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Constraining the Genesis of Barite Deposits of the Gombe Hill, Using Stable Isotopes and Fluid Inclusion Studies

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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 was conducted. The result shows a melting ice temperature between −24 and −15°C (salinities of 13.6 to 24 wt. % NaCl equiv.) and a homogenization temperature ranged between 60 to 155°C. Isotopic analysis of barite showed $\delta^{34}S_{VCDT}$ ranges from +18.1‰ to +19.8‰. Sulfur and Oxygen isotope data for the barite 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 12.2‰ for the mineral. 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 its subsequent deposition within the veins and fractures of the Bima Formation.

Keywords: Barite, sinistral, displacement, paleogeographic, stable isotopes, microthermometry

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Introduction

In the sedimentary domain of northeastern Nigeria, the following types of ore deposits were most likely formed during the Cretaceous: the manganese and barite deposits of the Hawal Massif (which is bounded to the north by tertiary sediments of the Chad Basin and to the south by Cretaceous sediments of the Benue Trough (which separates Hawal Massif from Adamawa Massif)) in the Mubi area [1]; the barite and anhydrite deposits of the Gombe hill in Gombe state [2]; barite deposits such as those of Gulani and the Gombe hill, in Yobe and Gombe states [2, 3]; the limestone of Ashaka in Gombe state [4]; the gypsum deposits of Fika, Ngalda, and environs, all around Potiskum – Yobe State [5, 6]; Pb- Zn sulphides, such as [7, 8] and uranium deposits in detrital sequences of the Guburunde horst and around [9, 10]. These deposits, most of which are in the states of Yobe, Gombe, and Adamawa, are epigenetic and occur in basins of Mesozoic to Cenozoic age connected with the entrance of the Benue Trough, with the exception of the manganese deposit, which is primarily syngenetic (Fig. 1). The stratigraphic correlation indicates a relative age for these deposits that varies from Mesozoic to Cenozoic, and it is important to remember that none of these deposits have been accurately dated. Even though the sedimentary deposits have not been geochronologically determined, we can still make educated guesses about when sediment-hosted ore deposits might have been deposited in relation to the region's orogenic pulses [11–13], which could indicate either Pre-Cretaceous or Syn-Cretaceous deposits.

According to some writers [8, 14], the Upper Benue basin's most distinct regional anatomy displays a preferential distribution of the various mineralogical types of the associated deposits in the following ways: 1) Fluorite deposits occur in the basin at shallower stratigraphic levels and are associated with diluted and relatively cool fluids; 2) Pb–Zn and barite exist deep inside the basin and are generated from the hottest and most salty mineralizing brines across the entire axis of the Benue Trough.

The barite deposits of the Gombe hill are a glaring illustration of the first scenario mentioned above. The mining area is situated along Biu Road in the Bicije district, on the outskirts of Gombe town. The barite resources, which are located on wide anticlinal features like the Gombe hill, have been mined artificially for the past ten or so years. The mineralization is found in small, subvertical, open fractures that are rarely longer than 50 meters. The primary structural trend of the Benue Trough, N600E, is typically oblique to the fracture system that contains the mineralization. Located in severely fractured zones near the Granite/Bima, these enormous stratiform bodies are made up of smaller, non-economic ore bodies containing pyrite occurrences and high-purity barite. The Pre-Cambrian granite hosts them.

The purpose of the geology, microthermometry, and isotope data presented in this research is to determine the part basinal brines played in the formation of the barite deposits in the Gombe hill in northeastern Nigeria.

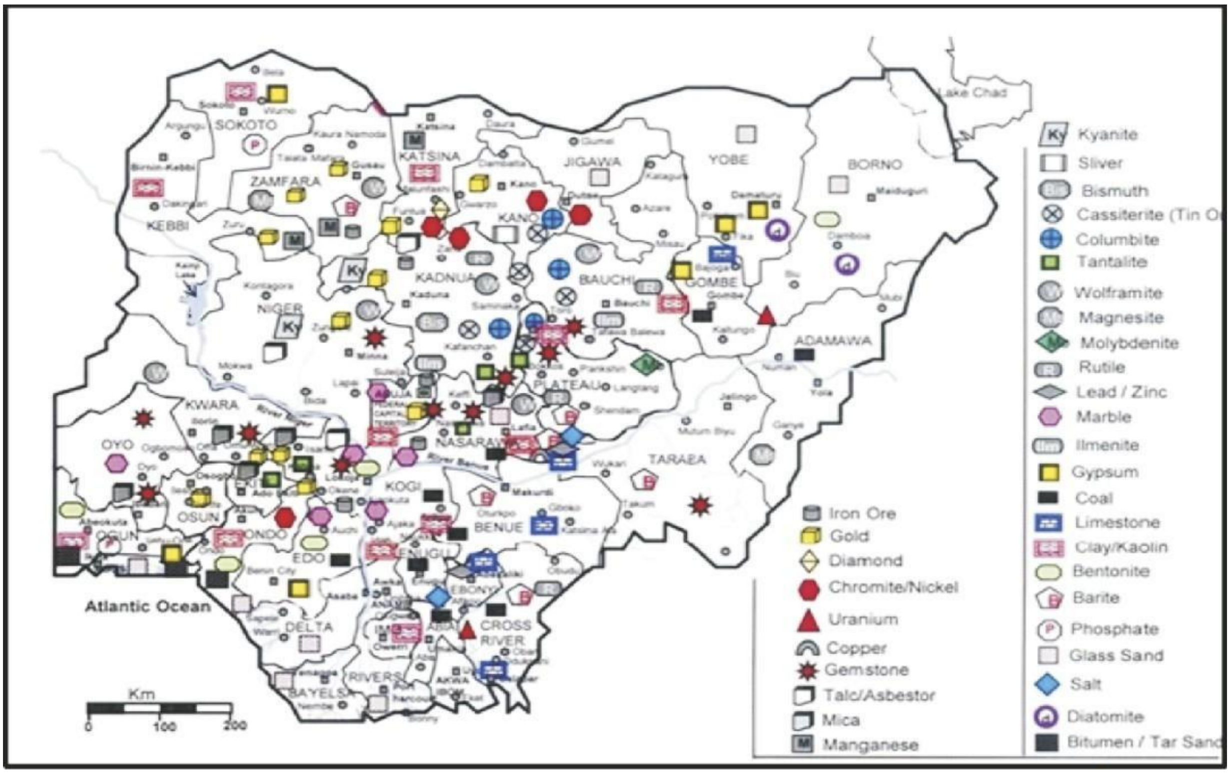

Figure 1: Map showing mineral occurrences in Nigeria [15]

The Gombe hill 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 hill 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 [11, 12, 16].

The Benue Trough is geographically subdivided into Lower, Middle and Upper (Fig. 2) Troughs [17, 18]. 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.

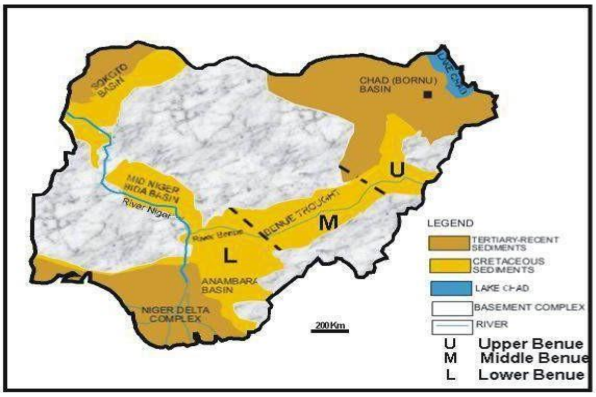

Figure 2: Geological Map of Nigeria showing the Benue Trough [18]

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 Upper Benue Trough is also generally divided into three (3) sub-basins [17, 19]; 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.

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.

Materials and Methods

The geochemical investigations will involve analysis of the ore samples employing sulphur, oxygen isotope analysis and fluid inclusion studies. The geochemical techniques employed for this work include the following;

a) Mass Spectrometry
b) Petrological Microscope, Microthermometry and Raman Microprobe.

Sulphur and oxygen isotopic analysis was conducted on nine (9) representative samples of barite at the Queen's Facility for Isotope Research, Department of Geological Sciences, Miller Hall, Queen's University Kingston, Ontario, Canada. All results were calibrated to certified reference materials, reported in standard permil notation (‰) and relative to the following international standards of Vienna Standard Mean Ocean Water (VSMOW) for $\delta^{18}O$, and Vienna Canyon Diablo Troilite (VCDT) for $\delta^{34}S$. Precision, in permil notation (‰) is based upon duplicate sample analyses. Accuracy (std.dev.), reported in permil notation (‰), is based upon primary or secondary standard analyses of 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: 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.

In sample preparation, ten (10) fluid inclusion sections (approximately 3 x 5 cm) were produced for these studies at the Queen's Facility for Isotope Research, Department of Geological Sciences, Miller Hall, Queen's University Kingston, Ontario Canada. Important factors to take note of in preparing the sections depending on transparency of the host crystal and the inclusion size and abundance, include the quality of polishing and the ideal thickness (typically, between 90 and 120 μm).

The petrographic study of the textural relationship between fluid inclusions and host rock begins with a careful identification of the different populations of fluid inclusions in such a way that the fluid inclusions analyzed is a true representative of the characteristic fluid of the process. The petrographic study will provide information about populations of fluid inclusions, such as number of inclusions, their types and chronology with respect to geological processes. This study conducted systematically is the key for proper decision – making about the type of fluid inclusions to be examined further [20–22].

The device for the Microthermometry study of the fluid inclusions is carried out using a heating and cooling stage. This allows for increasing or decreasing temperatures over a wide range (between -200 and +1500^{o}C approximately). This stage is placed on a microscope, so that the phase changes occurring as well as the temperature at which these changes taking place can be observed. From the start, the fluid inclusions are cooled to the lowest temperature that the stage can achieve and subsequently, the phase changes taking place are observed while the temperature rises towards room temperature again.

For the composition of the fluids: a confocal Raman microscope which uses an optical arrangement that inserts a limiting aperture at an image plane. This approach serves to limit the Raman signal entering the spectrograph to a very specific, sharply in focus, volume in the sample. The resulting Raman spectrum is characteristic of that isolated region alone, eliminating or strongly reducing Raman signals from out-of-focus regions in the field of view.

Information on the geofluids that influence the petrogenesis in this research work will include the temperature of entrapment of the fluid (also seen as the temperature of formation of a fluid inclusion), salinity of the fluid (expressed as weight - NaCl equivalent) and the composition of the fluid (eutectic composition).

Results and Discussion

Stable isotopes studies and fluid inclusion microthermometry analysis

Nine (9) barite samples were analyzed for their sulphur and oxygen isotope values (Table 1). The sulphur isotope ($\delta^{34}S$) values expressed in per mil (‰) relative to Canon DiabloTroilite of sulphate (barite) yield $\delta^{34}S$ 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 (Fig. 3) alongside the standard plots reported by Rollinson [23]. These values indicate that the source of the sulphur and by extension the mineralizing fluids were from the modern seawater.

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

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 (Fig. 4) alongside the standard plots reported by Rollinson [23]. 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.

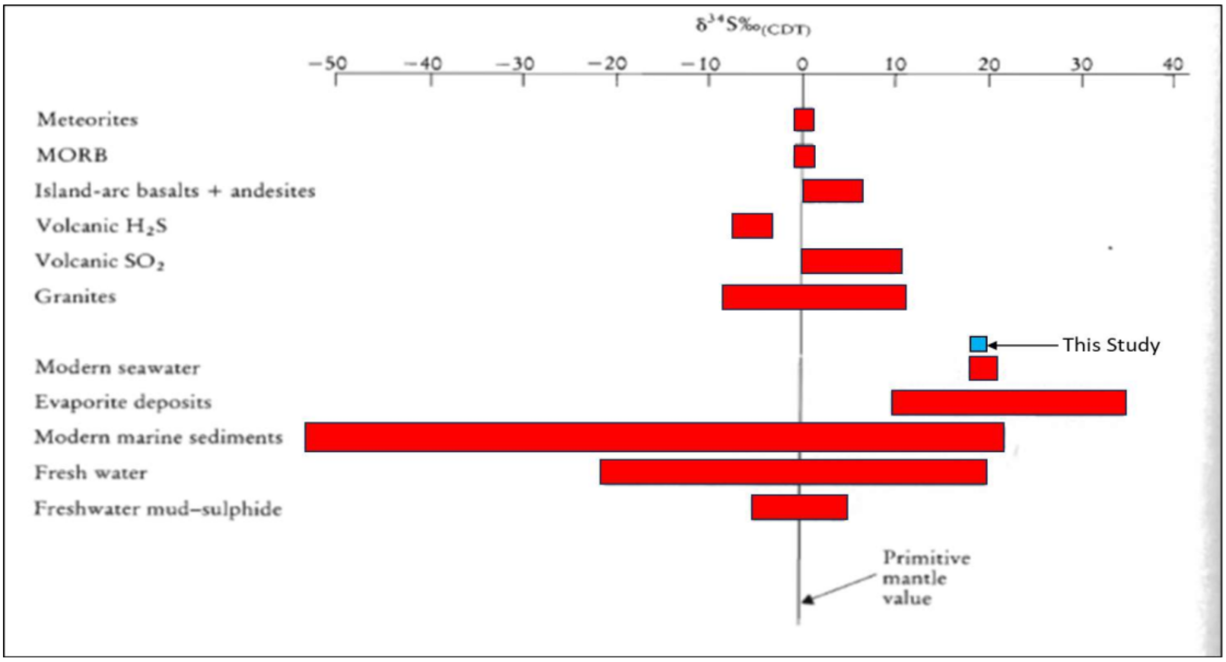

Figure 3: Natural sulphur isotope reservoirs, barite values of the barite mineralization sub-system are also plotted (modified after [23])

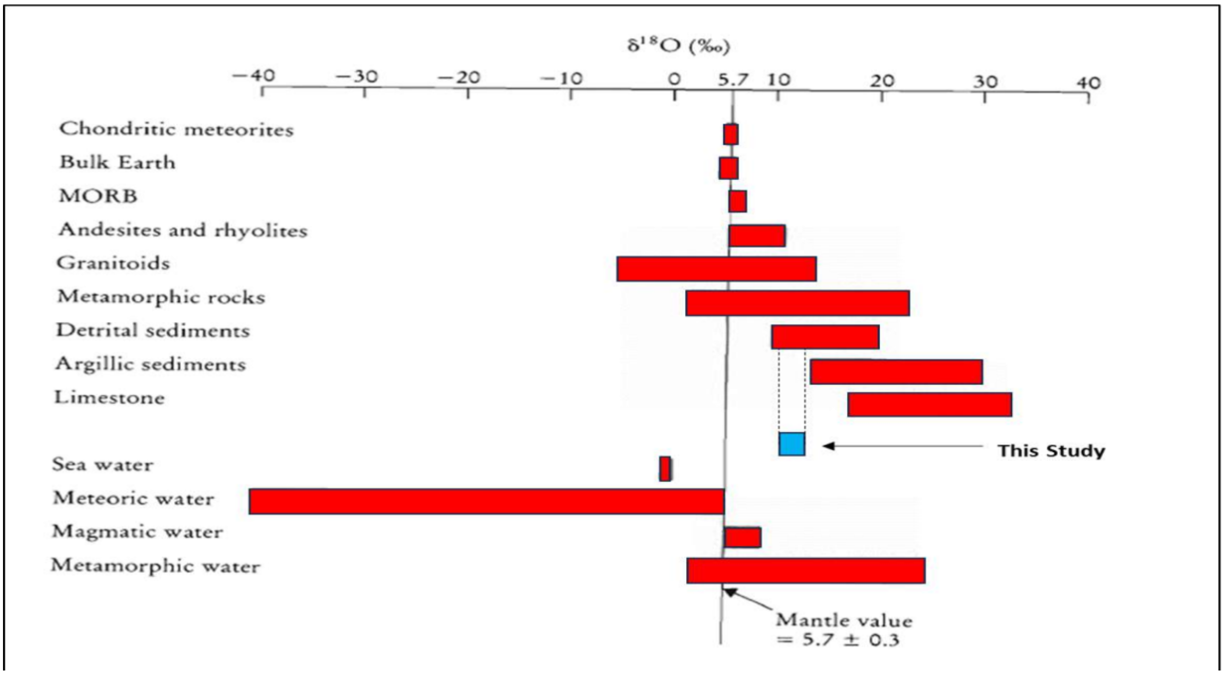

Figure 4: Natural oxygen isotope reservoirs, barite values of the barite mineralization sub-system are also plotted (modified after [23])

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_4may 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 distinguished the mode of barite formation, and in view of this, Fig. 5 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 (e.g. diagenetic barites, marine barites, cold seep barites and hydrothermal barites). Fig. 5a therefore, placed the barite of the Gombe in-lier as hydrothermal barite type. Figure 6 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 (Fig. 6a).

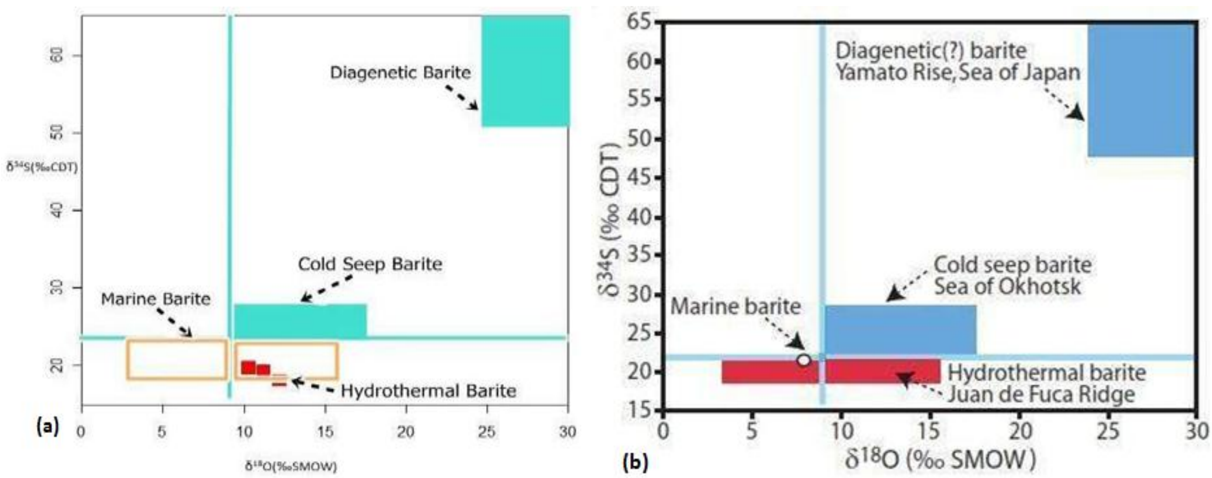

Figure 5: Plot of the sulphur and oxygen isotopic composition of; (a) Barite samples of the Gombe Hill, (b) JuandeFuca O and S isotope data [24, 25], respectively. Sea of Okhotsk data [26], Sea of Japan data [27]. Average Marine barite data [27].

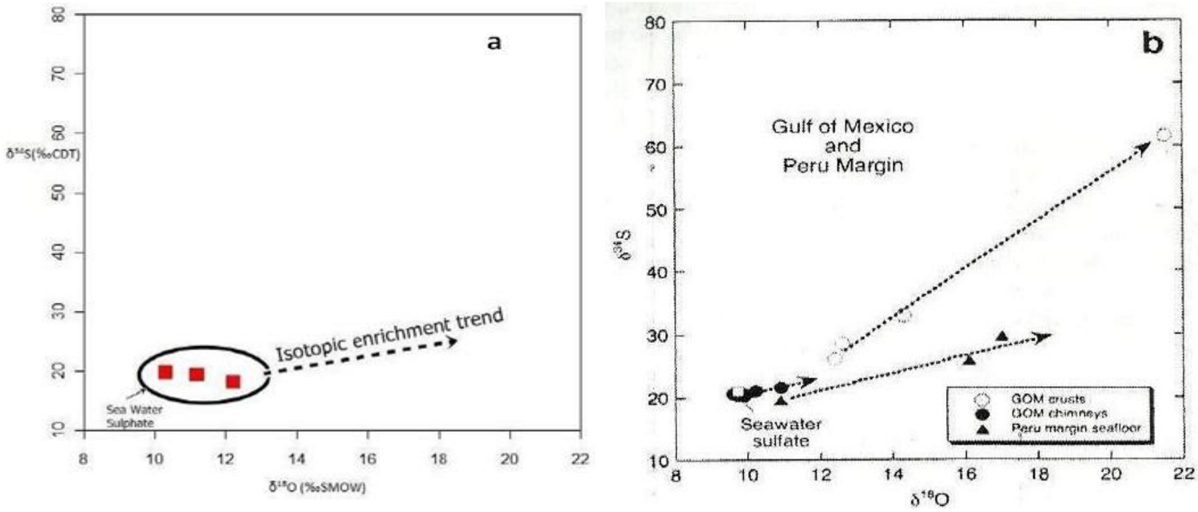

Figure 6: Sulphur and Oxygen-isotopic composition of barite from modern continental margins: (a) Barites of Gombe Hill deposited within veins, (b) Barites deposited beneath the sea floor as cements or crust show large isotopic enrichment relative to normal seawater sulphate

Table 2: Results of fluid inclusion studies of barite and fluorite mineralization of the study area (Gombe Hill)

S/N	Location	Material	Sample I.D.	Results of Fluid Inclusion Range Th (°C)	Average Th (°C)	Range Tmi (°C)	Average Tmi (°C)	Salinity (wt% NaCl Equiv.) (Range)
1	Gombe Hill UBT, Study Area	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
2	Gombe Hill UBT, Study Area	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

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

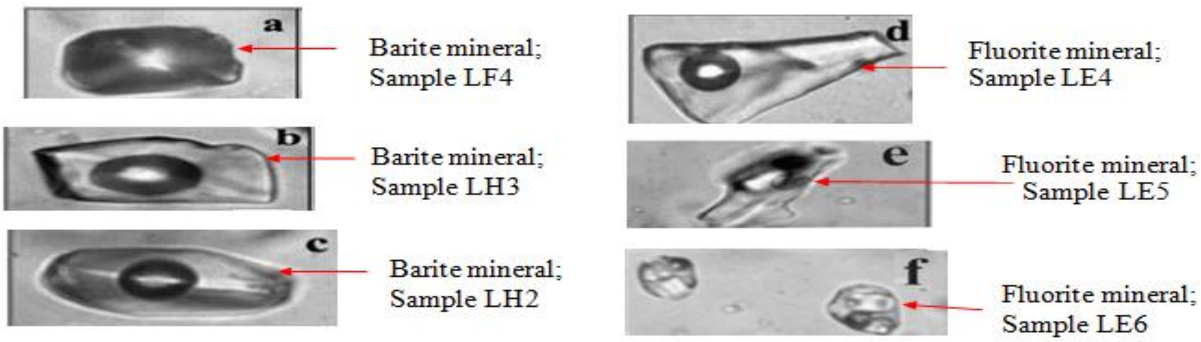

Figure 7: Primary Inclusion with rounded to rectangular shapes in Barite Sample (a–c) and fluorite samples (d– f)

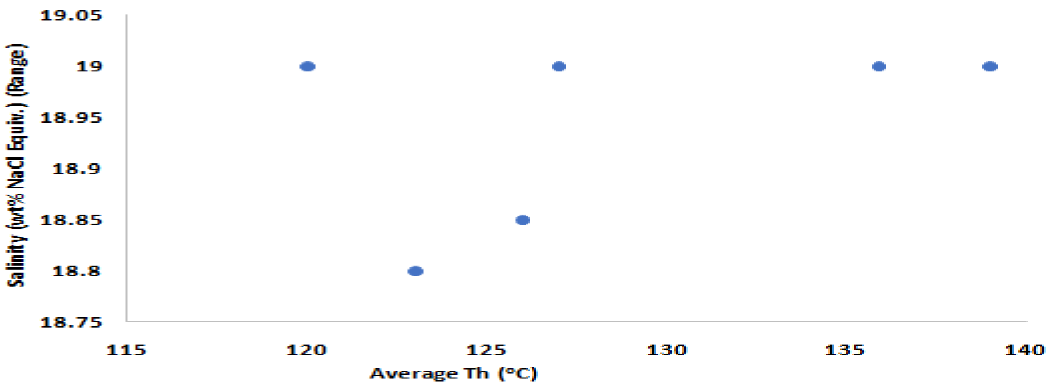

Figure 8: Homogenization temperature (Th) vs. salinity diagram for fluid inclusion data of barite and fluorite mineralization of the study area (Gombe Hill)

The results of fluid inclusion studies of three barite separates/mineral-grain showed 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 – 155^{o}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 (Fig. 7) 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 – 142^{o}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 (Fig. 8) 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.

Gombe hill is abasement 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 by many authors notably [28, 17, 29].

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 [7]. 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. [30] 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 YauriAu 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 and oxygencon form to the views of [31, 32] 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 5and 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 seawater. Researchers [33, 34] 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 – 155°C for barites and 125 – 142°C for fluorite (low temperature conditions).

Previous genetic hypotheses suggested for the Benue ore fluids include (1) a magmatic hydrothermal origin [35]; (2) formation from juvenile and connate brines [36] and (3) circulating connate brine source [14]. 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 for sediment-hosted Irish base-metal mineralization seems attractive to these 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. This study favors a basinal-brine expulsion model for the Upper Benue ore fluids. Sudden dewatering accompanying overpressures in sedimentary basins [37] 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 Sand stones) 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;

i. The barite 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.
ii. The hydrothermal fluid seems to have supplied the barium ions (Ba^{2+}), while the sea water supplied the sulphate ions (SO_4^{2-}).
iii. The high density of the hydrothermal fluid favours precipitation of barite minerals.
iv. 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 60-155°C for the mineralizations.
v. 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.
vi. Values arrived at, indicate that the source of the oxygen were from the sedimentary basin of the host rocks (detrital sediments).
vii. The fluid inclusion data show that the mineralizing fluids were saline brines containing recognizable Na, Ca and Cl and that fluid temperatures ranged between 60-155°C for barites and 125–142°C for fluorite (low temperature conditions).
viii. This study favors a basinal-brine expulsion model for the Upper Benue ore fluids. The sudden dewatering accompanying overpressures in sedimentary basins is the mechanism for the expulsion of metal-bearing fluids through permeable fractures in the Cretaceous sediments.
ix. 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 deposits.

Conflict of interest: The authors wish to declare that there is no conflict of interest in this research work.

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