Geological Setting and the Role of Basinal Brines for the Origin of Barite and Fluorite Deposits of the Gombe Hill, N.E Nigeria

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ABSTRACT: *The opening of the Benue Trough as well as the sinistral displacement of the strike slip fault that displaced the whole inlier has been well documented in recent times. Fractures formed are believed to have occurred at the time of the opening of Benue Trough in the Cretaceous. Large sedimentary basins were developed with carbonates and continental platforms that provided ideal conditions for the formation of suits of sandstone hosted, stratiform deposits such as barite, celestine and fluorite of Cretaceous age. Barite is the economic mineral and the shape of the ore bodies is considered massive, the gangue minerals are calcite, traces of celestine and silica. A fluid inclusion and stable isotope analysis (S and O) for barite and fluorite were conducted. The result shows a melting ice temperature between −24 °C and −15 °C (salinities of 13.6 to 24 wt. % NaCl equiv.) and a homogenization temperature ranged between 60 °C to 155 °C. Isotopic analysis of barite showed* $\delta^{34}S_{VCDT}$ *ranges from +18.1‰ to +19.7‰ (average of 19.03‰), fluorite sulphur isotope value gives 17.7‰ to 19.6‰ (average of 18.6‰). Sulfur isotope data for the barite and fluorite from the study area is consistent with a sulfur source formed during the Cretaceous, which coincides with the age of the Bima Formation. The oxygen isotope analysis showed a range between* $\delta^{18}O_{VSMOW}$ *9.9‰ and 12.3‰ for both minerals. Fluid inclusion microthermometry and isotopic measurements lead us to conclude that brines from the Upper Benue Basin led to the replacement of the evaporite strata (gypsum) by barite and fluorite and its subsequent deposition within the veins and fractures of the Bima Formation.*

Keywords: barite, basinal brines, sinistral, displacement, paleogeographic, stable isotopes, microthermometry.

INTRODUCTION

The ore deposit types which were likely formed during the Cretaceous in the sedimentary domain of northeastern Nigeria are: The manganese and barite deposits of Hawal Massif [bounded by tertiary sediments of the Chad Basin to the north, Cretaceous sediments of the Benue Trough to the south (which separates Hawal Massif from Adamawa Massif)] in Mubi area (Vandi and Benkhelil, 2019); the barite and anhydrite deposits of the Gombe hill in Gombe state (Haruna, 2013); barite deposits such as that of Gulani and the Gombe inlier, in Yobe and Gombe states (Haruna, 2013 and El-Nafaty, 2015); limestone of Ashaka in Gombe state (Udeh, 2021); gypsum deposits of Fika, Ngalda and environs, all around Potiskum – Yobe State (Bukar *et al.*, 2011; Jazuli and Moisule, 2015); Pb- Zn sulphides, such as (Sharma *et al.*, 2006;Saleh *et al*., 2024) and uranium deposits in detrital sequences of the Guburunde horst and around (Haruna *et al.*, 2017, Haruna, 2017). With the exception of the manganese deposit, which is largely syngenetic, these deposits are epigenetic and occur in basins of Mesozoic – Cenozoic age associated with the opening of the Benue Trough, with the majority of them in the states of Yobe, Gombe and Adamawa (Figure. 1). It is pertinent to note that none of these deposits have been properly dated and the stratigraphic correlation suggests a relative age for these deposits that ranges from Mesozoic to Cenozoic. Despite the lack of geochronological determinations for the sedimentary deposits, we may, nevertheless, speculate about a tentative timing for the deposition of sediment-hosted ore deposits relative to the orogenic pulses in the region (González-Sánchez *et al.*, 2009 and 2015), suggesting a possibility of either Pre- Cretaceous or Syn- Cretaceous deposits.

The clearest regional anatomy within the basin (Upper Benue), for which certain authors (Saleh *et al*., 2024, Olade and Morton, 1985, Olade,2019 and El-Nafaty, 2015), shows a preferential distribution of the different mineralogical types of the associated deposits as follows: 1) Pb–Zn and barite occur deep in the basin, and are formed from the hottest and most saline mineralizing brines within the whole axis of the Benue Trough; and 2) Fluorite deposits occur in the basin at shallower stratigraphic sections and are associated with dilute and relatively cool fluids.

A clear example of the first case above is the barite deposits of the Gombe inlier. The mining area is located at the outskate of Gombe town in Bicije area, along Biu Road. The barite deposits have been mined artisinally for about a decade now, and they are localized on broad anticlinal structures such as the Gombe inlier. 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. They are massive stratiform bodies composed of high-purity barite and smaller, non-economic ore bodies of pyrite occurrences, hosted by the Pre-Cambrian granite and occurs within highly fractured zones located very close to the Granite/Bima sandstones contacts. The extent of this zones ranges from 15m-25m and 4m-5m wide.

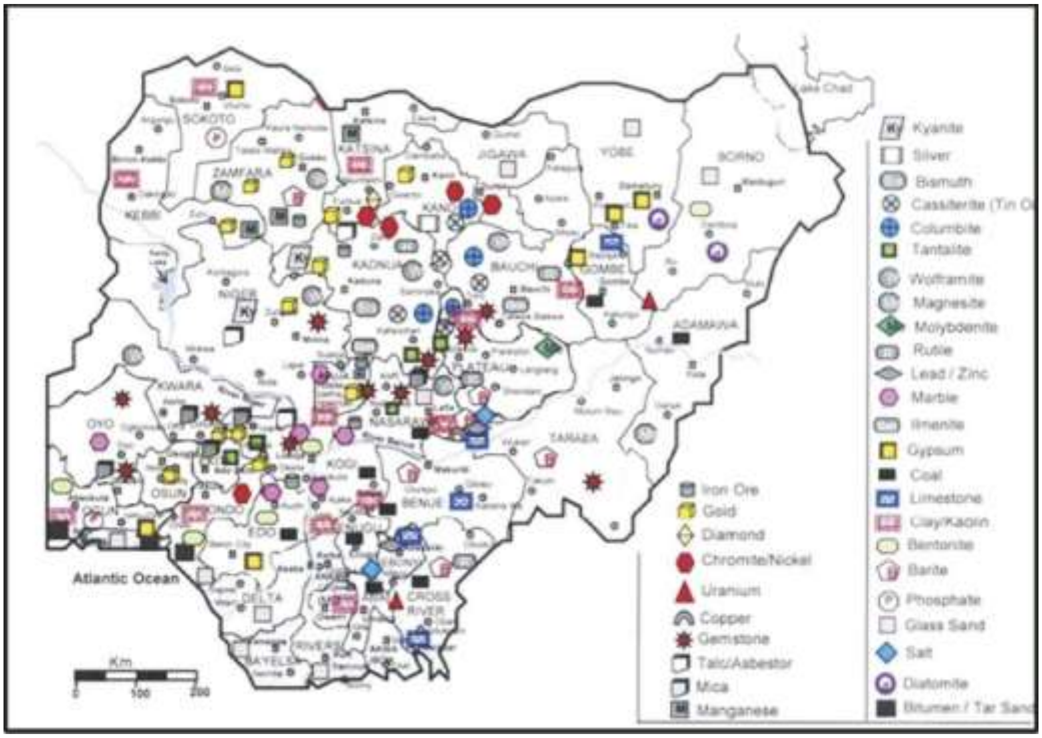


Figure 1: Map Showing Mineral Occurrences in Nigeria (After Olade, 2019).

The data provided based on geology, micro thermometry and isotope provided in this paper is geared towards defining the role played by basinal brines for the origin of barite deposits of the Gombe inlier, Northeastern Nigeria.

Geology and Setting

Structural and Paleogeographic features of the Upper Benue Trough during the Mesozoic (Cretaceous) was responsible for; 1) The opening of the Atlantic due to extension related to the break-up of Gwandwanaland at the Cretaceous and the rifting apart of the Gombe inlier block into a sinistrial displacement strike-slip blocks that provoked the subsequent formation of the several faults, fractures and veins which serves as conduits for mineralization fluids and structures for the barite mineralization. 2) This opening of the Atlantic determined the formation and architecture of the Upper Benue Trough, among others, with subsequent development of broad sedimentary platforms on raised blocks during the Cretaceous, which was responsible for formation of lithological units of carbonates and evaporites.

The Gombe inlier is bounded by the evaporitic deposits of gypsum to the West around Nafada, Fika and extending towards Ngalda, North of Potiskum in Yobe state, the younger Tertiary volcanic of Kaltungo across the Yola arm and volcanic of Biu to the East and North respectively. The rocks of the Basement Complex of the North-East lie to the South of the inlier around Kirfi-Alkaleri across into Bauchi, the home of the famous Bauchites.

Such merits and demerits of paleo-geographic features were limited by normal faults (in a horst and graben arrangement), anticlines and synclines. These features control emplacement of sedimentary brines into portions of the stratigraphic section, wherein the formation of gypsum, fluorite, anhydrite and barite occurred (González-Sánchez *et al.*, 2009; Alonso- García *et al.*, 2011).

The Benue Trough is geographically subdivided into Lower, Middle and Upper (Figure 2) Troughs (Benkhelil, 1989, Abubakar *et al.*, 2008). It is a sedimentary basin extending from the Gulf of Guinea in the South to the Chad Basin in the North. The origin and tectonic history of the Benue Trough is associated with the break-up of the continents of Africa and South America (the break-up of Gwandwanaland). This break-up was followed by the drifting apart of these continents, the opening of the South Atlantic and the growth of the Mid-Atlantic Ridges.

The Upper Benue Trough is also generally divided into three (3) sub-basins (Benkhelil,1989 Zaborski, 1997); namely, the Lau/Gombe Sub-basin, the Yola Sub-basin and the Gongola Sub-basin. Sedimentation in the Lau/Gombe Sub-basin began with the deposition of the continental Bima sandstones in the Albian. This was followed in the Cenomanian by the deposition of the transitional Yolde Formation and in the Turonian by the deposition of marine sediments of the Pindiga Formation. A period of unconformity followed after the Turonian, in the Mid-Santonian, by the deposition of Gombe sandstones in the Maastrichtian. Sedimentation ended with the deposition of the continental Kerri-kerri Formation in the Paleocene. In the Yola Sub-basin, sedimentation started or began with the deposition of the Bima sandstones in the Albian, transitional Yolde in the Cenomanian and marine Dukkul/Jessu/Sekuliye/Numanha/Lamja in the Turonian. Then, during the Santonian, there was folding, faulting, uplift and hiatus which were followed by intensive magmatism and volcanism. In the Gongola Sub-basin, sedimentation began with the continental Bima sandstones, followed by the Cenomanian Yolde which is in this area positively (+) or negatively (-) present. Overlying the Bima and/or the Yolde in the Gongola Sub-basin is the marine- Cenomanian- Turonian Gongila Formation. On top of the Gongila (Ashaka area) is Late Turonian – Coniacian Fika shales. This was followed by folding, faulting and uplift. Then began the deposition of the of the continental Gombe sandstones in the Maastrician and lastly the Kerri-Kerri Formation in the Paleocene.

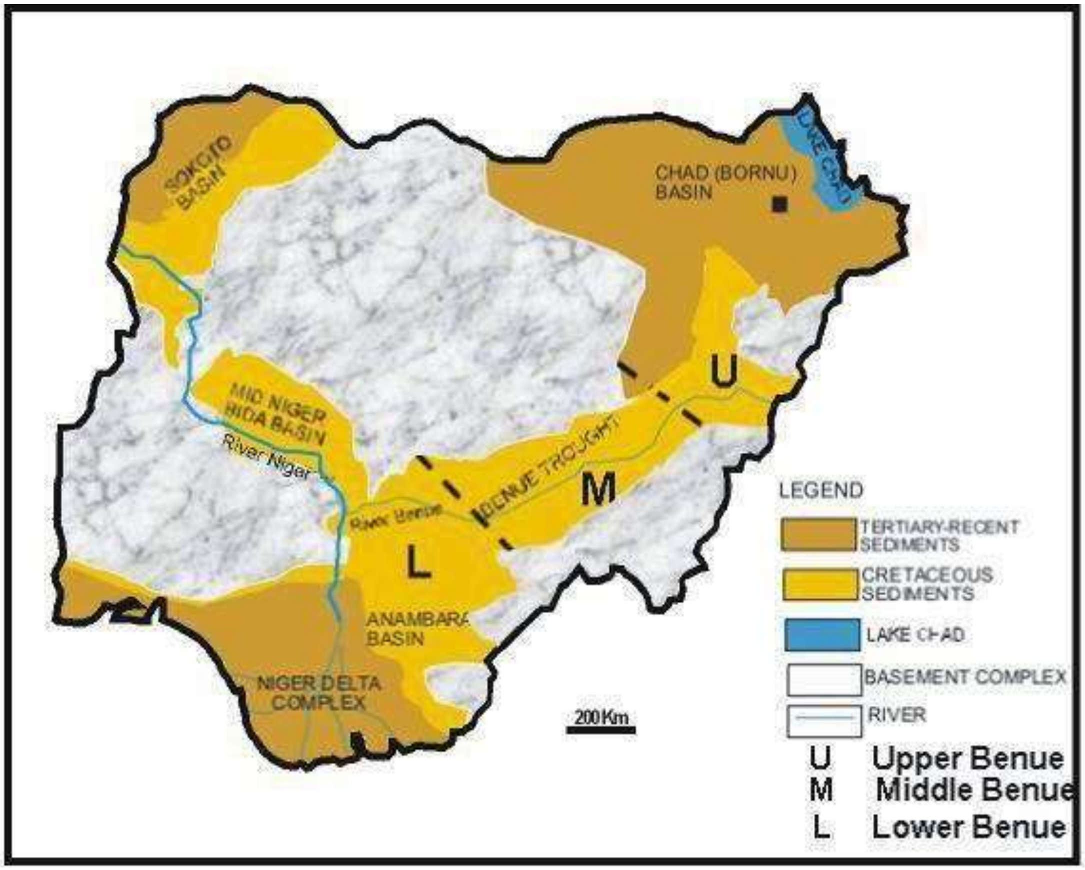


Figure. 2: Geological Map of Nigeria showing the Benue Trough (After Abubakar et al., 2008)

Gypsum occurs abundantly within the shales in the Fika and Pindiga Formations of the Upper Benue Trough. Carbonates in form of limestones occur extensively throughout the length and breadth of the Benue Trough where they are being exploited for cement manufacturing. In the Upper Benue Trough, the limestones in the Gongila Formation are being exploited for cement manufacturing in the Ashaka cement factory at Ashaka, Gombe State. There have been reported occurrences of hydrocarbons in the Anambra Basin and in the Gongola Basin.

MINERALIZATION

The barite deposits of the Gombe inlier 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^{0}E, 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 *et al.*, 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 (Figure. 3). The Cretaceous barite veins are generally stratiform; therefore, the deposits should be sought at the stratigraphic level of the Albian Bima sandstone where the mineralization is confined. This is consistent with any mineralization in many geological environments.

Figure 3: Massive Barite Minerals.

Stable Isotopes Studies and Fluid Inclusion Microthermometry Analysis

Nine (9) barite and six (6) fluorite samples were analyzed for their sulphur and oxygen isotope values (Table 1). The sulphur isotope (δ^{34}S) values expressed in per mil (‰) relative to Canon Diablo Troilite of sulphate (barite) yield δ^{34}S compositions with a short range of 18.1 – 19.8‰ (CDT) for the nine samples (Table 1), averaging 18.7‰. The plot of these values is presented in (Figure. 4) alongside the standard plots reported by Rollinson, (1993). These values indicate that the source of the sulphur and by extension the mineralizing fluids were from the modern seawater.

The oxygen isotope ($\delta^{18}O$) values also expressed in per mil (‰) relative to standard Mean Ocean Waters of sulphates (barite) yields $\delta^{18}O$ compositions with a short range of 10.1 – 2.2‰ (SMOW) for the nine samples, averaging 10.9‰. The plot of these values is presented in (Figure 5) alongside the standard plots reported by Rollinson, (1993). These compositions reflect formational influences of the detrital sediments which are known to have formed under fluviatile environment of deposition. These values therefore, indicate that the source of the oxygen were from the sedimentary basin of the host rocks.

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

Sample ID	Mineral Type	$\delta^{34}S$‰	$\delta^{18}O$‰
LH2	Barite	(VCDT) 19.7	(VSMOW) 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
LB1	Fluorite	17.7	10.8
LE3	Fluorite	18.9	11.2
LE4	Fluorite	18.4	12.1
LE5	Fluorite	17.9	12.3
LE6	Fluorite	19.6	9.9
LK4	Fluorite	19.1	11.4

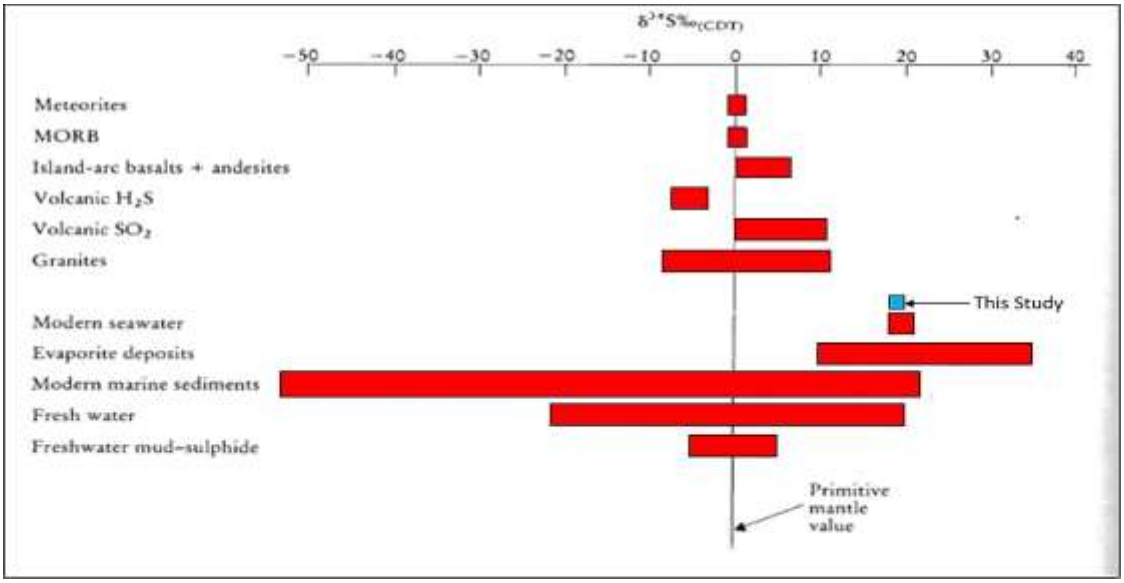


Figure. 4: Natural sulphur isotope reservoirs, barite values of the barite mineralization sub-system are also plotted (modified from Rollinson, 1993).

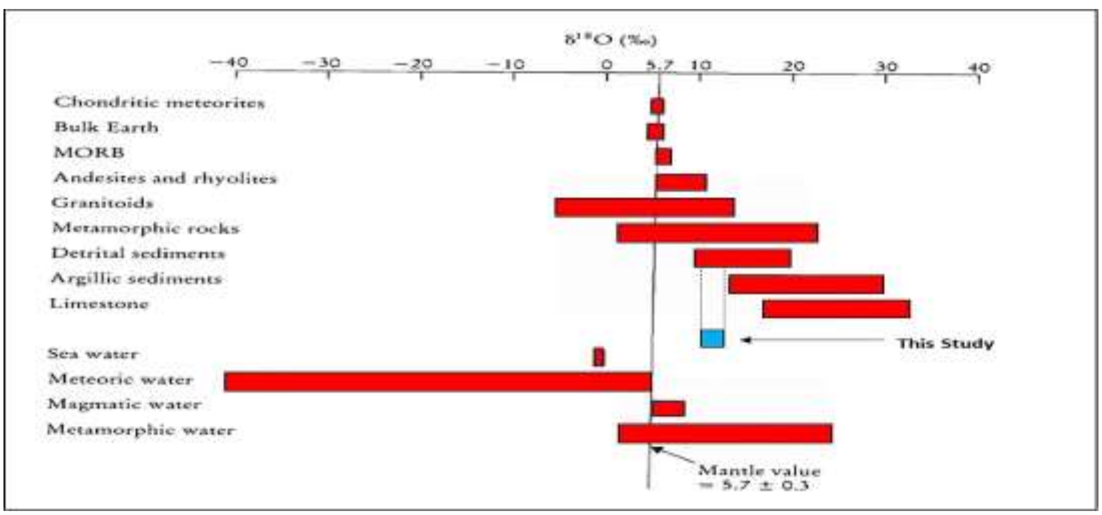


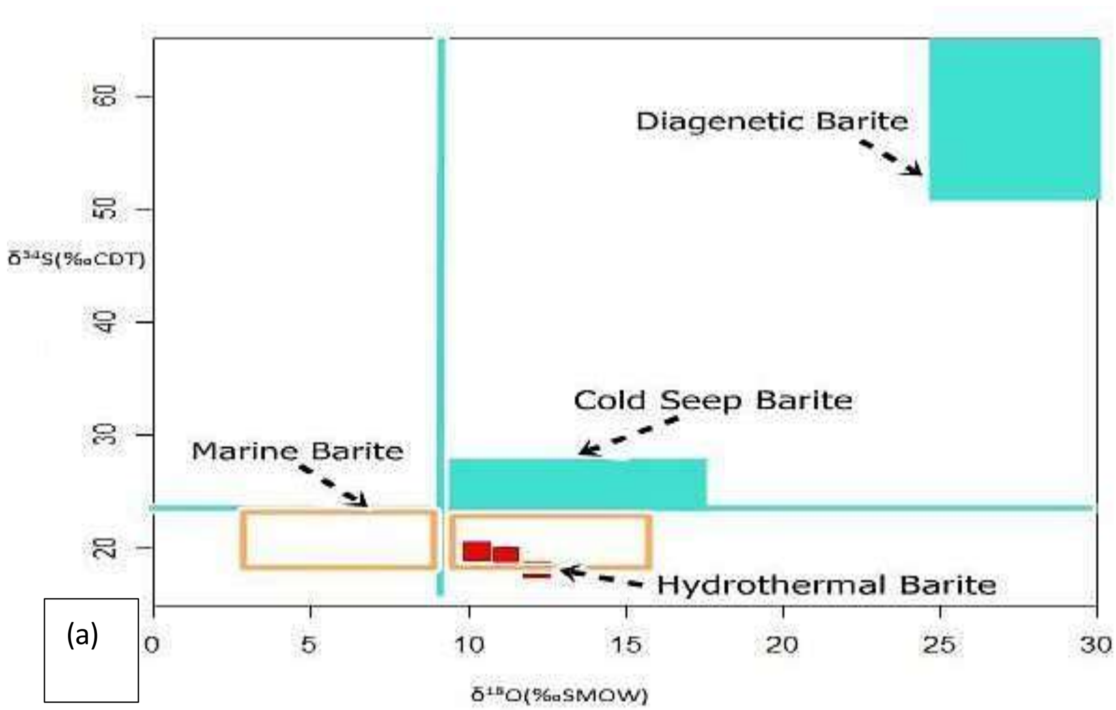
Figure.5: Natural oxygen isotope reservoirs, barite values of the barite mineralization sub-system are also plotted (adopted from Rollinson, 1993).

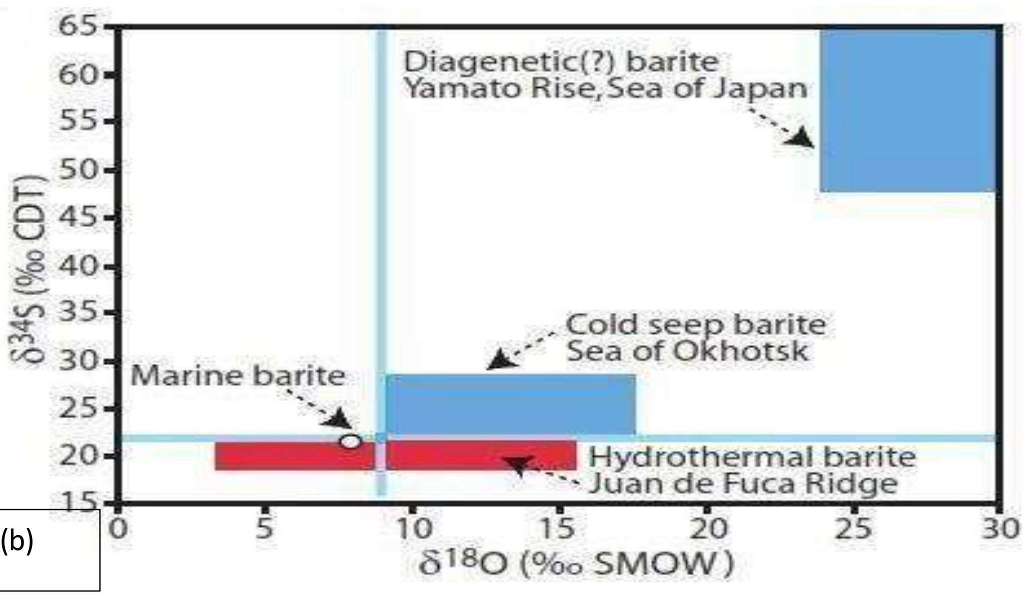
Sulphur isotopic composition ($\delta^{34}S$) of barite is quite similar to the SO_4 from which it precipitated. Accordingly, barite $\delta^{34}S$ will record the sulphur isotopic compositions of the formation fluids.

Depending on the mode of barite formation, several potential sources of SO_4 may be available for barite precipitation, including sea water SO_4, magmatic sulphate, pore water SO_4 modified by microbial reduction, SO_4 from calcium sulphate minerals and SO_4 produced by the oxidation of reduced sulphur species.

The sulphur isotopic signature in barite can be used to distinguish the mode of barite formation, and in view of this, (Figure. 6) is a plot of $\delta^{34}S$ vs $\delta^{18}O$ which discriminates barites into different type depending on their sulphur and oxygen isotopic compositions (diagenetic barites, marine barites, cold seep barites and hydrothermal barites). Figure. 6 a therefore, placed the barite of the Gombe in- lier as hydrothermal barite type.

Figure. 7 **is a** plot of Sulphur and Oxygen-isotopic composition of barite from modern continental margins. The isotopic enrichments of the Gombe barite are relative to normal seawater sulphate [Figure. 7 (a)].

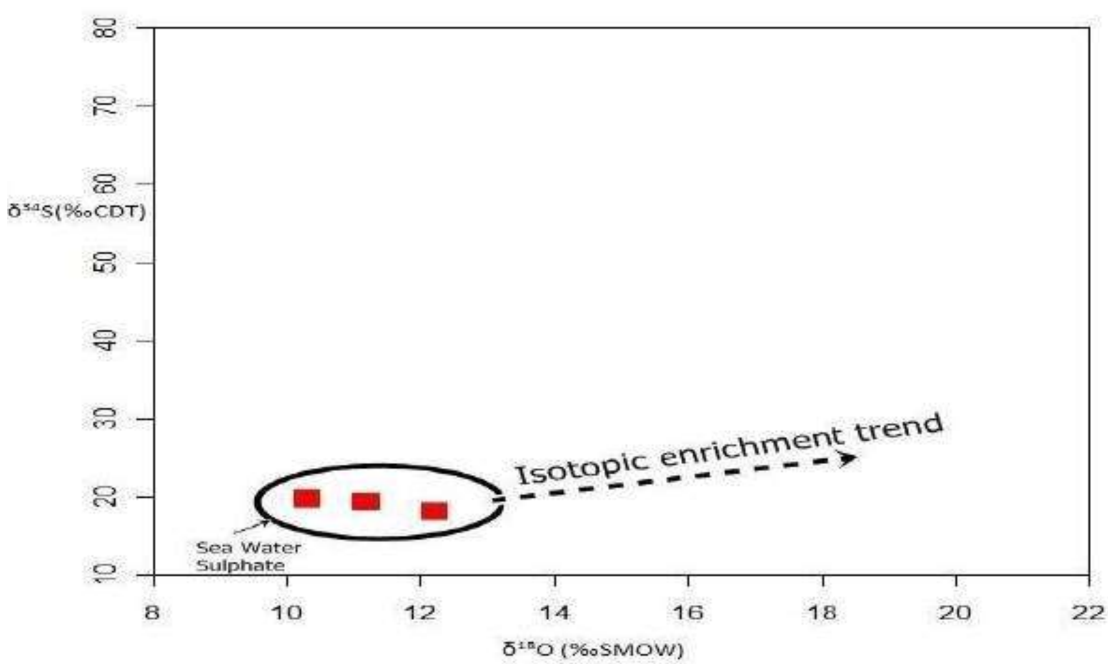




(b)

Figure. 6: Plot of the Sulphur and Oxygen Isotopic Composition of; (a) Barite samples of the Gombe Hill, (b) Juan de Fuca O and S isotope data (Good fellow *et al.*, 1993 and Paytan *et al.*, 2002), respectively. Sea of Okhotsk data (Greinert *et al.*, 2002), Sea of Japan data (Turchyn and Schrag, 2004). Average Marine barite data (Turchyn & Schrag, 2004).

(a)



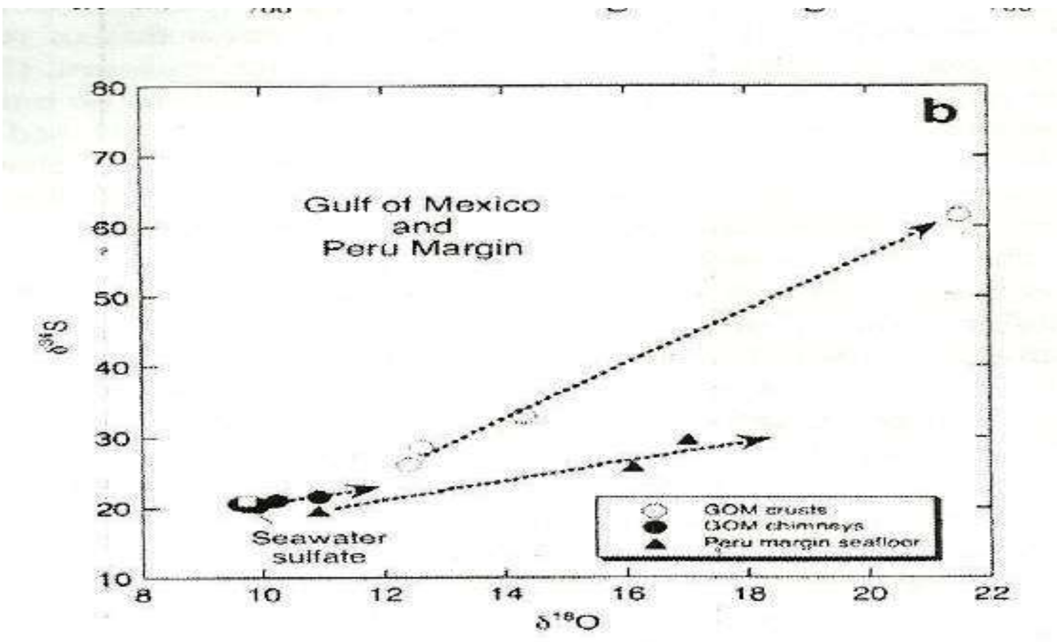


Figure 7: Sulphur and Oxygen-isotopic composition of barite from modern continental margins: (a) Barites of Gombe Hill deposited within veins, (b) Barites deposited beneath the sea floor as cements or crust show large isotopic enrichment relative to normal seawater sulphate.

Table 2: Results of Fluid Inclusion Studies of Barite and Fluorite Mineralization of the Study Area (Gombe Hill).

Barite	LF4	60 -155	123	-15 to -24	-18.2	13.6 - 24
	LH3	60 -155	120	-15 to -24	-20	14 - 24
	LH2	70 – 155	126	-15 to -24	-20.3	13.7 - 24
Fluorite	LE6	137 – 141	139	-7.5 to -15.5	-11.5	14 - 24
	LE5	137 - 142	135.9	-17 to -21	-19.0	14 - 24
	LE4	125 – 129	127	-18 to -22	-20.4	14 - 24

S/No.	Material	Sample I.D	Results of Fluid Inclusion Range Th (^{0}C)	Average Th (^{0}C)	Range Tmi (^{0}C)	Average Tmi (^{0}C)	Salinity (wt% NaCl Equiv.) (Range)
1	Gombe Hill UBT, Study Area						
2	Gombe UBT, Study Area						

Th = Homogenization Temperatures; Tmi = Melting Ice Temperatures; UBT = Upper Benue Trough; Equiv. = Equivalent.

The results of fluid inclusion studies of three barite separates/mineral-grain shows that the diameters of the inclusions vary from 1 to 5 µm. Most of the primary fluid inclusions homogenize to liquid between -15 and -24 °C (Table 2). They have a homogenization temperature range of 60^{0} - 155^{0}C. The salinity of the fluids estimated using freezing temperatures of the two-phase [liquid &vapour] range from 14^{0} to 24^{0} equivalent wt.% NaCl. The fluid inclusion data show that the mineralizing fluids were saline brines containing recognizable Na^{++}, Ca^{++} and Cl^{-} ions.

The three fluorite samples LE4, LE5 and LE6 are also primary inclusions that contain two phases (vapour – liquid). The shapes of the inclusions are also rounded or rectangular like that of the barite inclusion. They occur solitarily or as clusters (Figure 8) in the inner and outer structures of the fluorite mineral-grain. The diameters of the inclusions vary also from 1 to 5 µm. Most of the primary fluid inclusions homogenizes to liquid between -7.5 and -22 °C (Table 2). They have a homogenization temperature range of 125^{0} - 142^{0}C. The salinity of the fluids estimated using freezing temperatures of the two-phase fluid inclusions range from 14^{0} to 24^{0} equivalent wt.% NaCl.

Projection of the fluid inclusion data on a temperature vs. salinity diagram (Figure 9) enables identification of a fluid of relatively to moderate low temperature and variable salinity fluid. The above results place the barite and fluorite of the study area as belonging to the low temperature environment and a moderate salinity surface-derived fluid occurring in the vein systems.

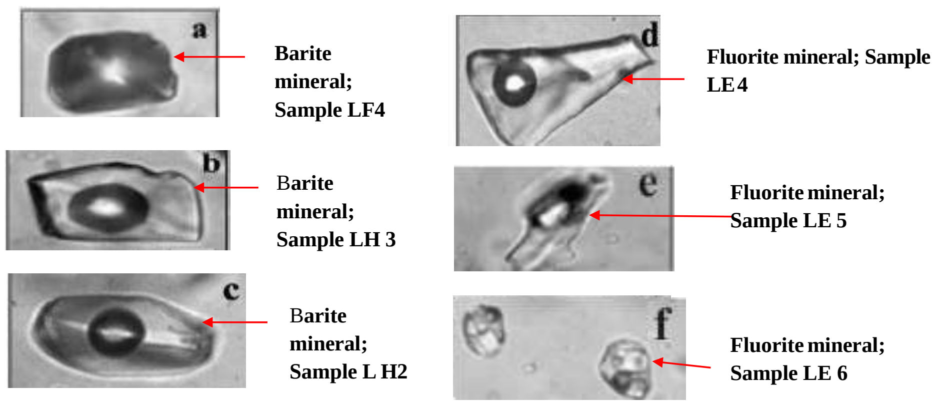


Figure 8: Primary Inclusion with rounded to rectangular shapes in Barite Sample (a– c) and fluorite samples (d – f)

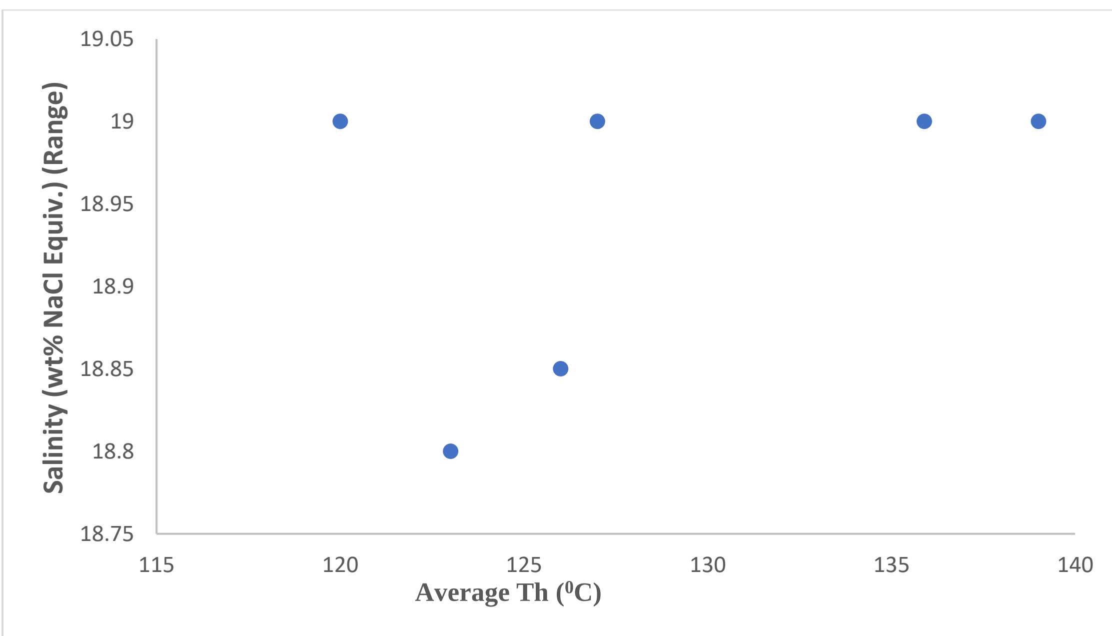


Figure 9: Homogenization Temperature (Th) vs. Salinity Diagram for Fluid Inclusion Data of Barite and Fluorite Mineralization of the Study Area (Gombe Hill).

DISCUSSION

Gombe hill is a basement dome that exposes most of the sequence of rocks in the Gongola basin of the Upper Benue Trough from the oldest Bima Sandstone to Gombe Sandstone. The sedimentary sequence forms an off-lapping relationship from the inlier with the oldest rock (Bima Sandstone) on the inlier and the youngest (Gombe Sandstone) farther away from the inlier. The inlier was faulted during the division of the Gondwanaland in the Cretaceous to form a strike-slip fault. The offset of the faulted blocks forms a sinistral strike- slip fault. This movement is consistent with model for the opening of Benue Trough as a 'pull-apart' basin (Benkhelil, 1986 and 1989, Maurin *et al.,* 1985).

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. This narrow range of data shows that Ba-rich solutions sub-systems were dominated by sulphate from a homogenous source (Sharma *et al.,* 2006). The above result rules out the magmatic source of the sulphur from the Basement inlier and nearby Tertiary/Quaternary volcanic rocks of the Biu Plateau as well as sulphur source from the local country rocks around the inlier. Garba (2000 a & b) in a similar situation has reported stable sulphur ($\delta^{34}S$) compositions of the range 1.5-9.4‰ (CDT) that reflect a metamorphic fluid sulphur source for the Bin Yauri Au mineralization, NW Nigeria, ruling out the magmatic source of the sulphur despite the proximity of the gold mineralization to a granitoid.

The stable oxygen ($\delta^{34}S$) isotope compositions range of 9.9-12.2‰ (SMOW) of the sulphate (barite) for the nine samples reflect formational influences of the detrital sediments which are known to have formed under fluviatile environment of deposition. These values therefore, indicate that the source of the oxygen were from the sedimentary basin of the host rocks.

The above conclusions for sources of sulphur (barite and fluorite) and oxygen (barite and fluorite) conform to the views of Rye and Ohmoto (1974) and Ohmoto and Rye (1979) who grouped the hydrothermal ore deposits into three categories:

(a) Deposits with $\delta^{34}S$ values near zero should derive their sulphur from igneous sources, including sulphur released from magmas and sulphur leached from sulphides in igneous rocks.
(b) Deposits with $\delta^{34}S$ values near 20‰ should derive their sulphur from ocean water.
(c) Deposits with $\delta^{34}S$ values between 5‰ and 15‰ may receive their sulphur from local country rocks or from mixture of (a) and (b).

Therefore, the barite with values of 17.2-19.8‰ derived their sulphur source from the modern sea water. Akande and Abimbola (1987) and Akande *et al*. (1989), reported a $\delta^{34}S$ values of -10 to 21‰ in sulphide minerals from the Pb-Zn-F-Ba mineralization of the Benue Trough, implying ore solution of probable evaporitic environment and sulphur of basinal source.

The fluid inclusion data show that the mineralizing fluids were saline brines containing recognizable Na, Ca and Cl and that fluid temperatures ranged between 60^0 - 155^0 °C for barites and 125^0 – 142^0

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for fluorite °C (low temperature conditions).

Previous genetic hypotheses suggested for the Benue ore fluids include (1) a magmatic hydrothermal origin (Farrington 1952), (2) formation from juvenile and connate brines (Ofodile 1976), and (3) circulating connate brine source (Olade and Morton 1985). The lack of close spatial association of igneous intrusions with Upper Benue veins except for the pegmatite dikes of the study area (Upper Benue) makes the first hypothesis untenable. The present data also refute the second hypothesis in view of the lack of igneous affiliations and the fluid inclusion data. However, brine circulation could have taken place under the influence of a shallow convective system driven by magmatic intrusions. Although our data suggest that the ore components were probably derived from compaction and brine release in the basin, the deeply circulating convection cells model of evolution (Russel 1978) for sediment-hosted Irish base-metal mineralization seems attractive to this mineralizations. However, the abundance of distinct epigenetic features and lack of syngenetic ores in the study area argue against a syn- sedimentary mode of origin. Lastly, the model of Russell (1978) applies mainly to syngenetic exhalative ore deposition, it is also therefore incompatible with the precipitation of sulfides in steeply dipping fractures. This study favors a basinal-brine expulsion model for the Upper Benue ore fluids. Sudden dewatering accompanying over pressures in sedimentary basins (Bethke 1984) could be the mechanism for the expulsion of metal-bearing fluids through permeable fractures in the Cretaceous sediments. The clear stratigraphic distribution of the Benue veins within Albian sediments (the Bima Sandstones) and their absence in the overlying Turonian sediments (Yolde Formation) suggest that the Cenomanian tectonism and uplift in this part of the trough probably initiated fractures through which the metal-bearing brines were expelled to form the barite and fluorite deposits.

CONCLUSION

This research work has provided some far-reaching answers to the questions raised, some of which include;

1. The barite and fluorite mineralization took place under oxidizing conditions as a result of reaction between the ascending hydrothermal fluid through the deep-seated fractures and the sea water that flowed into the fractures.
2. The hydrothermal fluid seems to have supplied the barium ions (Ba^{2+}) and the calcium ions (Ca^{2+}) while the sea water supplied the sulphate ions (SO_4^{2-}).
3. The high density of the hydrothermal fluid favours precipitation of barite while low density of hydrothermal fluid would favour the precipitation of fluorite.
4. Fluid inclusion and isotope data from the Benue Trough have shown that the mineralizing fluids were sulphate-rich brines having evolved from evaporitic sequences with fluid temperatures ranging from 60^o - 155^0C for the mineralizations.
5. The stable sulphur ($\delta^{34}S$) isotope compositions range of 17.2-19.8‰ (CDT) of the sulphate indicate modern sea water source of the sulphur.
6. Values arrived at, indicate that the source of the oxygen were from the sedimentary basin of the host rocks (detrital sediments).

7. The fluid inclusion data show that the mineralizing fluids were saline brines containing recognizable Na, Ca and Cl and that fluid temperatures ranged between 60^0 - 155^0 °C for barites and 125^0 – 142^0 for fluorite °C (low temperature conditions).
8. This study favors a basinal-brine expulsion model for the Upper Benue ore fluids.
   i. The sudden dewatering accompanying over pressures in sedimentary basins is the mechanism for the expulsion of metal-bearing fluids through permeable fractures in the Cretaceous sediments.
9. The Cenomanian tectonism and uplift in this part of the trough probably initiated fractures through which the metal-bearing brines were expelled to form the barite-fluorite deposits.

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