Communication in Physical Sciences 2020, 5(3): 315-324

Petrography and Geochemical Studies of Eyingba Lead-zinc Mineralization, Lower Benue Trough

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Received 29 May 2020/Accepted 20 June 2020/Published online: 25 June 2020

Abstract *The geochemical studies of the Enyingba lead-zinc mineralization in the Lower Benue Trough was carried out to unravel the elemental association of the mineralization along with the gangue minerals and to further define the provenance of the mineralization. The petrographic studies of galena, sphalerite, chalcopyrite, siderite was carried out with Carborundum grit of 90, 120, 400, 600, 800, 1000, 1200 and magnesium oxide (MgO) to ensure smoothening of samples on a polishing machine. The geochemical result obtained indicated the enrichments of Pb (400000ppm) in galena, Zn (200000ppm) in sphalerite, Cu (100000ppm) for chalcopyrite. Chondrite normalization for galena/barite/siderite plots reveals positive Eu and Tm anomalies in galena, with a gentle slope depletion from the LREEs to HREEs. Chalcopyrite/sphalerite normalization plots also indicated a positive spike for Eu and Tm which imply that they are co-genetic. The concentrations of these elements in their ores reveal their abundance, high grade and could be exploited in large scale. Exsolution blebs from polished section also emphasize paragenetic relationship. The positive anomalies observed on the chondrite plots are due to hydrothermal fluid inputs in the mineralization since REEs are believed to be the least soluble and relatively immobile during weathering and hydrothermal alterations.*

Key Words: *Benue trough, petrography, geochemical studies, lead-zinc mineralization*

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1.0 Introduction

According to Fatoye *et al.* (2014), the Benue Trough of Nigeria is a sedimentary basin that extends from the Gulf of Guinea in the southwest to the Chad Basin in the northwest for about 800 km in length and 150 km in width. Olade and Morton (1985) had stated that epigenetic Pb-Zn deposits of the southern Benue Valley (Nigeria) are localized within Cretaceous sediments of an intracontinental rift basin. They carried out fluid inclusion studies of vein minerals from the Abakaliki and Ishiagu orebodies and observed that sphalerite and quartz were deposited at relatively low temperatures (102–175 °C), with ore-fluid salinity mostly in the range of 17–25 equiv. wt% NaCl. Consideration of geotectonic setting of the Benue trough with respect to the mode of occurrence and fluid-inclusion characteristics, mineralization in the trough is attributed to setting of the connate brines into motion by a high geothermal gradient accompanying continental rifting (ref). Other researchers also link lead–zinc mineralization in the lower Benue trough to hydrothermal and epigenetic (Farrington , 1952;Nwachukwu , 1972; Obarezi *et al* ., 2013; Olade, 1975,1976; Orajaka, 1965, 1972; Orazulike, 1994). The mineralization in the area was linked to fracturing and joining. Generally, lead-zinc mineralization is often associated with ores of galena and sphalerite respectively and in most cases, the mineralization occurs in form of dissemination, stockwork and narrow beds with poorly developed wall-rock silicification. The Enyingba, Ameri and Ameka lead-zinc lode–host other co- minerals such as siderite, chalcopyrite, malachite, azurite, pyrite, quartz, sericite, cerussite and anglesite. Hence this has over the years compelled the activities of both artisanal and small-scale mining of the lead-zinc mineralization within the lower Benue trough.

The increase in demand for these ores have necessitated the review of the historical and general as well as the mineralization and petrogenesis/metallogenesis of lead-zinc ore in the Enyingba district which forms the focus of this work.

2.0 Materials and Methods

2.1 Local geology

The Eyingba, Ameri, and Ameka is marked by undulating range of shale outcrops, which serve as the host for Pb-Zn mineral deposits (Nnabo, 2015). The area forms part of the "Abakaliki anticlinorium" and is generally underlain by the Abakaliki shales of the Asu River Group. The lower Cretaceous Abakaliki shale is exposed in the area. The sedimentary rocks are predominantly black calcareous (calcite-cemented) shale with occasional intercalation of siltstone (Nnabo, 2015).

The study area is the mining district of Enyigba (Fig. 1) which is located about 14 km southeast of Abakaliki town and lies between latitudes 6° 07' N and 6°12' N and longitudes 8° 05' E and 8° 10E' . The area is in a conspicuously low-lying to gently undulating terrain within the Cross River plain. The lithostratigraphic units underlying Enyigba, Ameri and Ameka and its environs are mainly the sediments of Abakaliki shale (Asu–Rver Group) (Nnabo, 2015). These units are delineated on the basis of their lithologic character, structures and stratigraphic positions. Two units were identified and were designated as unit A and unit B (Table 1). Unit A is made up of muddy, dark, grey/greenish and black shale while unit B comprises of grey-brown and reddish shale. The shales are mostly weathered and this accounts for their reddish colour.

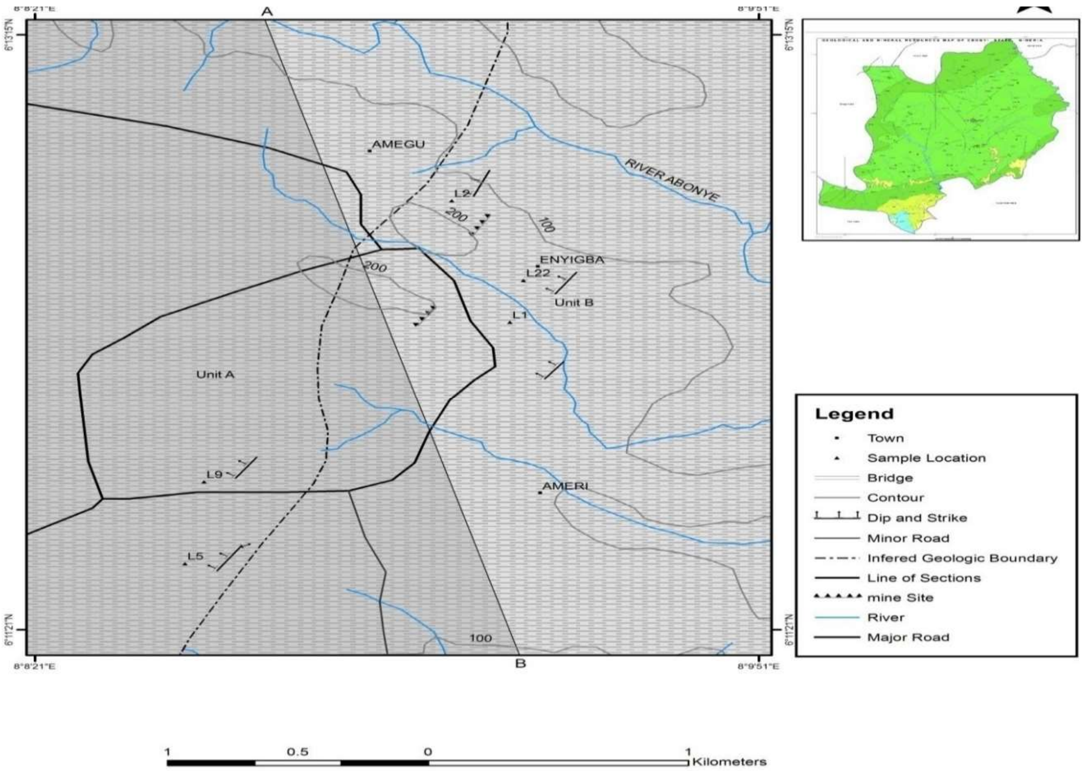

Fig 1: Geologic map of Eyingba district

2.2 Sampling and methods

For the petrographic studies, eleven representative samples were selected for polished section preparation. The samples were cut into thicknesses of 3cm and then trimmed. Each sample of galena, sphalerite, siderite and chalcopyrite were successively polished with carborundum grit (in the presence of water) of 90, 120, 400, 600, 800, 1000, and 1200 grades respectively. Further smoothening was done by re-polishing the samples with magnesium oxide to polish the samples on a polishing machine. Finally the samples were polished with glycerol on a text-net cloth to enhance a fine surface, then mounted on a Nikon LV100POL reflected light microscope for studies.

Table 1: Stratigraphic and Correlation Table for Eyingba district.

Group	Formation	Member	Age	Description
Azu River group	Abakiliki shale	B	Albian	Grey to dark brown reddish shale with carbonaceous stains; weathered surface is dark in colour
Azu River group	Abakiliki shale	A	Albian	Dark grey/greenish and black shale muddy fissile, glauconite and micaceous, easily breaks into fragments
Crystalline basement	Precambian	A	Unexpected igneous and metamorphic rocks	

Geochemical analysis was carried out using a laser ablation inductive couple plasma which used metaborate/tetraborate fusion. Samples of galena sphalerite, siderite, chalcopyrite and baryte where digested using nitric and hydrochloric acid in HNO_3: HCl ratio of 1:3. This was carried out by ACME Analytical Laboratory Limited, Vancouver, Canada. The major oxides were digested by lithium metaborate/ tetraborate fusion, while the Trace and Rare earth elements were digested using HNO_3-HCl acid and lithium metaborate/ tetraborate fusion.

Prepared samples were mixed with $LiBO_2/Li_2B_4O_7$ flux. The cooled bead was dissolved in ACS grade nitric acid and analyzed by ICP-MS. Loss on ignition (LOI) was determined by igniting a sample split then measuring the weight loss. Total carbon and sulphur were determined using the method recommended by Leco (TC003).

3.0 Result and Discussions

3.1 Petrographic studies

The photomicrograph of chalcopyrite, galena and sphalerite (obtained from the studied area) are shown in the photomicrographs presented in Fig. 2

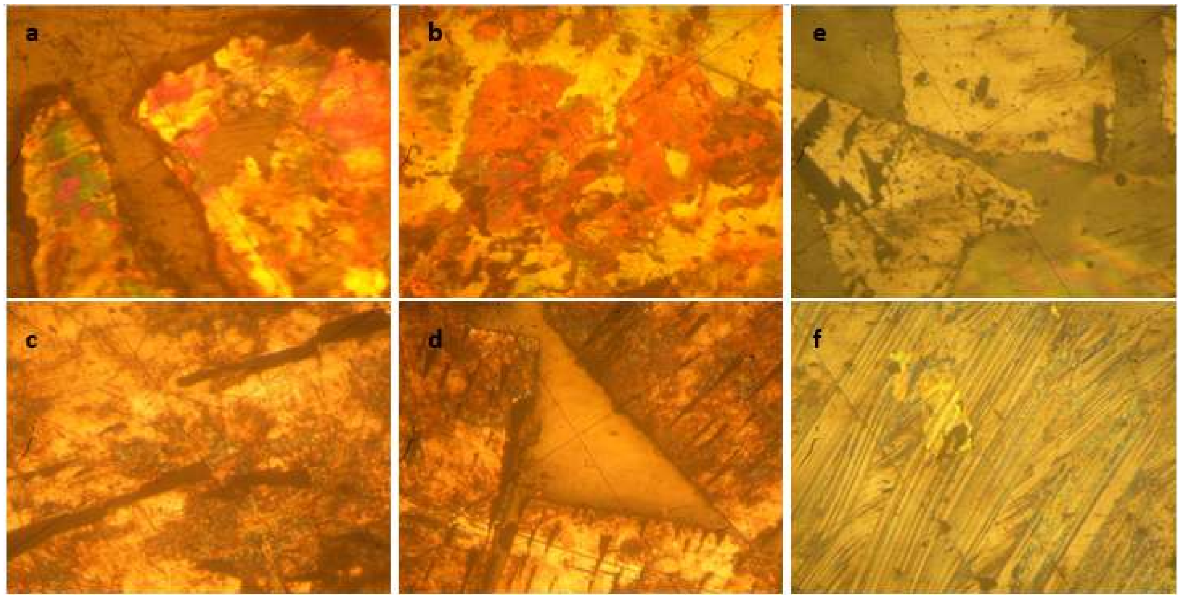

Fig 2: Photomicrographs of (a & b)-Chalcopyrite, (c & d)-Galena and (e & f)-Sphalerite

The photomicrographs of chalcopyrite (Fig. 2a & b) reveal forms of irregular yellow masses, with dotted dark patches that suggest bleb of galena at the time of cooling. There’re also sections with greenish to reddish coloration that shows a wide definition of chalcopyrite.

The photomicrograph of galena (Fig. 2c & d) show traces of exsolution blebs of sphalerite. The section also appears yellow in colour with discordant mineral (sphalerite) alignments showing perfect cleavages. Some sections show larger dark patches indicating fractures with terminated mineral edges like sphalerite.

Sphalerite (ZnS) in (Fig 2e & f) suggest existent in its pure state but highly discordant. It displays grey to brown colour as well as traces of yellow to green imprints on the sections which are indications of other accessory minerals like pyrite showing golden yellow. Some cleavages are also observed.

3.2 Geochemical studies

Concentrations of major oxides obtained from geochemical analysis of the samples are presented in Table 2. Several plots from the values obtained have helped in establishing certain facts. Plots of Na_2O/Al_2O_3 versus K_2O/Al_2O_3 defined the environments of deposition as sedimentary-metasediment in origin (Fig. 3).

There's abundance of silicon oxide (SiO_2) when compared to calcium oxide (CaO) (Fig 4) and potassium oxide, K_2O (Fig 5). For the major oxides, SiO_2 record the highest value in all the samples analysed as (74.45 ppm), followed by Al_2O_3 (34.56 ppm).

Table 2: Geochemical composition of galena, sphalerite, chalcopyrite and siderite

Oxide (%)	Galena			Sphalerite			Chalcopyrite	Siderite
	Minimum	Maximum	Mean	Minimum	Maximum	Mean		
SiO_2	3.82	21.15	**13.95**	9.61	21.15	**15.56**	12.230	19.110
TiO_2	0.02	0.10	**0.06**	0.11	0.70	**0.37**	0.650	0.400
Al_2O_3	0.40	2.23	**1.34**	2.26	5.60	**3.87**	34.560	3.460
Fe_2O_3	3.44	6.87	**4.39**	0.08	3.74	**1.39**	0.640	0.060
MgO	0.09	0.32	**0.16**	0.07	0.17	**0.12**	0.210	0.030
CaO	0.04	0.12	**0.08**	0.03	0.08	**0.05**	0.090	0.030
Na_2O	0.03	0.08	**0.06**	0.08	0.14	**0.1**	0.090	0.070
K_2O	0.04	0.45	**0.17**	0.01	0.44	**0.16**	0.030	0.010
P_2O_5	0.03	0.05	**0.04**	0.03	0.03	**0.03**	0.010	0.020
MnO	0.04	0.37	**0.13**	0.06	0.25	**0.13**	0.930	0.070
Cr_2O_3	0.00	0.04	**0.01**	0.00	0.00	**0.00**	0.002	0.003
LOI	10.8	14.1	12.95	11.1	14.2	12.77	17.300	13.000

Silicon (IV) oxides was found to be most abundant with mean percentage concentration of 13.95, 15.56 , 12.230 and 19.110 in galena, sphalerite, chalcopyrite and siderite respectively. Concentration of silicon (IV) oxide in galena ranged from 3.82 to 21.15 % but from 9.61 to 21.15 in sphalerite. Mean concentration in chalcopyrite was 12.23 % and 10.11 % in siderite. Mean concentration of Cr_2O_3 in galena was 0.01%. However, mean concentrations of 0.002 and 00.003 % were found in chalcopyrite and siderite respectively. Mean concentration of TiO_2 was least in galena (0.06 %) and highest in chalcopyrite (0.650 %). Al_2O_3 had highest percentage concentration in chalcopyrite (34.56 %) and least in galena (1.34%) while mean concentrations of Fe_2O_3 in galena, sphalerite, chalcopyrite and siderite were 4.39, 1.39, 0.640 and 0.060 % respectively. The corresponding values obtained for MgO were 0.16, 0.12, 0.210 and 0.030 respectively while that of Na_2O were 0.06, 0.1, 0.09 and 0.07 % respectively. Mean K_2O concentrations in galena, sphalerite, chalcopyrite and siderite were 0.17, 0.16, 0.03 and 0.01 % respectively while that of P_2O_5 were 0.04, 0.03, 0.01 and 0.02 % respectively. Manganese exhibited moderate mean concentrations of 0.13, 0.13, 0.930 and 0.070 in galena, sphalerite, chalcopyrite and siderite

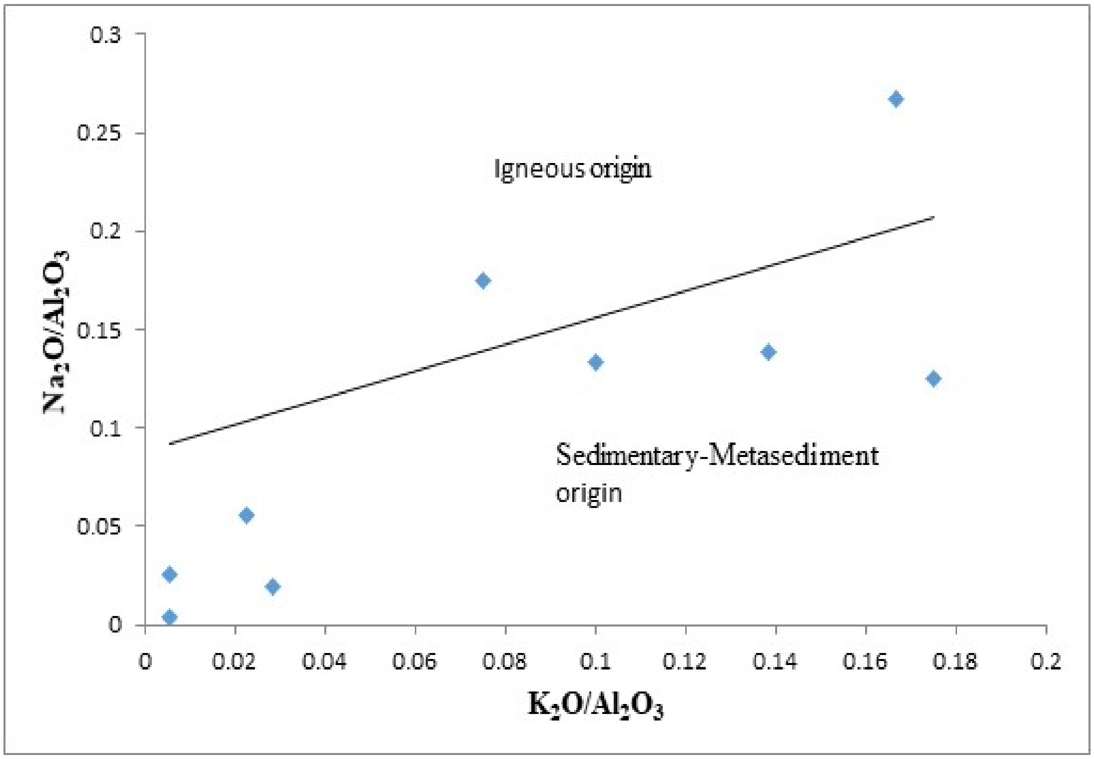

Fig. 3: Plot of environment of deposition after Garels and Mckenzie 1971

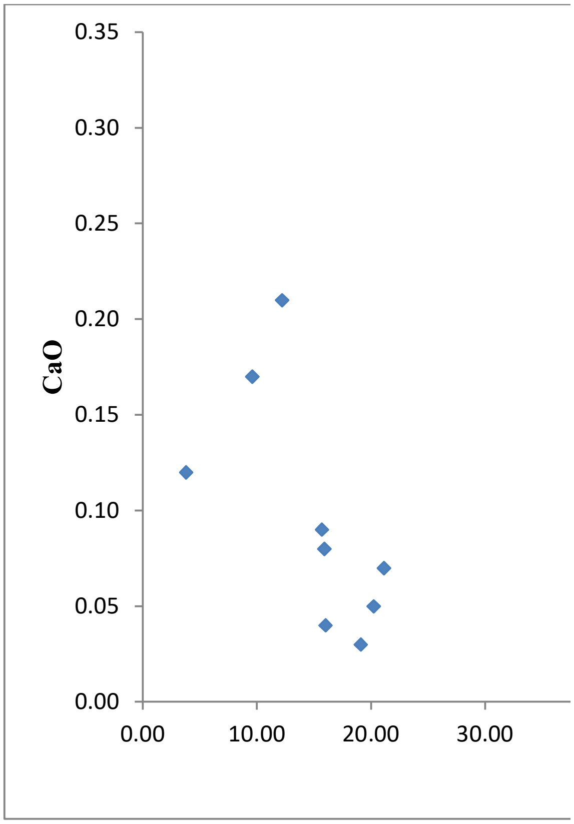

Fig. 4: CaO/SiO_2 concentration modified after Brown et al 1979

Trace elements

Results showing concentrations of Trace elements (Table3) indicates the trace elements enrichments of Pb (400000ppm), galena, Zn (200000ppm) and sphalerite, Cu (100000ppm) for chalcopyrite. Measured high concentrations of Pb, Zn, and Cu they constitute the base elements of their ores. These high values depict their abundance in nature hence could be explored in large scale. Ni in chalcopyrite is relatively high at 609.9(ppm). Mn shows appreciable abundance in the ores especially in the chalcopyrite sample at 7229 (ppm). Cd also records very high values ranging from (0.1ppm) in baryte to (1324 ppm) in F

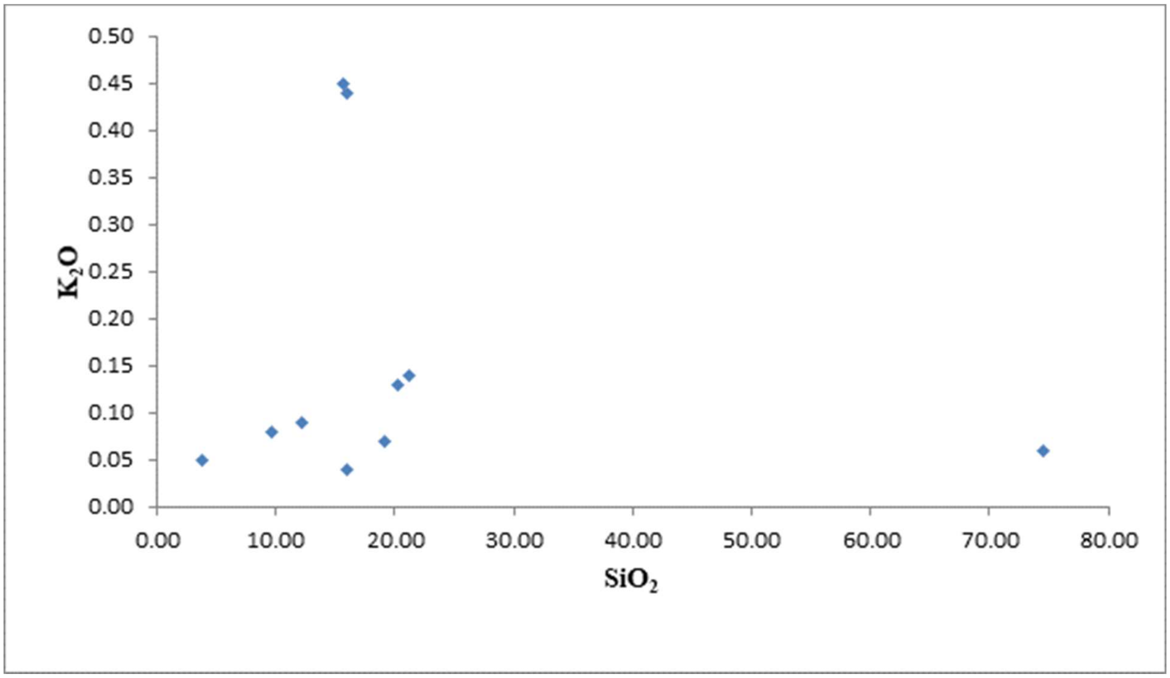

Fig 5: Fig. 5: K_2O/SiO_2 concentration in the oxides after Middlemost, 1975.

Table 3: Trace elements in galena, sphalerite, siderite and chalcopyrite

Trace Element	Galena			Sphalerite			Siderite	Chalcopyrite
	Min	Max	Mean	Min	Max	Mean		
Ba	24	59	43	23	69	45	18	33
Ni	0	78	30	1	73	33	1	636
Co	4	60	42	30	50	43	44.8	39
Ga	1	19	13	9	15	12	12.6	8.2
Nb	0	2	1	0	2	1	0.5	0.7
Rb	1	23	12	3	20	10	3.5	2.6
Sr	10	23	17	10	29	18	8	19.8
V	0	26	11	10	39	26	0.1	0.1
Zr	7	16	11	7	20	12	5.1	8.5
Cu	1505	10467	3883	1915	6893	3596	1924.3	100000
Pb	40000	40000	40000	40000	40000	40000	40000	38353
Zn	8858	200000	152215	175856	200000	191952	200000	128569
Ag	34	222	86	33	144	77	54.4	252.5
Ni	11	74	28	14	50	26	14.3	609.9

Co	3	90	62	29	88	61	68	35.2
Mn	305	2787	997	307	1947	948	592	7229
Cd	30	1299	917	478	1324	957	1078.9	357.7
Sb	57	385	150	51	269	140	103.2	32.6
Bi	3	26	9	3	18	9	5.4	3.5
Cr	1	2	1	1	1	1	1.8	1.4
Hg	0	20	13	5	18	12	13.09	5.23
Ga	0	30	20	10	29	20	22	10
Se	12	57	24	11	39	22	15	30

3.3 Rare earth element

According to Schneider *et* al. (1998), the rare earth element patterns of samples are known to preserve their original geochemical characteristics. They are also believed to be the least soluble and relatively immobile elements during weathering and hydrothermal alterations (Ibe and Obiora, 2019) and are thus useful factor in describing provenance. Measured concentrations of rare earth elements in galena, siderite, chalcopyrite, sphalerite and baryte samples are presented in Table 4. Four samples of galena, baryte and siderite were normalized with respect to chondrite (Fig 6). Barite shows a relatively even slope from the LREEs to the HREEs. Siderite also show a gentle slope without any form of anomaly.

Galena shows some anomalous variations in certain elements especially Eu and Tm having high positive enrichment.

Table4: Geochemical data for the rare earth elements in Eyingba

	Galena			**Sphalerite**			**Baryte Mean**	**Chalco Mean**	**Siderite aver**
Rees	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**			
La	1.8	6.7	4.35	2	4	2.966667	26.9	2.2	13.3
Ce	0.8	11.7	6.45	2.2	5.2	3.4	53.7	2.4	21
Pr	0.1	1.34	0.7125	0.22	0.52	0.333333	6.66	0.3	2.38
Nd	0.4	4.5	2.45	0.6	2.1	1.333333	25.5	1	8.8
Sm	0.07	0.82	0.405	0.1	0.32	0.176667	3.82	0.31	1.43
Eu	0.03	0.25	0.15	0.04	0.11	0.07	1.28	0.17	0.35
Gd	0.17	0.65	0.3925	0.16	0.31	0.21	2.76	0.47	0.96
Tb	0.03	0.08	0.0525	0.02	0.03	0.026667	0.33	0.12	0.12
Dy	0.16	0.34	0.255	0.15	0.25	0.19	1.69	0.89	0.46
Ho	0.03	0.07	0.055	0.03	0.05	0.04	0.36	0.22	0.08
Er	0.06	0.27	0.18	0.04	0.11	0.083333	1.27	0.63	0.22
Tm	0.02	0.1	0.0425	0.1	0.1	0.1	0.2	0.07	0.03
Yb	0.06	0.22	0.15	0.07	0.11	0.09	1.39	0.41	0.27
Lu	0.01	0.1	0.04	0.01	0.1	0.07	0.23	0.05	0.04
Y	0.7	2	1.375	0.6	1.1	0.9	11.3	4.4	2.7

The spider diagram (Fig. 7) is a normalization plot of the REEs with respect to the three sphalerite samples and a chalcopyrite sample. A positive spike of Eu in chalcopyrite and sphalerite was observed. The sphalerite enrichment in HREEs occurred simultaneously in the three samples suggesting a common provenance. Tm records 0.1 ppm for the three sphalerite samples with the high positive spikes which marks a broad difference from others with lower values of 0.03, 0.02 and 0.07 ppm respectively. Chalcopyrite maintains alternating values ranging from 0.07 - 4.4 ppm, hence the observed anomalous enrichment. These are all indications of hydrothermal infiltration at the time of mineralization. Gulcan and Ahmet (2004) reported that such anomalies are indications that the minerals formed from hydrothermal fluids with different compositions at the time of precipitation. Comparative study of the high anomaly in the galena trend and the chalcopyrite normalization in Figs. 6 & 7 suggest two phases of volcano-sedimentary activities. The first episode being direct precipitation of the minerals and the second, a re-dissolution of the initially precipitated minerals, thus, a differing provenance with respect to their episodes

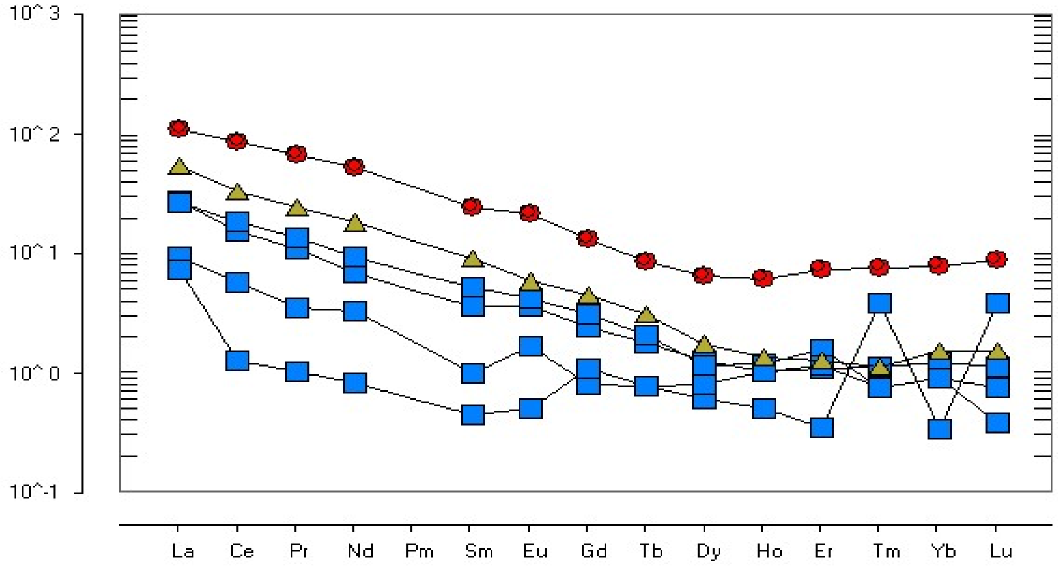

Fig.6: Chrondrite normalized rare earth element pattern for samples of four Galena samples, Baryte and Siderite modified after Boyton 1984. Galena (Blue), Barite (Red), Siderite (Brown).

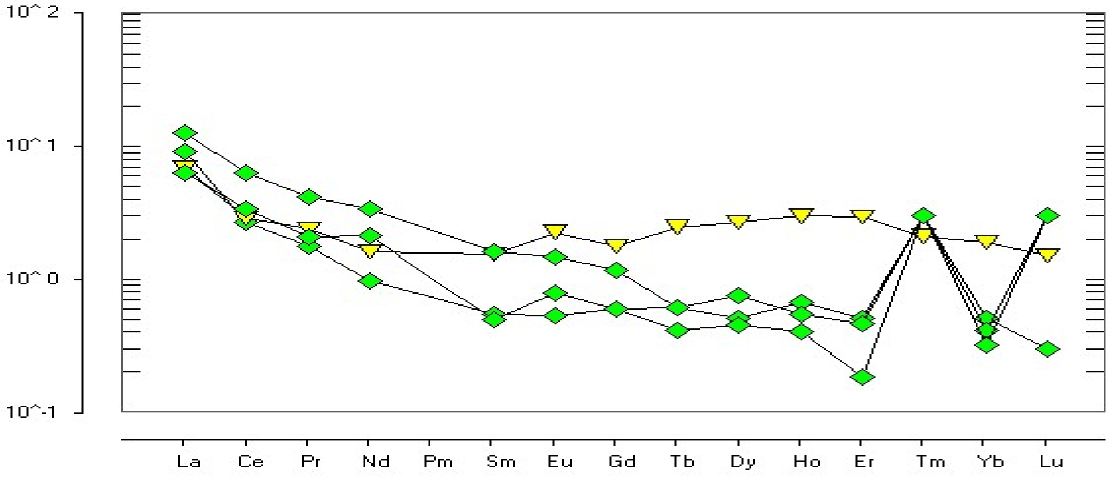

Fig. 7 : Chrondrite normalized rare earth element pattern for three samples of sphalerite samples of a chalcopyrite sample modified after Boyton 1984. Chalcopyrite-Green, Sphalerite-Yellow

3.4 Minerals paragenesis

Ores are generally products of physiochemical processes that involve interaction, distribution, dispersion and deposition of one or more valuable minerals from which metals could be extracted. Since ores are essentially rocks, the origin of the ore deposit is related to various geochemical process and reaction that lead to their formation, transformation and modification of rocks.

Akande *et al*. (1990) reported a sequential evolution of the vein mineralization, suggesting three stages which include the pre-ore fracturing and brecciation of Albian shales and the precipitation of framboidal as well as colloform aggregate of pyrite, siderite and quartz. This stage is vital as it is preparatory for the later mineralization process. The next stage was the ore formation of sphalerite, galena, and copper bearing minerals such as the chalcopyrites, chalcosite, energite and digenite. This stage ushered in the third stage which was the deposition of a second generation of octahedral galena, sphalerite and marcasite.

Further probing of the mineral association in the study area reveals that pyrite mineralization was dominant before the introduction of the lead-zinc rich solutions, hence the mineralization process was not simultaneous but sequential notably in the order of pyrite-galena-sphalerite-rich top and chalcopyrite rich base leading to a decrease of the lead and zinc values and increasing copper at deeper levels as recorded in analysed samples with values greater than 40000ppm in Pb, 200000 in Zn and 1920.6ppm respectively in a galena sample (Tables 3 & 4).

The subject of the origin of the ore forming fluid has over time remained controversial, however fluid inclusion studies of the Abakaliki-Ishiagu vein by Olade and Morton (1985), Akande and Abimbola (1987) indicated that the solution responsible for the lead-zinc-copper mineralization were hot saline brines of about (150-230 °C). The mineralogical composition and association are compatible with those of ore deposits that were formed from basinal brines (eg. Mississippi Valley Type Deposit), and is believed to be epigenetic.

4.0 Conclusion

Considering the lateral extension of the trough, the hydrothermal solution in a bid to be deposited through the rock will lose heat and metal content as distance increases, this could be at low temperature (epithermal deposits) or intermediate temperature (mesothermal deposits), hence, the abundance of metal content in the hydrothermal solution, presence of solution capable of precipitating mineral matters, availability of openings (veins) in the rocks permitting movement of hydrothermal solution from the source to the site of deposition, chemical reactions causing the deposition of the ores were all factors that aided the epigenetic formation of the Eyingba Pb-Zn mineralization of the Lower Benue Trough. Polished samples show visible exsolution blebs depicting a paragenetic source. Some polished galena show traces of sphalerite sections, sphalerite with golden yellow color surmised as pyrite. Chalcopyrite sections show blebs of galena, which could be linked closely to their common source of formation.

There is a general gentle slope from the LREEs to the HREEs. The positive anomalies on the chondrite plots on Eu, Gd, Tm for the samples are due to hydrothermal fluid inputs in the mineralization since REEs are believed to be the least soluble and relatively immobile during weathering and hydrothermal alterations. The high concentrations of (Pb, Zn, Cu, Mn, Ni) in their ores reveal their abundance and high grade, hence mechanized and large-scale mining is recommended.

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