

GEOCHEMISTRY AND Nd - ISOTOPIC COMPOSITION OF HIGH SILICA RHYOLITES IN THE NEOARCHAEAN MUSOMA-MARA GREENSTONE BELT, NORTHERN TANZANIA: EVIDENCE FOR THE PRESENCE OF OLDER CONTINENTAL CRUST

S Manya

Department of Geology, University of Dar es Salaam, P.O. Box 35052 Dar es Salaam
Email: shukrani73@udsm.ac.tz

ABSTRACT

The high silica rhyolites of the Kibasuka Hill, although volumetrically minor, constitute a part of the greenstone sequence in the northern Musoma-Mara Greenstone Belt (MMGB). They are characterized by high silica contents (63.38 – 81.65 wt %), very low CaO (≤ 0.05 wt %), variable to unusually high K₂O (2.24 – 13.43 wt %) most likely attributed to strong sericite alteration, and their major element composition are similar to those of melts derived from partial melting of metapelites. The samples have very low concentration of the transition elements, Sr (3.13 – 48.4 ppm) and very large negative Europium anomalies ($Eu/Eu^ = 0.27 - 0.48$). Their Nd isotopic composition reveals that the T-depleted mantle (DM) model ages of the samples are highly variable and range from 2867 Ma to 4015 Ma. These geochemical features are consistent with generation of the high silica rhyolites by partial melting of a heterogeneous source composed of juvenile Archaean rocks of the MMGB and older sedimentary rocks. The later imparts to the rocks very old mean crustal residence ages. Partial melting of this suite took place at low-pressure conditions within the continental crust where plagioclase was a stable phase. Such old T_{DM} model ages of up to 4015 Ma recorded by the high silica rhyolites suggest the presence of very old continental crust in the MMGB which has so far not been identified anywhere else in the Tanzania Craton.*

INTRODUCTION

The Musoma-Mara greenstone belt (MMGB) of northeastern Tanzania is a Neoarchaean granite-greenstone terrane forming part of the Tanzania Craton (Barth 1990, Borg and Shackleton 1997). The MMGB is a depository of medium to large scale gold deposits with two operating mines at Buhemba and North Mara. Several gold projects are at advanced stages of exploration in various places in the MMGB. Previous documented and published geological works are limited to geological mapping done in the 1960's by Gray and Macdonald (1964) for the northern MMGB, and Mulgrew (1964) for the southern MMGB. Because of the economic potential of the MMGB, the area has recently attracted many geochemical studies including those of Manya *et al.*

(2006), Manya *et al.* (2007a, b), Kazimoto (2007) and Mtoro (2007). These works have largely contributed to our modern understanding of the geological evolution of the MMGB and factors that control gold mineralization through geochemical and geochronological studies.

The focus of the present study is the rhyolitic volcanic rocks which form part of the northern MMGB stratigraphy. These rocks were mapped by Gray and Macdonald (1964) and were considered to belong to the lower unit of felsic volcanic rocks of the Nyanzian Supergroup (Fig. 1). The geochemical and Nd isotopic composition of the rocks is presented with a view to unravel their petrogenesis.

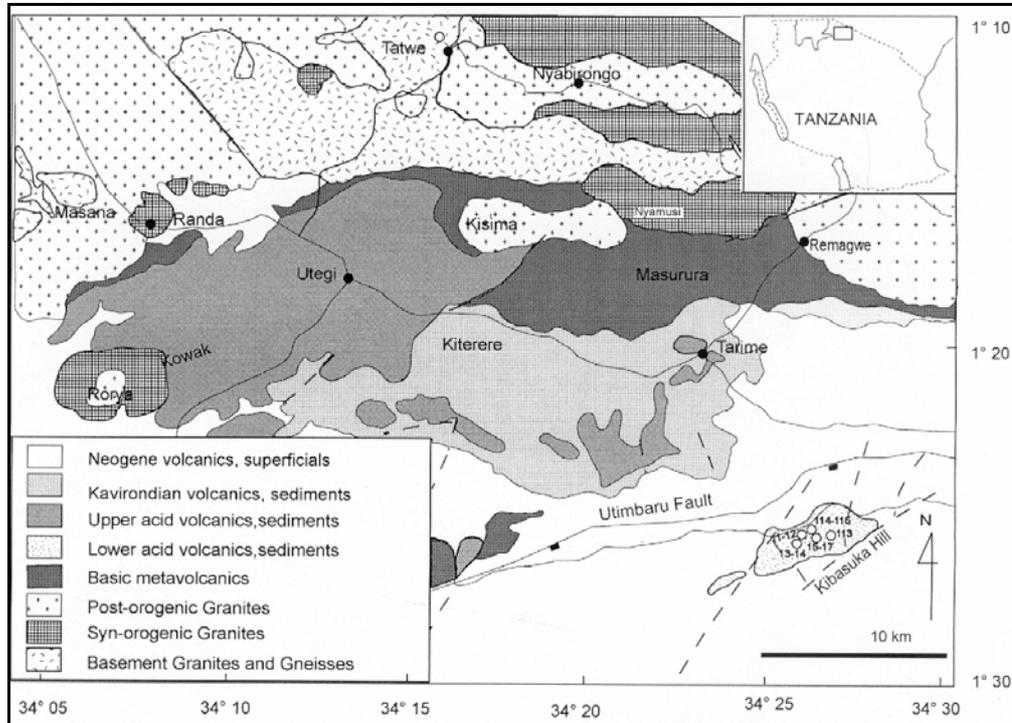


Figure 1: Geological map of northern Musoma-Mara greenstone belt of northern Tanzania (after Gray and Macdonald 1964) showing the sample locations.

Geological background

The Musoma-Mara Greenstone Belt (MMGB) is located in the northeastern part of the Neoproterozoic granite-greenstone terrane of the Tanzania Craton, east of Lake Victoria. The geological setting for the Tanzania Craton and its subdivision into the high-grade Dodoman belt of central Tanzania and the low-grade granite-greenstone terrane of northern Tanzania is described elsewhere (e.g. Borg and Shackleton 1997, Manya *et al.*, 2006). The geology of the northern MMGB has been described by Stockley (1936), Gray and Macdonald (1964) and recently by Manya *et al.* (2006), Manya *et al.* (2007a, b). Gray and Macdonald (1964) subdivided the supracrustal rocks of northern MMGB into four units (from bottom upwards): the mafic volcanic rocks, lower felsic volcanic rocks (the object of this study), the upper felsic volcanic rocks of the Nyanzian Supergroup and the

metasedimentary rocks of the Kavirondian Supergroup. Manya *et al.* (2006) reported ion microprobe U-Pb zircon ages for the MMGB rocks and showed that the mafic volcanic rocks are high-Mg andesites that are dated at between 2676 ± 12 and 2669 ± 9 Ma whereas the upper felsic volcanic rocks are dated at 2668 ± 30 Ma together with the Na-rich granitoids that yielded an age of 2668 ± 11 Ma. The Kavirondian volcanic rocks yielded a zircon U-Pb age of 2667 ± 8 Ma (Manya *et al.* 2006). The MMGB magmatism was concluded at 2649 ± 36 Ma by the intrusion of post-orogenic K-rich granites (Manya *et al.* 2006).

The study area forms part of the greenstone sequence in northern MMGB and the study is focussed on rhyolitic volcanic rocks that are volumetrically minor, as they are restricted to the Kibasuka Hill, an isolated, fault-bounded hill that is on the

downthrown side of the major Utimbaru Fault (Fig. 1). These rocks represent a narrow band of recrystallized, sheared and altered felsic volcanic and volcanoclastic rocks (including lapilli tuff) that are interlayered with detrital sedimentary rocks. Because of their fault-bound nature, the stratigraphic relationship with other rock units in the belt is hard to establish.

MATERIALS AND METHODS

A total of 10 rhyolitic samples were carefully sampled from fresh outcrops (see figure 1 for sample locations) at Kibasuka Hill. The samples were thin-sectioned and petrographically studied. The samples were analyzed for major elements at the Pheasant Memorial Laboratory (PML) for Geochemistry and Cosmochemistry of the Institute for the Study of the Earth's Interior of Okayama University at Misasa, Japan using a Phillips PW2400 X-ray Fluorescence spectrometer on glass beads made from mixing 0.5g of powdered sample and 5g of lithium tetraborate as flux (Takei, 2002). Analytical reproducibility between replicate analyses were better than 0.5 %. Trace elements were measured using a Yokogawa PMS2000 Inductively Coupled Plasma Mass Spectrometer (ICPMS) fitted with a flow injection system using the methods of Makishima and Nakamura (1997) for Rb, Sr, Y, Cs, Ba, REE, Pb, Th and U; Makishima *et al.* (1999) for Zr, Hf, Nb and Ta. Analytical reproducibility between replicate analyses was better than 7% for trace elements.

Five selected samples were also analyzed for Nd isotopic compositions in a static multicollection mode, as well as Sm and Nd concentrations using a Finnigan MAT262 mass spectrometer at the PML, Misasa, Japan. The analytical procedures for chemical separation and mass spectrometry are described in Makishima and Nakamura (1991). Typical analytical reproducibility

was 0.006 % for Nd isotopes. Normalizing factors to correct for isotopic fractionation during analyses are $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$. Replicate analyses of the La Jolla Nd standard gave $^{143}\text{Nd}/^{144}\text{Nd} = 0.511849 \pm 7$ (2σ , $n = 7$, measurements taken over a period from May to August 2003). Typical blank values are 0.25 and 5 pg for Sm and Nd, respectively and are therefore negligible.

RESULTS

Petrographic studies of the samples show that they are composed of feldspar and/or quartz phenocrysts set in a fine-grained groundmass of quartz, feldspar and sericite. Accessory phases include sphene and zircon in some specimens. Plagioclase is strongly altered to sericite and in some samples it has lost its tabular habit (Fig. 2).

Major and trace element compositions for the 10 rhyolitic samples are presented in Table 1. These samples are generally silica-rich rocks and based on SiO_2 contents, they can be broadly subdivided into two groups: the low silica group, comprising of two samples (TA 15 and TA 113) that have lower SiO_2 values of dacitic composition (63.38 – 66.05 wt %) and the high silica group that consists of eight samples with a fairly restricted range in SiO_2 contents (79.35 - 81.65 wt %). The low silica samples correspond with unusually high K_2O values (12.85-13.43 wt %) most likely attributed to strong sericite alteration; whereas the high silica samples have low to moderate (2.24-7.10 wt %) K_2O contents. This suite shows extremely low CaO contents (≤ 0.05 wt %) along with all other major element compositions which are less than 0.90 wt %. On the Winchester and Floyd (1977) classification scheme which is based on immobile elements (Fig. 3), these samples can be classified as rhyolites and based on their high silica contents, the samples are in this study termed as high-silica rhyolites.

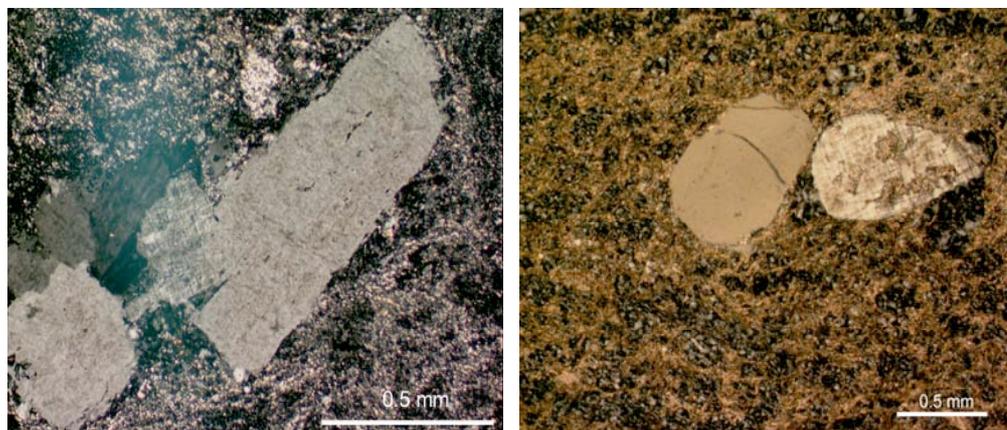


Figure 2: Thin sections of samples TA 113 (left) and TA 15 (right) showing plagioclase and quartz (in TA 15) phenocrysts surrounded by a fine grained matrix mainly composed of sericite.

Table 1: Major (wt %) and trace (ppm) element compositions for the northern MMGB high silica rhyolites

	TA11	TA 12	TA 13	TA 14	TA15	TA 16	TA 17	TA 113	TA 114	TA 115
SiO ₂	80.0	80.3	81.3	81.6	63.4	80.3	81.0	66.1	79.4	79.4
TiO ₂	0.38	0.11	0.10	0.10	0.18	0.10	0.10	0.16	0.39	0.41
Al ₂ O ₃	10.6	12.4	11.2	10.3	19.9	10.8	10.5	18.0	11.0	11.5
Fe ₂ O ₃	0.50	0.63	0.90	0.36	0.51	0.24	0.38	0.33	0.51	0.89
MnO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
MgO	0.21	0.40	0.62	0.23	0.43	0.26	0.32	0.38	0.21	0.28
CaO	0.04	0.02	0.01	0.00	0.00	0.01	0.01	0.00	0.04	0.05
Na ₂ O	0.17	0.10	0.08	0.11	0.39	0.15	0.17	0.18	0.13	2.51
K ₂ O	7.02	2.24	3.89	6.59	13.43	7.10	6.51	12.85	6.93	3.36
P ₂ O ₅	0.06	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.06	0.06
LOI	0.90	3.43	1.85	0.92	1.44	1.21	1.07	1.54	1.07	1.68
SUM	99.8	99.6	100	100.2	99.7	100.2	100.1	99.5	99.7	100.1
Mg#	46	55	58	56	63	68	62	69	45	38
Cr	6.11	bl	4.40	4.60	bl	5.27	bl	4.82	bl	bl
Ni	3.34	0.41	0.67	0.57	2.67	bl	0.60	1.41	0.91	1.70
Zr	354	194	205	177	327	148	176	299	348	394
Hf	11.3	5.88	6.77	5.86	10.7	4.88	5.67	9.14	11.15	12.33
Nb	22.9	9.98	10.94	9.08	15.4	6.83	9.08	13.2	23.8	26.4
Ta	1.79	0.94	1.03	0.79	1.38	0.68	0.73	1.17	1.86	1.97
Rb	94.3	82.9	136	125	280	115	117	178	88.1	49.5

Sr	48.4	6.90	3.13	7.80	8.27	9.40	8.69	14.6	32.8	44.1
Y	163	30.7	31.6	21.0	41.5	19.9	22.9	33.3	111.7	139.0
Cs	1.15	0.81	1.42	0.97	2.11	1.42	1.53	2.24	1.23	0.98
Ba	1168	153	162	547	620	572	566	508	603	578
La	63.3	36.7	54.2	46.6	46.5	23.1	24.1	25.1	72.3	108.3
Ce	135	78.7	116	91.7	98.4	51.4	55.9	52.4	151	151
Pr	16.0	8.63	12.5	9.45	9.97	4.87	5.33	5.23	18.4	21.3
Nd	67.3	31.2	45.8	33.4	34.3	17.5	18.7	17.9	75.6	82.6
Sm	15.4	5.28	8.63	5.54	6.74	3.04	3.66	3.95	17.0	17.2
Eu	2.41	0.59	0.66	0.57	1.01	0.41	0.50	0.53	2.79	2.56
Gd	15.9	4.34	6.55	4.28	6.54	2.81	3.57	4.25	18.2	17.2
Tb	2.75	0.72	0.94	0.64	1.15	0.50	0.61	0.86	3.28	2.86
Dy	18.7	4.55	5.37	3.65	6.77	3.17	3.92	5.56	19.7	18.0
Ho	3.77	1.06	1.12	0.77	1.27	0.69	0.83	1.18	4.09	3.86
Er	10.5	3.13	3.19	2.23	3.05	1.99	2.26	3.04	11.1	10.4
Tm	1.74	0.56	0.53	0.36	0.44	0.34	0.37	0.45	1.76	1.68
Yb	12.1	4.20	3.76	2.59	2.78	2.35	2.60	3.05	10.5	11.6
Lu	1.64	0.58	0.56	0.37	0.37	0.33	0.37	0.42	1.58	1.59
Pb	12.8	3.51	3.11	5.01	9.71	4.46	4.64	7.72	7.49	5.04
Th	19.2	14.8	18.6	16.2	28.8	13.4	14.4	25.4	19.1	16.8
U	3.94	3.69	3.64	2.57	5.33	2.72	3.86	4.99	5.73	4.41
La/Yb	5.2	8.8	14.4	18.0	16.8	9.81	9.27	8.21	6.86	9.37
Eu/Eu* (La/Yb)	0.47	0.37	0.27	0.36	0.46	0.43	0.43	0.40	0.48	0.46
CN	3.53	5.90	9.71	12.1	11.3	6.62	6.25	5.54	4.63	6.32

bl = below detection limit

The samples have very low concentrations of transition elements, some of which showed to be below detection limit. The concentrations of the Large Ion Lithophile Elements (LILE) are highly variable with Rb contents of 49.5 – 280 ppm, a feature which is also shared by Ba concentrations (153 – 1168 ppm). The samples have unusually low contents of Sr (3.13 – 48.4 ppm).

On chondrite-normalized diagrams (Fig. 4), the samples show patterns with slight enrichment of the LREE relative to HREE characterized by low La/YbCN = 3.53 -12.1. The samples display strongly negative Eu anomalies (Eu/Eu* = 0.27-0.48, average

0.41). On primitive, mantle-normalized spidergrams (Fig. 5), the samples display coherent patterns with moderate to strong depletion in Ba and Sr as well as Nb, Ta and Ti. The samples are also characterized by enrichment in Th, U and K but not Pb. Samples TA 15 and TA 113 plot together with the majority of the samples in the spidergram and not as an exclusive group as shown in major element composition.

The samples have $^{147}\text{Sm}/^{144}\text{Nd}$ ratios that range between 0.1020 and 0.1466 and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of between 0.510910 ± 8 and 0.511895 ± 10 . Their corresponding T_{DM} ages are highly variable and range from 2867 Ma to 4015 Ma (Table 2).

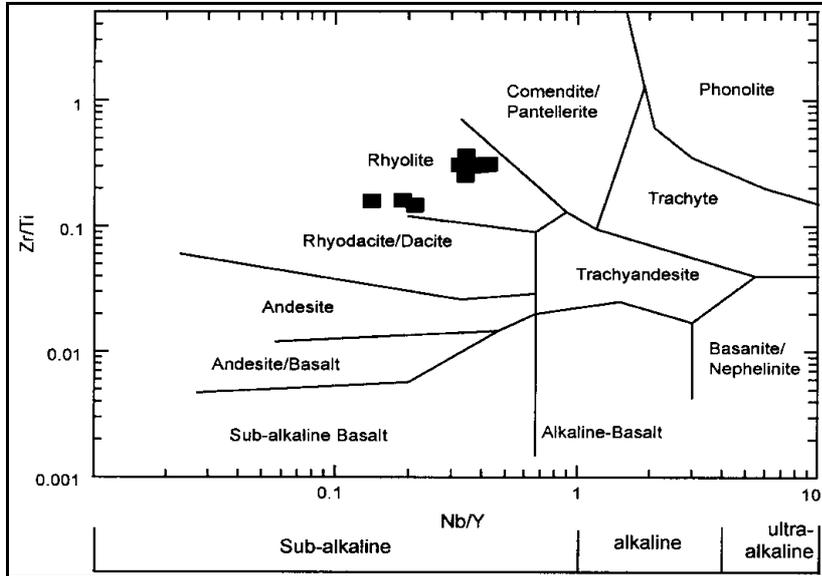


Figure 3: Zr/Ti – Nb/Y classification diagram of Winchester and Floyd (1977) for the northern MMGB high silica rhyolites.

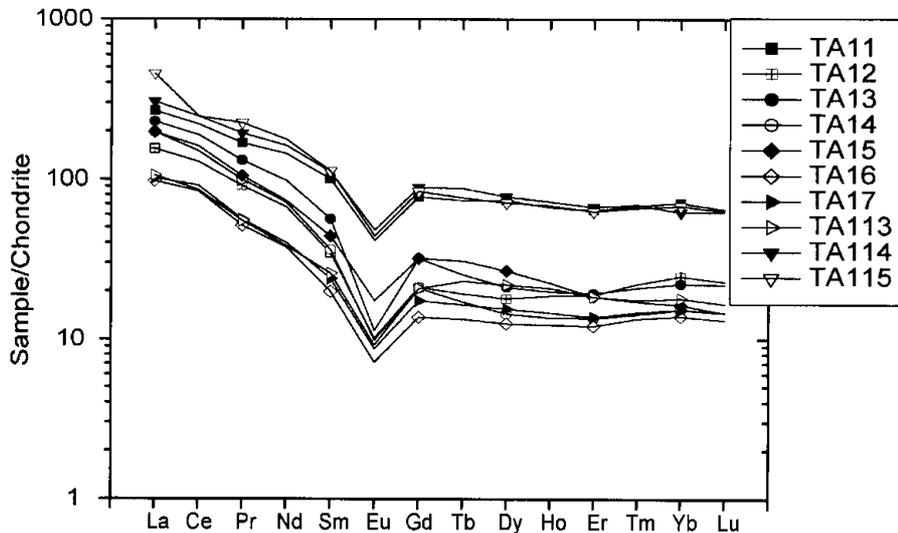


Figure 4: Chondrite-normalized rare earth elements diagram (normalizing values after Sun and McDonough 1989) for the northern MMGB high silica rhyolites.

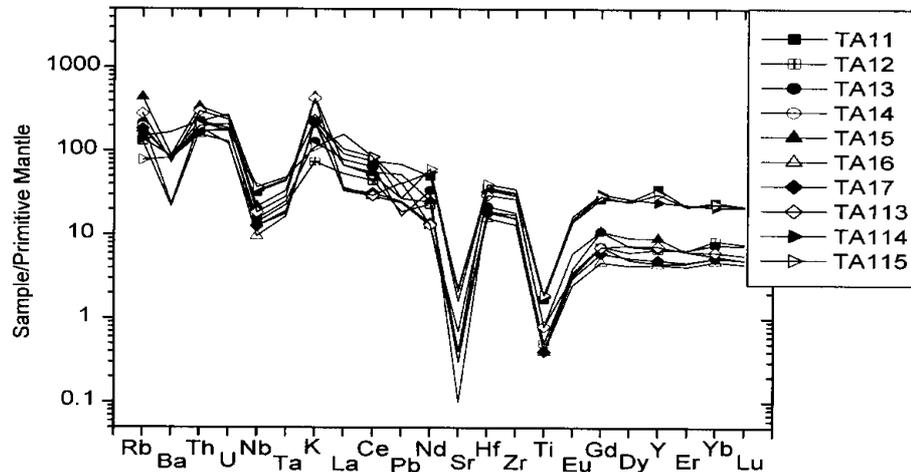


Figure 5: Primitive mantle normalized extended trace elements diagram (normalizing values after Sun and McDonough 1989) for the northern MMGB high silica rhyolites.

Table 2: Sm-Nd isotopic data for the high silica rhyolites from northern MMGB

Sample	Sm ppm	Nd ppm	$^{147}\text{Sm}/^{144}\text{Nd}$	$^{143}\text{Nd}/^{144}\text{Nd}$	T_{DM} (Ma)
TA 11	15.22	65.90	0.1396	0.5117039 ± 9	2975
TA 14	5.33	31.55	0.1020	0.510910 ± 8	3050
TA 15	6.63	33.62	0.1193	0.511041 ± 7	3393
TA 115	20.06	82.56	0.1466	0.511895 ± 10	2867
TA 113	4.22	19.33	0.1321	0.510989 ± 12	4015

Calculations are based on a decay constant of 6.54×10^{-12} per year for ^{147}Sm and DM values for Nd are $(^{143}\text{Nd}/^{144}\text{Nd})_{\text{today}} = 0.51316$, $(^{147}\text{Sm}/^{144}\text{Nd})_{\text{today}} = 0.2137$

DISCUSSION

The samples have very low concentrations of Cr and Ni suggestive of derivation from an evolved crustal source. They also possess extremely low contents of Sr (3 – 48 ppm, average 18 ppm) and strongly negative Eu anomalies ($\text{Eu}/\text{Eu}^* = 0.27 - 0.48$, average 0.41). The low Sr content is accompanied by low CaO contents. These elements are considered to be mobile under secondary processes (Rollinson, 1993) and this could be one way to explain their low abundances. Alternatively, their low abundance could reflect a primary feature. HFSE and REE are considered by many researchers (e.g. Jochum

et al., 1991) to be immobile during alteration of Archaean volcanic rocks, a fact supported by the coherent REE patterns of the samples (Fig. 4). Thus, the low contents of Eu, which together with Sr and CaO strongly partition in plagioclase mineral, suggest that plagioclase was involved in the magma generation/evolution either as a fractionating phase during magmatic differentiation or its retention as a residual phase during partial melting. Since these rocks are not spatially associated with other volcanic rocks in the MMGB, this suggests that the geochemical features reflect partial

melting of a plagioclase-bearing source rather than magmatic differentiation.

The low Sr contents of these rocks are associated with very low contents of CaO (< 0.05 wt %) but high contents of Ba (average 548 ppm). The fact that the high silica rhyolites have extremely low Sr but high Ba contents suggest that processes other than the presence of residual plagioclase during partial melting/and or plagioclase

fractionation are responsible for the low contents of CaO and Sr, as well as the negative Eu anomalies. These features can be attributed to formation of the high silica rhyolites by partial melting of metapelites, a fact supported by major element compositional similarities between melts derived from partial melting of metapelites (Patiño Douce and Johnston 1991) and the high silica rhyolites (Table 3).

Table 3: Comparison of the major element composition between the MMGB high silica rhyolites and the melts derived from the melting of a sillimanite-bearing metapelite

Element	Average MMGB high silica rhyolites	HQ36 melts at 10 kbar 950 °C ^a
SiO ₂	77.3	73.7
TiO ₂	0.20	0.40
Al ₂ O ₃	12.6	13.5
Fe ₂ O ₃	0.53	1.70
MnO	0.001	0.10
MgO	0.33	0.3
CaO	0.02	0.10
Na ₂ O	0.40	1.10
K ₂ O	6.99	6.90
P ₂ O ₅	0.03	0.00

^aExperimental results of Patiño Douce and Johnston (1991)

The concentrations of the REE in the rhyolites cover a wide range leading to variable La/Yb ratios of between 5.24 and 17.98 (average 10.67). In particular, The HREE contents in the high silica rhyolites are high and variable (Y = 20-163 ppm, average 58 ppm). Such a large variation in REE concentrations cannot be explained by formation from a homogenous source and as previously argued, the variation might be attributed to formation from a heterogeneous source.

In Sm-Nd system of dating, model ages are used to estimate the time at which a sample separated from its mantle source region and hence giving crust formation ages for igneous and meta-igneous rocks (Rollinson 1993). Studies of the initial ¹⁴³Nd/¹⁴⁴Nd

ratios from Precambrian terranes suggest that the continental crust formed from a depleted mantle and hence model ages are calculated with reference to the depleted mantle (DM) reservoir. Although the use of model ages is faced with several limitations (Rollinson 1993), they offer an insight into the crust formation ages especially with rocks whose REE were not disturbed as those of the Kibasuka Hill rhyolites. The calculated T-depleted mantle (DM) model ages for the high-silica rhyolites range from 2867 Ma to 4015 Ma. The T_{DM} ages of 2867 Ma are shared by other volcanic rocks of the MMGB including the high-Mg andesites, dacites and the Na-rich granitoids discussed in Manya et al. (2007a). The variable T_{DM} model ages demonstrate that the high silica rhyolites are derived from a mixture of

young and very old crust. The old T_{DM} model ages of up to 4015 Ma recorded by the high silica rhyolites suggest the presence of very old continental crust in the MMGB which has so far not been identified anywhere else in the Tanzania Craton. This signifies that the rhyolites could be derived from a heterogeneous source involving the juvenile material represented by the late Archaean igneous rocks of the MMGB which are represented by samples with younger T_{DM} model ages and the very old metapelites represented by samples with very very old T_{DM} model ages.

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