	<5	<5	<5	<5	<5	120	121	6
W	479	502	512	469	480	510	51	43	53
Co	<1	<1	<1	<1	<1	<1	17	17	b.d.
Cu	8	7	7	7	7	7	13	14	11
Mo	<1	<1	<1	<1	<1	<1	<1	<1	<1
Ni	7	6	8	5	6	7	47	41	<1.
Pb	203	216	219	202	210	217	7	8	4
Sc	1	1	1	1	1	1	17	16	1
Tl	<10	<10	10	10	<10	10	<10	<10	<10
Zn	10	7	8	8	8	9	25	25	2
Cd	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Ag	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
As	<5	6	6	<5	5	6	5	7	<0.5
Li	150	150	150	150	145	145	100	90	50

< :bdl: below detection limit (www.alsglobal.com).

Table-3: Rare earth elements of pegmatite intruded metasedimentary rock from Musha-Ntunga area (all values are in ppm).

Rock	Pegmatites						Qtz vein	Cassiterite ore	
Sample	MN23	MN24	MN25	MN26	MN27	MN28	MN Qtz vein	MN cass32	MN cass33
La	26.2	24.2	24.6	25.5	25.1	24.3	13.5	15.3	6.8
Ce	49.0	44.4	46.7	48.0	46.1	45.0	23.1	26.3	11.5
Pr	5.86	5.22	5.37	5.60	5.70	5.50	2.55	2.85	1.19
Nd	31.3	27.2	28.0	29.3	30.5	29.2	9.3	10	3.7
Sm	7.80	6.18	6.77	7.50	7.90	6.01	1.61	1.69	0.57
Eu	1.12	1.07	1.15	1.19	1.09	1.11	0.48	0.44	0.09
Gd	3.45	3.24	3.31	3.24	3.30	3.18	1.48	1.5	0.33
Tb	0.33	0.31	0.35	0.33	0.32	0.34	0.22	0.2	0.01
Dy	1.26	1.25	1.23	1.31	1.27	1.22	1.26	1.32	0.24
Ho	0.19	0.19	0.19	0.22	0.19	0.20	0.26	0.26	0.04
Er	0.46	0.44	0.53	0.54	0.48	0.51	0.85	0.83	0.12
Tm	0.06	0.08	0.08	0.08	0.07	0.08	0.13	0.09	0.02
Yb	0.44	0.45	0.48	0.57	0.45	0.47	0.73	0.72	0.11
Lu	0.08	0.08	0.07	0.09	0.07	0.08	0.12	0.12	0.02
ΣREE	127.55	114.31	118.83	123.47	122.54	117.2	55.59	61.62	24.74

Table-4: Elemental ratios of pegmatitic intrusion in metasedimentary rock from Musha-Ntunga area.

Rock	Pegmatites						Qtz Vein	Cassiterite ore	
Sample	MN23	MN24	MN25	MN26	MN27	MN28	MN Qtz vein	MN Cass 32	MN Cass 33
K/Rb	11.53	11.82	11.92	11.38	11.92	11.89	126.99	135.18	110.51
K/Cs	240.18	249.04	246.33	233.62	244.72	250.80	940.83	946.83	896.89
Rb/Ba	7.26	7.58	7.39	7.83	7.23	7.58	0.31	0.32	0.36
Rb/Sr	29.05	31.59	30.12	31.68	30.60	31.02	1.20	1.19	1.85
Rb/Cs	20.81	21.06	20.65	20.51	20.52	21.08	7.40	7.00	8.11
K ₂ O/Na ₂ O	17.16	16.50	16.66	17.16	17	16.66	1.07	1.03	1.95
Nb/Ta	0.42	0.43	0.49	0.46	0.44	0.50	0.41	0.40	1.33
Zr/Hf	3.52	3.50	3.42	3.56	3.89	3.39	10	11.19	45

Molar Na₂O – Al₂O₃ – K₂O plot

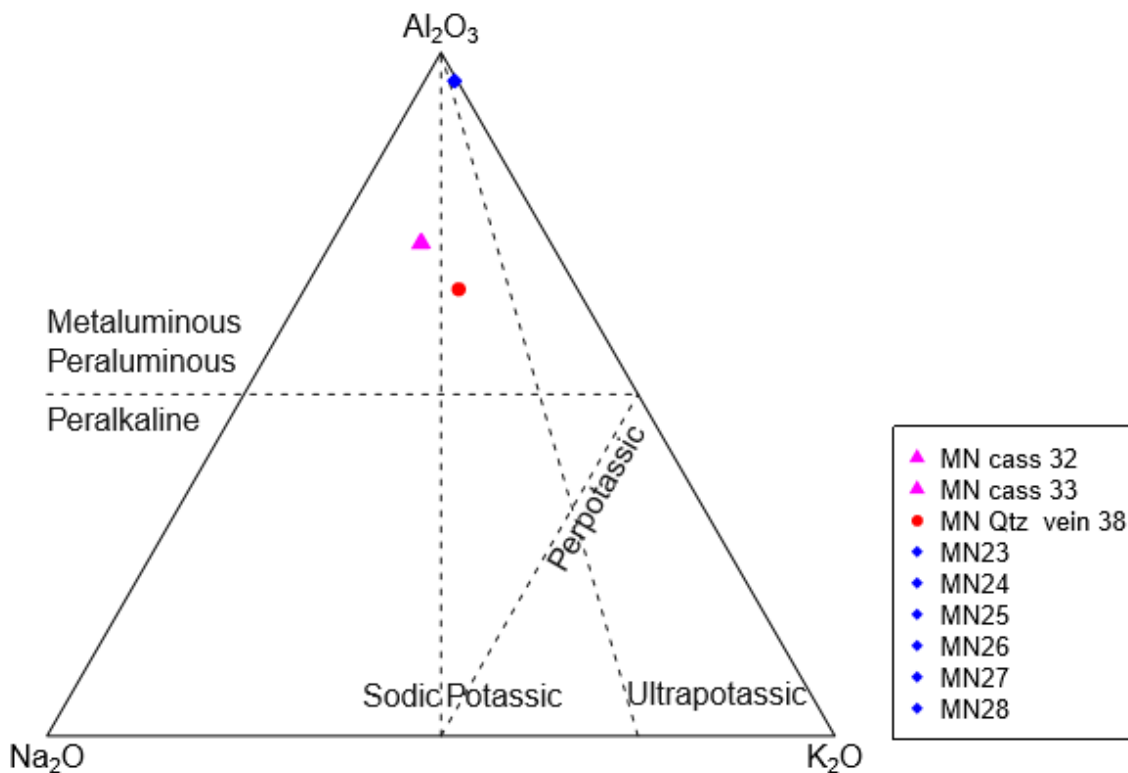


Figure-5: Molar Na₂O-Al₂O₃-K₂O plot of Pegmatites, cassiterite ores, and Quartz vein in Musha-Ntungwa area. Blue: pegmatite, Red: quartz Vein, and pink: cassiterite ore.

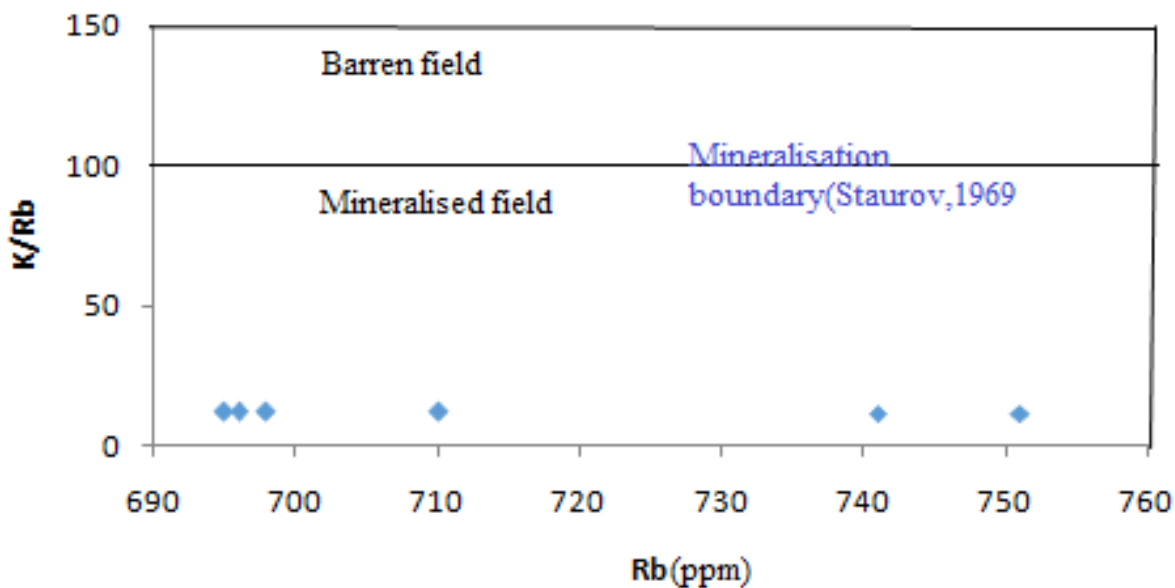


Figure-6: K/Rb versus Rb Plot of pegmatites from Musha-Ntungwa area²².

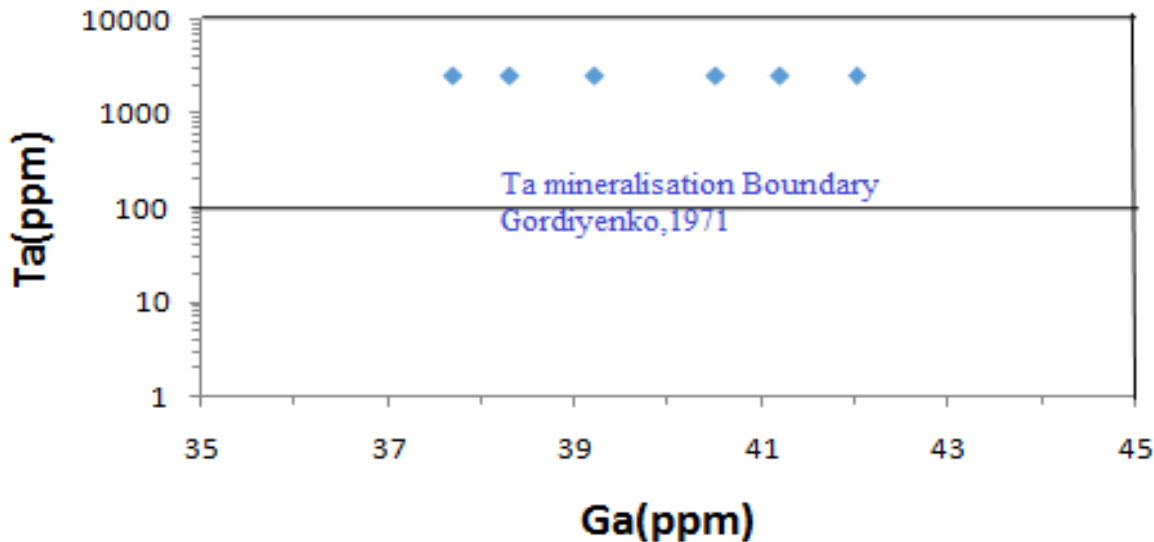


Figure-7: Ta versus Ga plot of pegmatites from Musha-Ntungwa area²³.

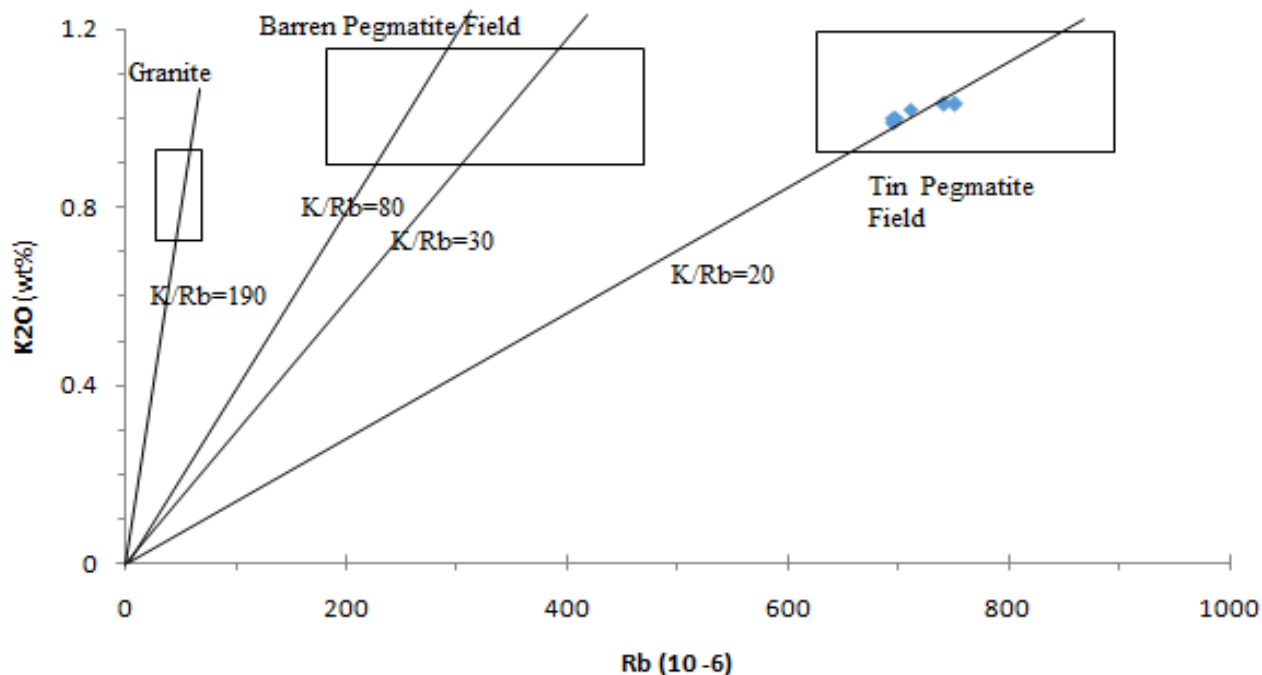


Figure-8: Rubidium versus Potassium contents of Tin-bearing, barren pegmatites and associated granites, samples of pegmatites from Musha-Ntungwa area.

The Tin-bearing pegmatites are distinguished by high enrichment of Rubidium content (Figure-9) with depletion of Sr and Ba chemical elements⁸.

Conclusion

The study area is lithostratigraphically dominated by schists in Musha formation of metasedimentary rocks which are intruded by mappable bodies of pegmatites and some discordant quartz veins. These altered pegmatites underwent the process of

metasomatism by hydrothermal fluids which induced the precipitation of rare metals potentially hosted in Pegmatites. This study revealed that these are (Tin) pegmatites bearing Tin and Niobium-tantalum. Petrography and Geochemical results of intrusive body of pegmatite in origin indicated the process of alteration occurred after the primary emplacement of ore, thus mineralized intrusion as confirmed by the signature of mineralization printed in plots of Ta vs. Ga and K/Rb vs. Rb. The area is nowadays mined by the mining company named Piran Rwanda Ltd.

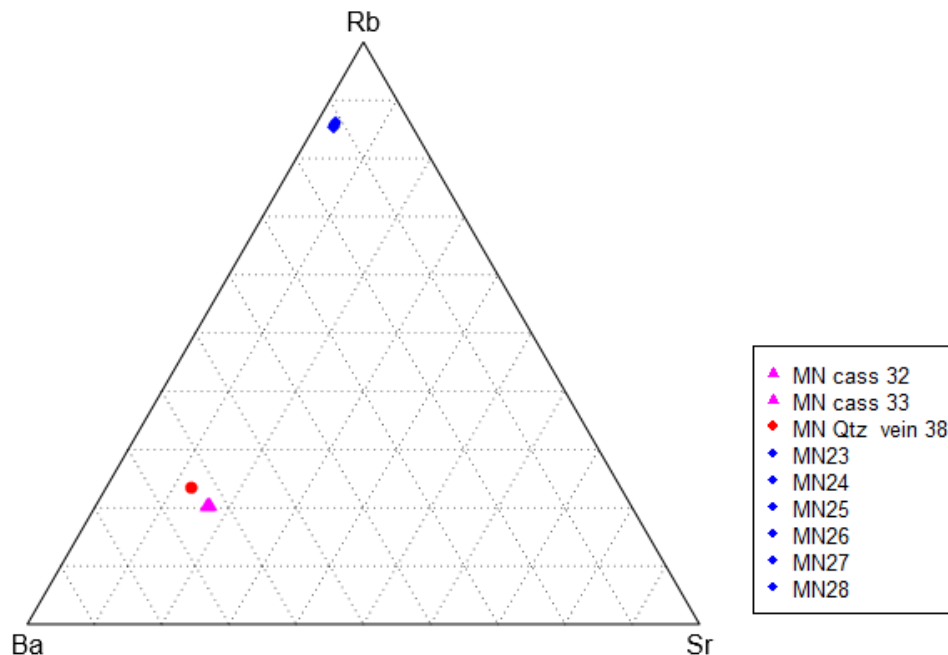


Figure-9: Ba-Rb-Sr ternary plot of pegmatites (blue at left top apex), Quartz vein(red) and Cassiterite ores(pink) from Musha Ntungwa area, Eastern province, Rwanda.

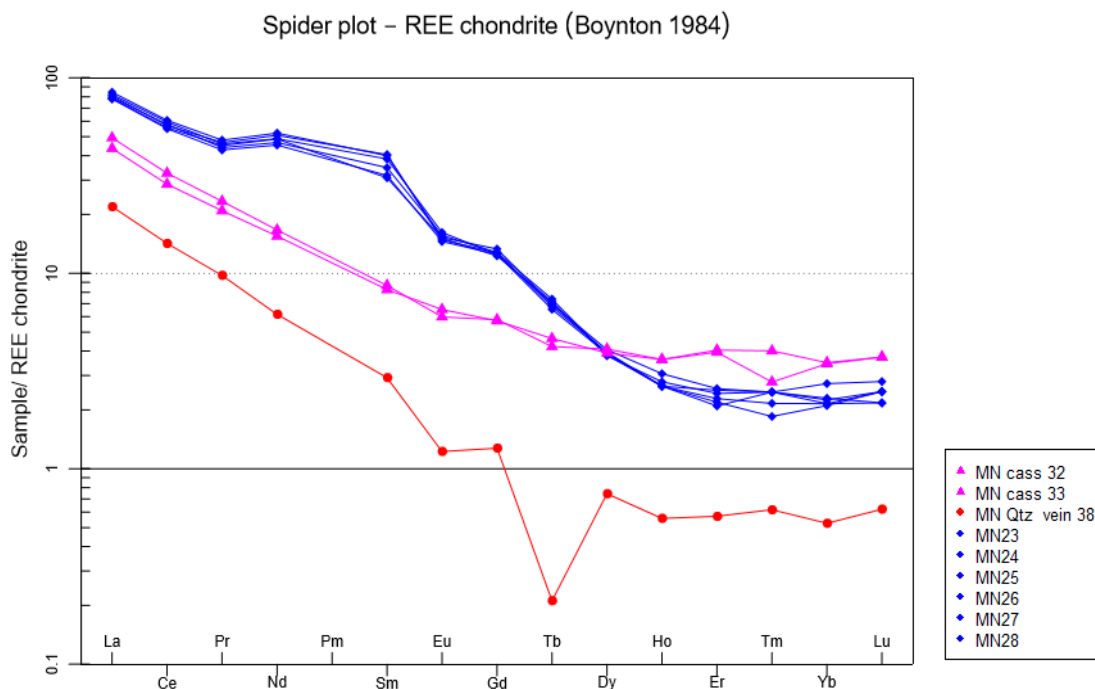


Figure-10: REE chondrite plot for pegmatite(blue), Cassiterite ore(pink) and quartz vein(red) from Musha-Ntungwa area, Eastern province, Rwanda²⁵.

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