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Sulphur Isotopic Studies of Barite Mineralization of The Gombe Hill, N. E Nigeria: Implication for Source and Formation Process

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Abstract: *Sulphur isotopic study on barite ($BaSO_4$) mineralizations, located on the Liji hill at Bicije on the outskirt of Gombe town, Gombe State, N.E Nigeria has shown that $\delta^{34}S$ values of barite vary from +17.2 ‰ to +19.8 ‰. The data, have demonstrated that the hydrothermal fluid seems to have supplied the barium ions (Ba^{2+}) while the sea water supplied the sulphate ions (SO_4^{2}). These isotopic signatures suggest mixing of sea water sulphate with Ba-carrying crustal fluid for barite deposits, with its initial deposition linked to the detrital sediments of the Bima Formation (host rock).*

Keywords: barite, fluorite, sinistral, displacement, stable isotopes, microthermometry.

INTRODUCTION

There are siliciclastic and calcareous host rocks that are associated with barite (Sharma et al., 2006; Haruna, 2013; El-Nafaty, 2015; Vandi et al. 2019). Along Biu Road, on the outskirts of Gombe town in the Bicije area, lie the barite deposits of the Gombe hill. Situated atop vast anticlinal formations of the Gombe hill, the barite resources have been exploited artisinally for almost ten years. Here, we provide a S-isotopic investigation of barite. The results are utilized to investigate the formation process and the source of S and Ba.

GEOLOGIC SETTING

The geology of the study area is located between Latitudes 10°15′ 0” N and 10°19′ 30” N and Longitudes 11°10′ 12” E and 11°14′ 42” E (Figure 1) in the Gongola Sub-basin. Sedimentation began with the continental Bima sandstones, followed by the transitional Cenomanian Yolde Formation. Overlying the Yolde is the Marine deposition of Late Cenomanian to Mid-Santonian Pindiga Formation which is in this area of study, absent. This was followed by folding, faulting and uplift. Then began the deposition of the of the continental Gombe sandstones in the Maastrichtian and lastly the Kerri-Kerri Formation in the Paleocene (Figure 2).

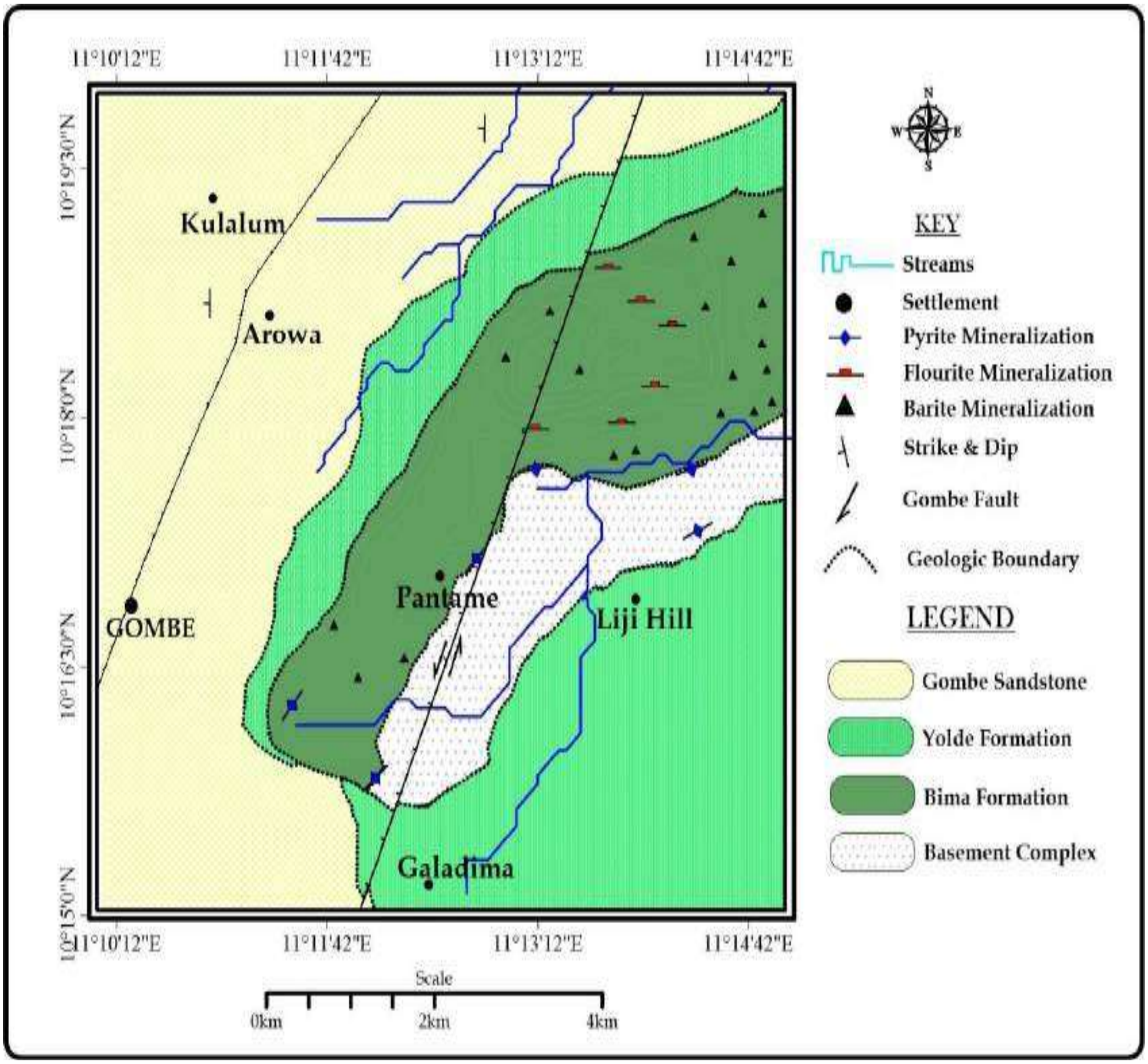

Figure 1: Geology of the Study Area.

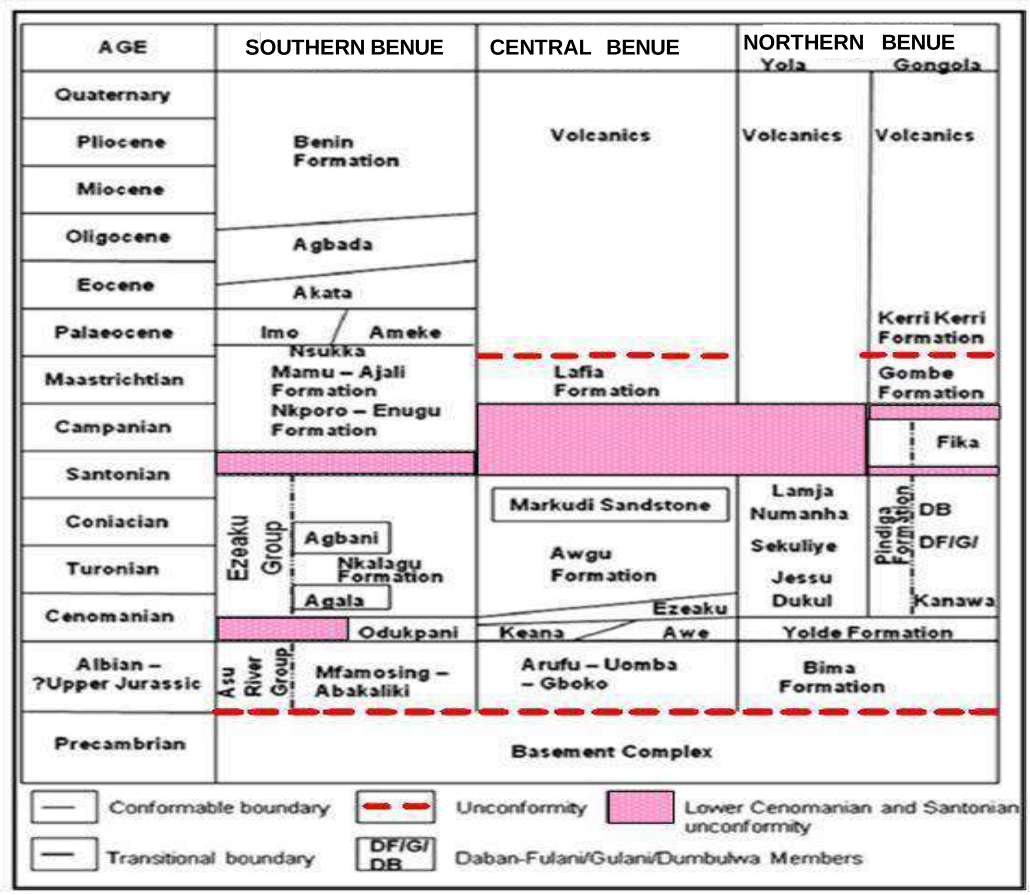

Figure 2: Stratigraphy of the Southern, Central and Northern Benue Trough (Modified After Abubakar, 2014).

PETROGRAPHY

The geology/lithologies encountered include Basement Complex rocks and Cretaceous sedimentary formation. The Basement Complex rocks occur as an inlier consisting of biotite granites and hornblende biotite granites. They are essentially composed of quartz and microcline and some orthoclase, plagioclase, biotite, hornblende and opaque minerals. Large quartz and microcline crystals are fractured. The Cretaceous sediment is the Bima sandstones. Barite veins are associated with the Bima sandstones whose large grains of quartz and microcline are also fractured and this has implication to the emplacement of the barite mineralization. The feldspathic Bima is non-kaolinized. The quartz and microcline crystals of both the granite and sandstones (sandstone host the barite mineralization) are fractured (Figure 3a & 3b), a situation which enhances the permeation of the hydrothermal solution through the host rock and the subsequent deposition of the sulphates in pre-existing (sandstone) fractures. The barite occurs as massive fracture filling veins (Daspan and Imagbe, 2010), while in the hydrothermally altered sandstones the minerals barite tends to be assimilated into silica matrix, they also fill up the pore spaces between the quartz and

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feldspars and within the fractures commonly found in these brittle minerals (barite), a silicification and hydrothermal activity/processes. The silicification and the hydrothermal activities came or started at the beginning of marine condition after the deposition of Bima Sandstone and Yolde Formation because Yolde Formation is partially silicified with Bima Sandstone on the Gombe Inlier especially at the flanks. The layer of Bima Sandstone on the inlier is too thin to be classified into Bima 1, 2 and 3 but it is suggestive to say that Bima 1 unconformably overlies the Basement Complex including the inlier (Haruna, 2013). In addition to the fractures, the hydrothermal solutions exploit cleavages and twin planes of microcline and the subsequent deposition of the sulphide (pyrite). The barite mineralization of the study area is most likely part of the northern extension of the main mineralization system of Pb-Zn- Ba-F metallogenetic belt of the Benue Trough as reported by several workers including Farrington (1952), Akande and Abimbola (1987), Wright (1989) and El-Nafaty (2015).

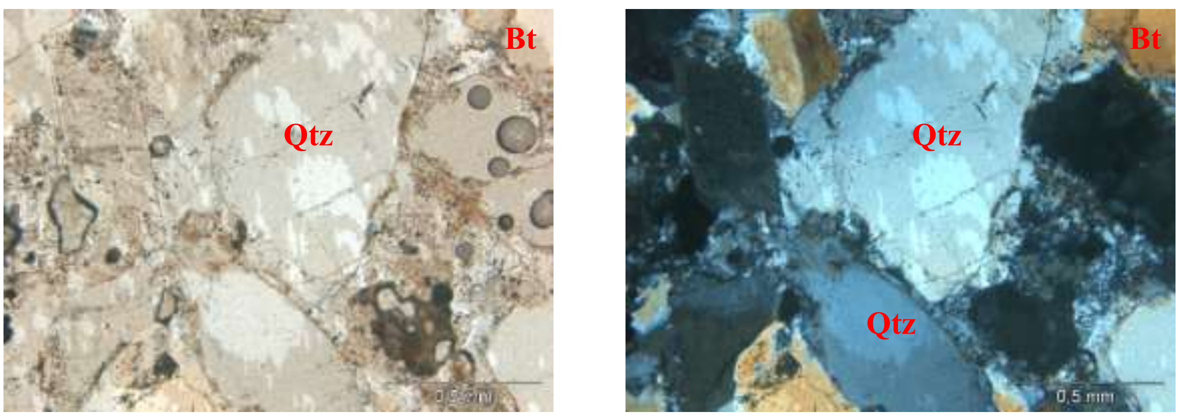

Fig. 3a (i) &(ii); Photomicrograph of Biotite Granite in Plain and Cross Polarized Light. Qtz = Quartz, Bt = Biotite.

Figure. 3b(i); Photomicrograph of a Sandstone sample in Plane Polarized Light. Qtz = Quartz, Slf = silicified or lithic fragments;

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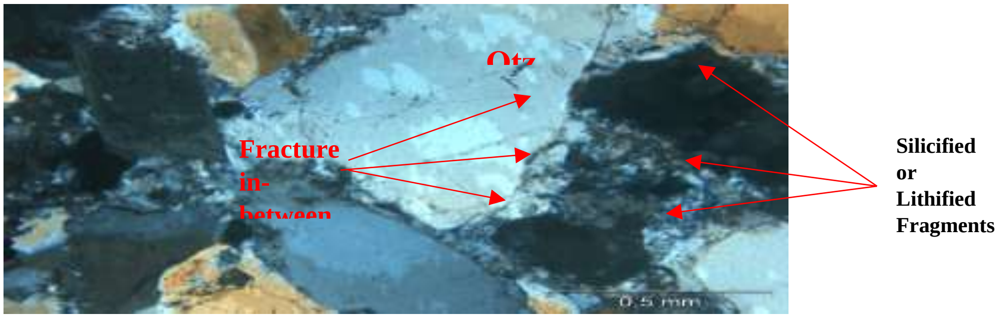

The petrographic studies shows that the megacryst of the barite were also recrystallized to blocky rectangular crystals of barite in the silica medium and in most cases, the megacryst of barite was assimilated into the matrix of silica either as fine grains or medium grains (Figure 4). This suggest that there could be another late hydrothermal fluid that invaded the Gombe hill and leached part of the barite and recrystallized them. The silicified sandstone contains a lot of cemented anhedral crystals of barite.

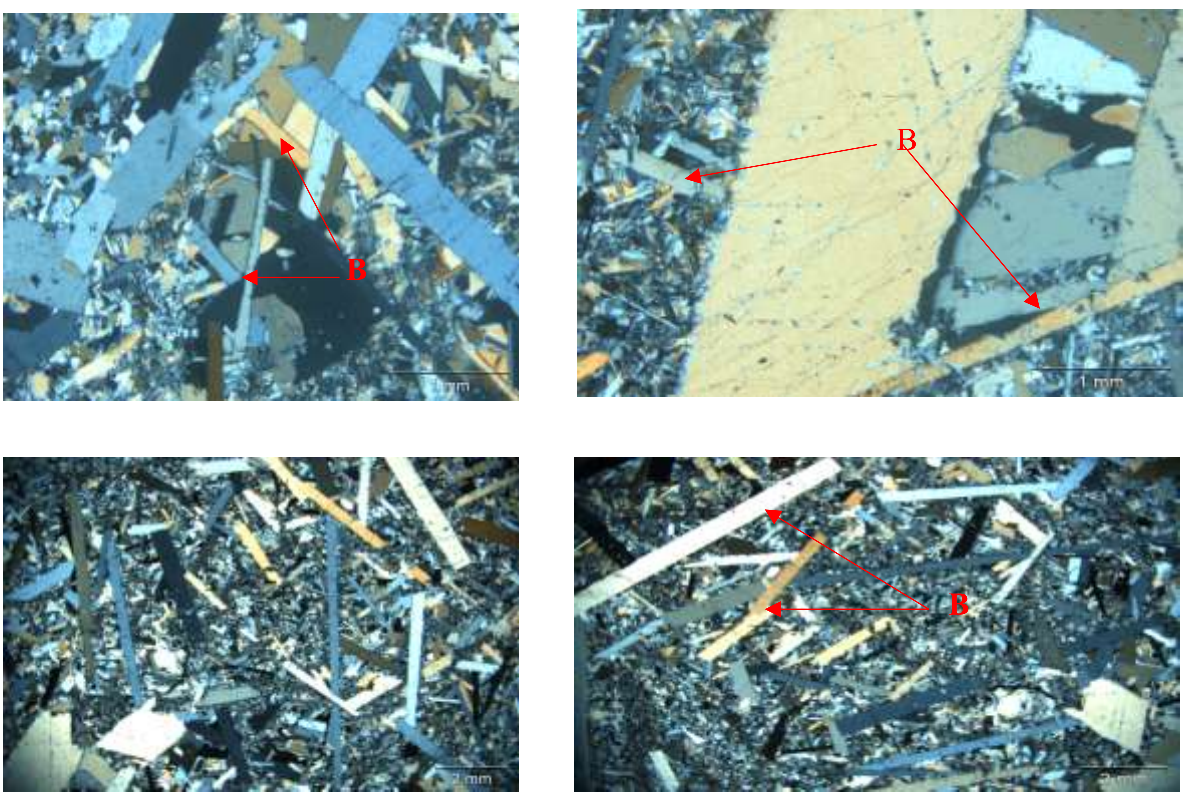

Figure. 4; Photomicrographs (I, II, III and IV) of a Sandstone sample in Cross Polarized Light. Notice Spindles of barite within disintegrated cementing matrix. Qtz = Quartz, Slf = silicified or lithic fragments and B = Barite.

MINERALIZATION

The barite deposits of the Gombe hill are massive and have a stratiform - epigenetic character. They are emplaced within sandstone veins. The mineralization occurs in sub-vertical open fractures of short extent rarely exceeding 50m. The fracture system bearing the mineralization is generally oblique to the main structural trend of the Benue Trough i.e. N60^0E, and the barite deposits show no evidence of metamorphism. Ore mineralogy is nearly pure barite and the gangue minerals are mainly coarse calcite, silica and trace amounts of celestine and alunite. The presence of silica cemented by barite is very obvious and pronounced. The contact between barite bodies and the silica is a narrow faded or washed-out alteration halo no bigger than 15 cm wide. This silicification is related to the barite mineralization (Akande, 1988). The barites are in aggregate forms which consist of fine-grained and euhedral crystals, 1 to 10 cm long with no apparent preferred orientation. The barite deposits are essentially massive and are devoid of vugs or other cavities (Plate i):

Plate i: Massive Barite Mineral devoid of vugs or cavities.

At the Queen's Facility for Isotope Research, Department of Geological Sciences, Miller Hall, Queen's University Kingston, Ontario, Canada, nine (9) veins of the Liji hills at Bicije, on the outskirts of Gombe town, were chosen to provide fresh white barite samples free of contaminants and impurities for sulphur and oxygen isotope analysis. All data were reported using standard permil notation (‰), calibrated to certified reference materials, and compared to the following international standards: Vienna Canyon Diablo Troilite (VCDT) for $\delta^{34}S$ and Vienna Standard Mean Ocean Water (VSMOW) for $\delta^{18}O$. The basis for precision in permil notation (‰) is the analysis of duplicate samples. According to primary or secondary standard analyses, accuracy (std.dev.), expressed in permil notation (‰), is 0.5‰ for $\delta^{18}O$ and 0.2‰ for $\delta^{34}S$.

(i)Sulphur: Samples were weighed into tin capsules and the sulphur isotopic composition measured using a MAT 253 Stable Isotope Ratio Mass Spectrometer coupled to a Costech ECS

4010 Elemental Analyzer. $\delta^{34}S$ values were calculated by normalizing the $^{34}S/^{32}S$ ratios in the sample to that in the VCDT international standard, values are reported using the delta (δ) notation in units of permil (‰) and are reproducible to 0.2 permil.

(ii)Oxygen (sulphates only): Samples are weighed into tin capsules and oxygen isotopic composition is measured using a MAT 253 Stable Isotope Ratio Mass Spectrometer coupled to a Thermo Scientific TC/EA High Temperature Conversion Elemental Analyzer. $\delta^{18}O$ values are calculated by normalizing the $^{18}O/^{16}O$ ratios in the sample to that in the VSMOW international standard. Values are reported using the delta (δ) notation in units of permil (‰), and are reproducible to 0.2 permil.

DISCUSSION

Sulphur is widely distributed in nearly all types of natural environments, viz. igneous, metamorphic and sedimentary rocks, organic substances, marine sediments and the oceanic water (Sharma *et al.*, 2006). It is one of the abundant elements in sea water, forming significant sulphate in the system. A general higher isotopic value for sulphur in sea water is because of heavier isotope partitioning for residual sulphate present therein, whereas lighter isotope of sulphur is fractioned into sulphides. d ^{34}S is low (0 ± 3‰) in primary magmatic sulphur in oceanic/submarine basalt (Field *et al.*, 1984) The widest range of d ^{34}S from –50 to +100‰ is found in sedimentary rocks. In the sediments of continental margins, platform areas and geosynclines, it is in the range of –12.5 to –17‰. In spite of a few exceptions like Kupersciefer, Germany (–5‰) and Eureka mine, New Mexico (–22 to –23‰) (Nielson, 1979), values of d ^{34}S are generally positive for sulphate of sea water/evaporate origin, as observed in many studies, including Leon Mexico (+12 to +17‰) (Kesler, *et al*., 1988), Seirra Del Guadarrma (+15‰) (Galindo, *et al.*, 1994), Aravalli (+17 to +21‰) (Deb, *et al.*, 1991) and Aberfeldy (+40‰) (Hall, *et al.*, 1991). Barite of the Gombe hill (present study) shows enrichment of isotopically positive sulphur, with d ^{34}S varying from +17.2‰ to +19.8‰ indicative of a modern sea water source of the sulphur (Table 1).

Table 1: Sulphur and Oxygen Isotopic Compositions of Barite minerals of the Gombe Hill Area.

Sample ID	Mineral Type	$\delta^{34}S$‰ (VCDT)	$\delta^{18}O$‰ (VSMOW)
LH2	Barite	19.7	10.3
LH3	Barite	19.3	11.2
LF4	Barite	18.1	12.2
LH1	Barite	19.5	10.7
LI2	Barite	17.9	10.1
LI3	Barite	17.2	10.5
LA2	Barite	19.8	11.3
LA3	Barite	19.1	9.9
LF1	Barite	18.4	12.1

This narrow range of data indicates that the barite-forming system was dominated by sulphate from a homogeneous source. This data rules out the possibility of involvement of any magmatic or sedimentary source for the sulphate, as well as sulphate derived from rivers or from isotope fractionation during mineral precipitate. Rather the data of d ^{34}S: +17.2‰ to +19.8‰ for the barite under study are well within the range suggestive of the Proterozoic Sea water. Claypool *et al*, (1980) demonstrated that d ^{34}S values for marine sulphate of different ages show variation with d ^{34}S values of sea water of about +30‰ during the Cambrian, probably reduced to +10‰ in the Permian. Later on, it increased irregularly during the Mesozoic and reached its present value of 20‰. Thus, substantial age information for the sulphate of marine origin can be obtained by Sulphur isotope geochemistry. Consequently, d ^{34}S values of the Gombe hill barite strongly favour involvement of Proterozoic Sea water for the deposition of barite.

Conversely, the source of sulphur (Figure 5) and the source of the metal barium (Ba) (Figure 6) has equally been deduced from the values (Table 1) and plotted as adopted from Rollinson (1993). These plots place the source of sulphur as originating from our Sea water and the metal barium (Ba) originating from the detrital sediments (Bima Sandstones) of the Gombe hill.

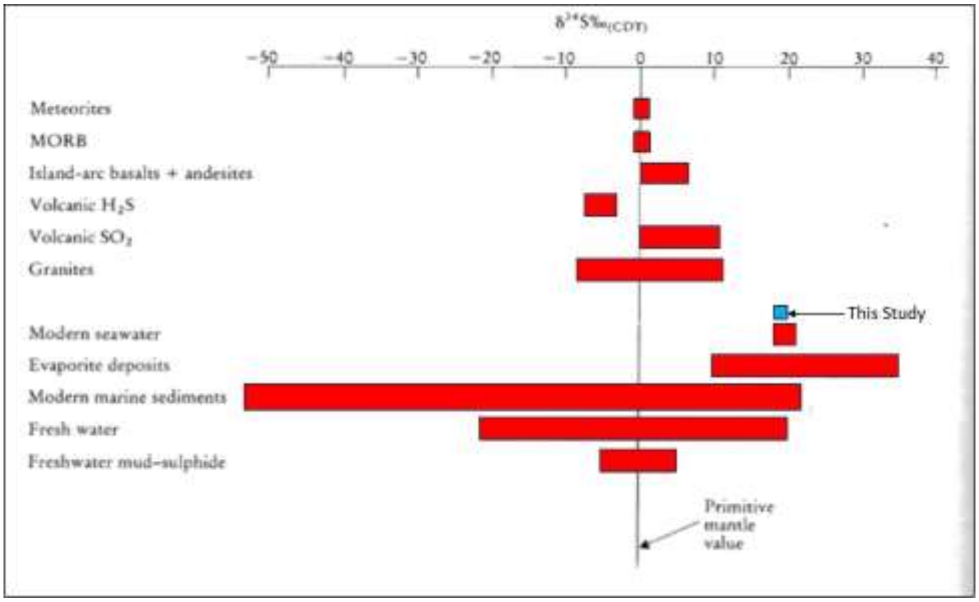

Figure. 5: Natural sulphur isotope reservoirs, barite values of the barite mineralization sub-system of the Gombe hill (modified from Rollinson, 1993).

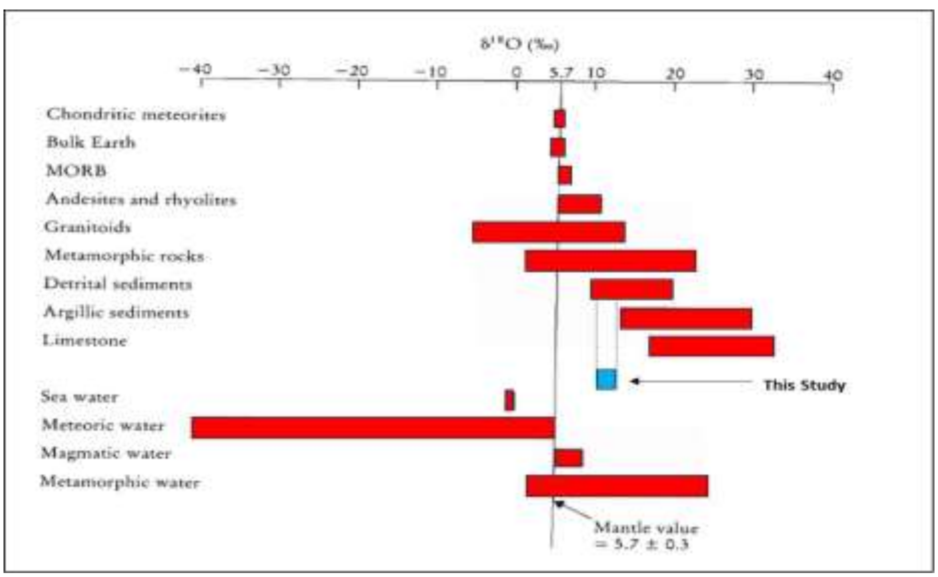

Figure 6: Natural oxygen isotope reservoirs, barite values of the barite mineralization sub-system of the Gombe hill (adopted from Rollinson, 1993).

It is suggested that Ba was derived from the leaching of silicate minerals present in the host rock, i.e. the Bima Sandstone. This agrees also with the conclusion of Ghosh (1991). Leaching of Ba from silicate minerals of the source area is plausible in view of the behaviour of Ba during fractional crystallization of magma, where Ba gets mostly confined to K-bearing minerals, such as feldspar and mica. Ba and K ions can replace each other at high P–T conditions, as they are isostructural. Consequently, 400–10,000 ppm Ba is found in granite, and K-feldspar in granite may contain up to 6% Ba (Smith and Brown. 1988). Hence, the Stable-isotope signatures of the Gombe hill barite present two distinct evidences. On this basis, we propose that the process of mixing of two fluids, viz. barium-rich crustal fluid and sea water sulphate, led to barite mineralization in the Gombe hill area.

CONCLUSION

This study shows that the hydrothermal fluid supplied the barium ions (Ba^{2+}) while the sea water supplied the sulphate ions (SO_4^{2-}). Evidences from petrography, suggest that there is another late hydrothermal fluid that invaded the Gombe hill and leached part of the barite and fluorite mineralization and recrystallized them. The silicified sandstone contains a lot of cemented anhedral crystals of barite. The stable sulphur ($\delta^{34}S$) isotope compositions range of 17.2-19.8‰ (CDT) of the sulphate (barite) indicate modern sea water source of the sulphur. Values arrived at, indicate that the source of the oxygen were from the sedimentary basin of the host rocks (detrital sediments). These isotopic signatures suggest mixing of sea water sulphate with Ba-carrying crustal fluid for barite deposits, with its initial deposition linked to the detrital sediments of the Bima Formation (host rock).

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