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GEOCHEMICAL CHARACTERISTICS OF IMMATURE COAL FROM IHIOMA, SOUTHEASTERN NIGERIA AND THEIR IMPLICATION FOR THE PALEOENVIRONMENT

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ABSTRACT

In this study investigation of lignite samples from coal mire of Ihioma mine in Orlu, Nigeria have been subjected to geochemical analysis to determine their mineralogical characteristics and paleodepositional environment. Petrographic data of composite samples indicate that these lignites were dominated by carbonate minerals followed by sulfides and clay minerals. Framboidal pyrites have also been recorded in the Ihioma lignite samples which reveals marine incursion into the study area. The occurrence of clay mineral in cell cavities, fissures and cracks indicates authigenic and syn-depositional origin of the Ihioma lignite. The study reveals Ihioma lignites deposited under terrestrial oxic to marine, terrestrial oxic to sub-oxic condition.

Keywords: *Geochemistry, Petrographic, Environment, XRD, XRF, Lignite, Ihioma.*

INTRODUCTION

Coal is one of the major sources of energy throughout the world. It comprises a complex heterogeneous mixture of organic (maceral) and inorganic (mineral matter) constituents (Saikia *et al.*, 2007; Vassilev and Tascón, 2003). Organic constituents are crucial to define the nature of coal, and it gives information about the suitability of coal for the different utilization processes (Singh and Singh, 2012; Singh *et al.*, 2015a; Taylor *et al.*, 1998; Ward, 1984, 2013). While the inorganic residue of coal results from combustion of coal at high temperature and it contains various minerals such as silicates, clay minerals, sulphides, phosphate, carbonates, sulphates, native elements (Vassilev and Vassileva, 1996; Ward, 2016), major, minor and trace elements (Singh *et al.*, 2015a). These inorganic constitutes may be used to decipher the paleodepositional condition of the coal-bearing basin. The significant concentration of mineral matter present in coal also imposes various environmental issues associated with coal utilization (Verma and Tripathi, 2000; Ward, 2002; Rajak *et al.*, 2018). It is mainly due to the presence of sulphur compounds such as pyrite and various sulphates of iron that reduces the combustibility of coal at high temperature. In addition, minerals like quartz increase health impact during coal mining operation (Erol *et al.*, 2013; Schatzel and Stewart, 2012), and it causes lung cancer through indoor coal combustion (Tian *et al.*, 2008). It also decreases the capacity for gas adsorption (Gurba *et al.*, 2003). For the removal of minerals from the coal, different approaches have been investigated (Juszczak *et al.*, 1995; Singh *et al.*, 2014).

Mineral matters also include trace elements and major elements such as aluminum, quartz, uranium, germanium recorded as a possible source with potential for economic recovery from the coal for modern industries (Ward and Dai, 2012). Several publications have discussed different aspects of mineral matter in coal (Vassilev and Tascón, 2003; Vassilev and Vassileva, 1996, 2009; Vorres, 1986; Ward, 2002). Moreover, minerals and trace elements in coals can provide vital information about depositional and post-depositional processes associated with the coal formation (Singh *et al.*, 2016; Ward and Dai, 2012). Considering the economic, environmental and utilization issues, the nature, origin and concentration of the mineral matters in the area have been studied.

Regional Stratigraphic Setting

Sub-bituminous coals are restricted to the Maastrichtian Mamu and Nsukka Formations which crops out in long narrow ridges in the NE–SW trending Anambra Basin (Fig.1) while lignites are restricted to Miocene – Oligocene Ogwashi – Asaba Formation crop out at Ihioma (Fig. 2). The Mamu and Nsukka Formations consist of alternating sandstones, sandy shales and mudstones with interbedded coal seams. The formation is underlain by the Campanian Enugu/Nkporo Shales (lateral equivalents), and overlain by the Ajali Sandstone (Middle Maastrichtian) and Nsukka Formation (Upper Maastrichtian to Danian. The Ogwashi – Asaba Formation consist of lignite, peat and intercalation of sandstone, clay and shale) (Fig. 2). Five sedimentary units are recognized in the Mamu Formation in the Enugu area, where the thickest exposed section (approximately 80 m) occurs (Simpsons, 1954; Reyment, 1965). From the base, the units consist of (i) a basal shale or sandy shale, (ii) sandstones with occasional shale beds, (iii) carbonaceous shales, (iv) coals and (v) sandy shales.

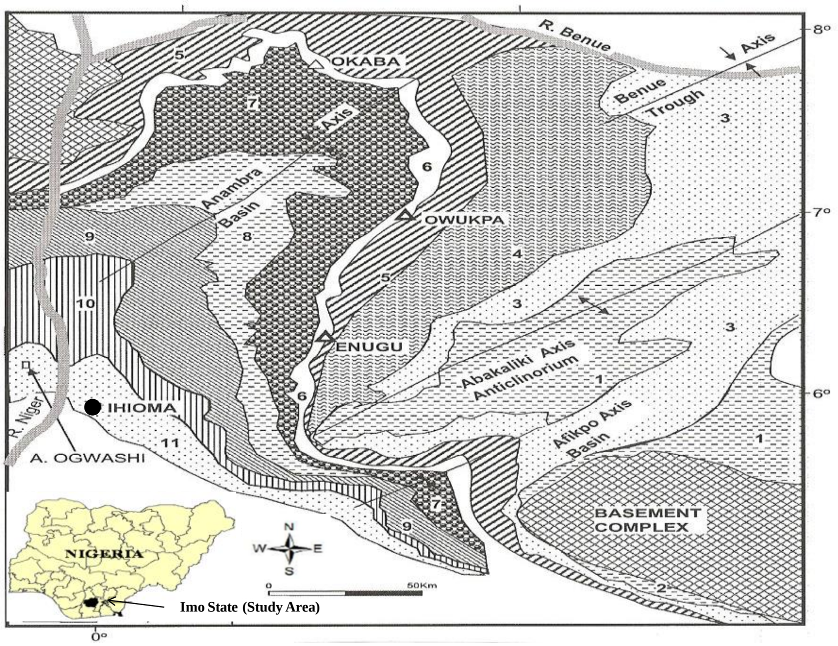

Fig. 1: Generalised geological map of the SE Nigeria (boxed areas of inset) showing the location of the coal deposits. Numbers indicate Cretaceous and tertiary formations shown as follows: **1.** Asu River Group; **2.** Odikpani Formation; **3.** Eze-Aku Shale; **4.** Awgu Shale; **5.** Enugu/Nkporo Shale; **6.** Mamu Formation; **7.** Ajali Sandstone; **8.** Nsukka Formation; **9.** Imo Shale; **10.** Ameki Formation and **11.** Ogwashi-Asaba Formation (modified from Akande et al., 2007)**.**

The Nsukka formation has well exposed section at Iyinzu in Ezimo area with thickness of about 55 m. The succession is paralic as demonstrated by the alternating marine and continental facies, and is considered to represent part of the "third marine cycle" in southern Nigeria (Short and Stauble, 1967; Hogue, 1977; Agagu, 1978). Sediments of the third marine cycle were mainly deposited in Campanian – Maastrichtian times in the Anambra and Afikpo Basins (Fig.1). These two post-Santonian basins were formed as successors to the Benue Trough, where deposition of the first and second marine depositional cycles took place in Albian – Santonian times (Figs 1 and 2). The third-cycle sediments were deposited during the initial growth of the proto-Niger Delta in the Late Cretaceous (Short and Stauble, 1967). An Early Paleocene transgression led to the termination of the proto-Niger Delta and the deposition of Imo Shale, which was succeeded by the regressive phases of the Ameki and Ogwashi Asaba formations.

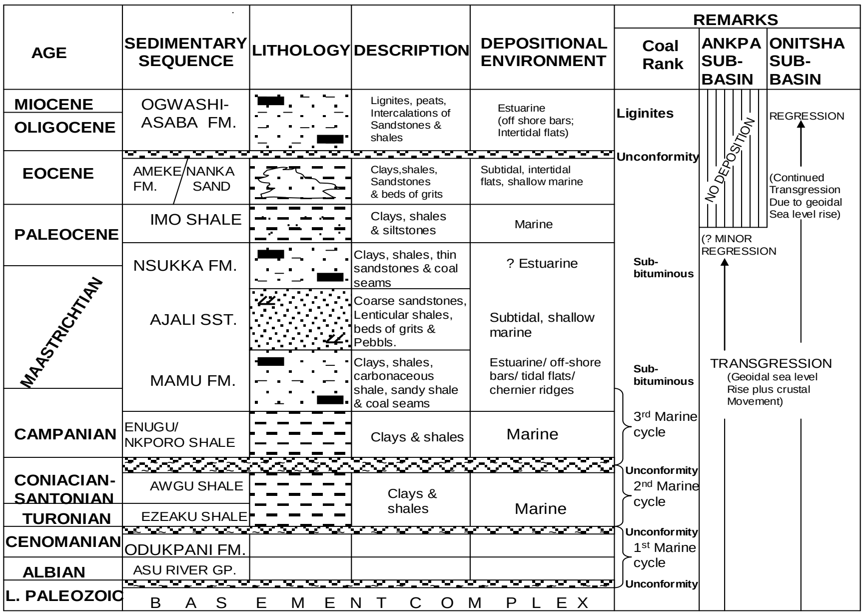

Fig. 2: The stratigraphy and environment of deposition of sediments in the Anambra Basin southeastern Nigeria.

MATERIALS AND METHODS

The lignite samples were collected using chisel and hammer and grab sampling method from the seam. The lignites having similar megascopic characteristics have been clubbed together to form a composite band for the analysis. In the laboratory, the samples were reshaped using a rotating steel cutter to eliminate surface that could be affected by alteration. Chips were cut from the samples and dried in an oven at 105 ^{o}C for 24 hours. The samples were crushed and pulverized in a rotating disc mill to yield about 50 g of samples for XRD and XRF analytical geochemistry.

The XRD analysis was carried out on powder lignite samples. The measurement has been taken within the angle range of 4 – 75^{o} 2θ. X'pert High score software digital was used for identification of minerals present in the Ihioma lignites. WD-XRF machine is used for detecting the major oxides and trace elements present in the sample.

RESULTS

Mineral matter assemblages visible under the microscope are dominated by carbonates (siderite), sulphides (pyrite, framboidal pyrite, disseminated as well as cracks and fissures fillings) and clay in fissures (Figure 3a-d) (Table 1). Carbonates are authigenic mineral in coal, and usually, Fe sulphides occur as siderite in massive and fine-grained lenses which are syngenetic in origin (Vassilev and Vassileva, 1996) (Figure 3a).

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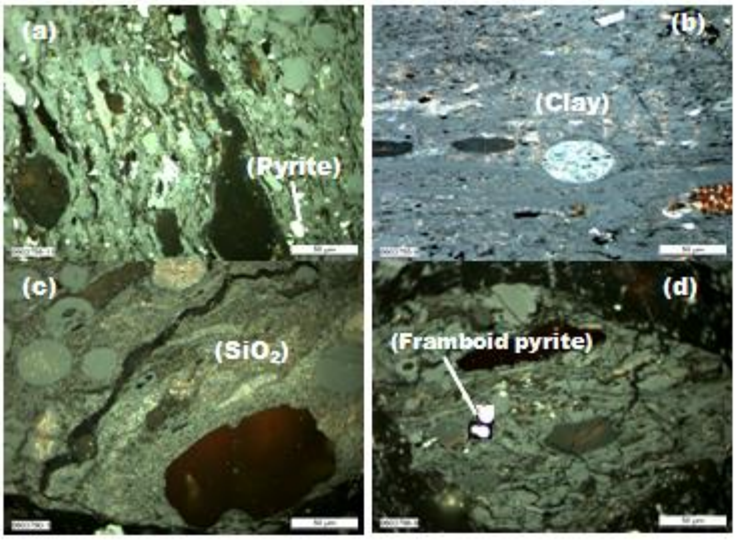


Figure 3: Microphotographs of mineral matters present in Ihioma lignite. Magnification x 5.

Table 1: Mineral matter concentration in Ihioma lignite samples based on microscopic observation

LOCATION	CARBONATE		PYRITE		ARGILLACEOUS	
	Range (vol%)	Mean (vol%)	Range (vol%)	Mean (vol%)	Range (vol%)	Mean (vol%)
Ihioma A	5.20 - 32.20	12.25	1.80 - 14.80	5.76	2.20 - 4.80	3.20
Ihioma B	2.50 - 14.40	5.34	0.60 - 4.80	1.98	0.20 - 1.70	0.96
Ihioma C	7.80 - 20.20	12.25	0.40 - 4.50	2.15	1.50 - 4.30	2.90

XRD and XRF analyses

Mineralogical studies based on XRD of lignite samples indicate quartz anorthite, albite, calcite and rutile (Figure 4).

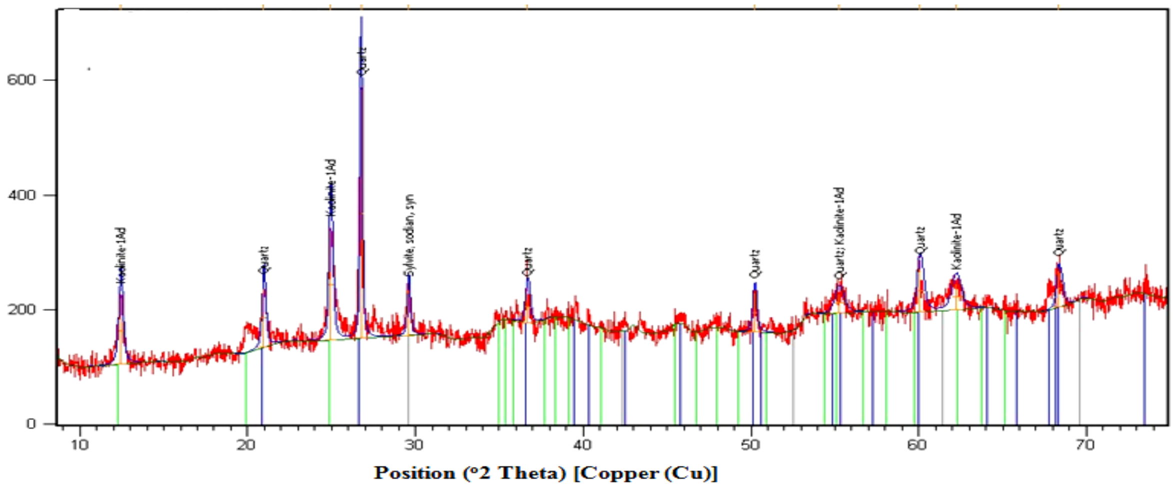
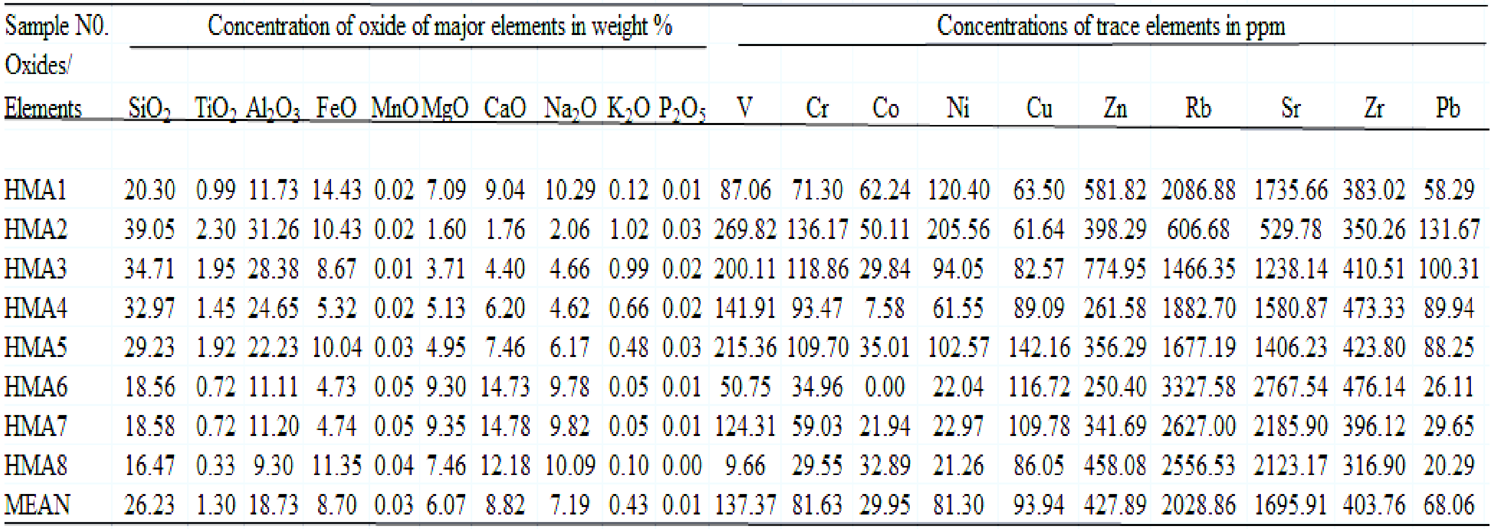


Figure 4: XRD diffractograph of Ihioma lignite.

The inorganic residue of the analyzed lignite samples comprises average SiO_2 (26.23%), Al_2O_3 (18.73%), FeO (8.70%), CaO (8.82%), Na_2O (7.19%), MgO (6.07%), TiO_2 (1.30%), K_2O (0.43%), P_2O_5 (0.01%), and MnO (0.03%). The average concentration (in ppm) of selected trace elements Rb (2028.86), Sr (1695.91), Zn (427.89), Zr (403.76), V (137.37), Cu (93.94), Cr (81.63), Ni (81.30), Pb (68.06), and Co (29.89) in the ash of lignite samples is recorded (Table 2).

Table 2: Geochemical analysis of major oxides and trace element concentrations of lignite ash using XRF technique.



Sample N0.	Concentration of oxide of major elements in weight %										Concentrations of trace elements in ppm									
Oxides/ Elements	SiO_2	TiO_2	Al_2O_3	FeO	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	V	Cr	Co	Ni	Cu	Zn	Rb	Sr	Zr	Pb
HMA1	20.30	0.99	11.73	14.43	0.02	7.09	9.04	10.29	0.12	0.01	87.06	71.30	62.24	120.40	63.50	581.82	2086.88	1735.66	383.02	58.29
HMA2	39.05	2.30	31.26	10.43	0.02	1.60	1.76	2.06	1.02	0.03	269.82	136.17	50.11	205.56	61.64	398.29	606.68	529.78	350.26	131.67
HMA3	34.71	1.95	28.38	8.67	0.01	3.71	4.40	4.66	0.99	0.02	200.11	118.86	29.84	94.05	82.57	774.95	1466.35	1238.14	410.51	100.31
HMA4	32.97	1.45	24.65	5.32	0.02	5.13	6.20	4.62	0.66	0.02	141.91	93.47	7.58	61.55	89.09	261.58	1882.70	1580.87	473.33	89.94
HMA5	29.23	1.92	22.23	10.04	0.03	4.95	7.46	6.17	0.48	0.03	215.36	109.70	35.01	102.57	142.16	356.29	1677.19	1406.23	423.80	88.25
HMA6	18.56	0.72	11.11	4.73	0.05	9.30	14.73	9.78	0.05	0.01	50.75	34.96	0.00	22.04	116.72	250.40	3327.58	2767.54	476.14	26.11
HMA7	18.58	0.72	11.20	4.74	0.05	9.35	14.78	9.82	0.05	0.01	124.31	59.03	21.94	22.97	109.78	341.69	2627.00	2185.90	396.12	29.65
HMA8	16.47	0.33	9.30	11.35	0.04	7.46	12.18	10.09	0.10	0.00	9.66	29.55	32.89	21.26	86.05	458.08	2556.53	2123.17	316.90	20.29
MEAN	26.23	1.30	18.73	8.70	0.03	6.07	8.82	7.19	0.43	0.01	137.37	81.63	29.95	81.30	93.94	427.89	2028.86	1695.91	403.76	68.06

DISCUSSION

The microscopic study of polished coal samples provides valuable information about common minerals in the coal, as well as their optical characteristics, size, distribution, crystalline structure and morphology (Vassilev and Tascón, 2003). Microscopically, the individual minerals are very difficult to identify due to this reason they have been identified as argillaceous, carbonate and sulphide minerals. They occur as strips and lenticles or may occupy cracks, fractures, and cleat infillings visible at microscopic as well as macroscopic level (Singh and Singh, 1995; Singh *et al*., 2019). In coal, mostly mineral matters are fine-grained, although with exception of occurrences of pyrite (Renton, 1982). A pyrite strip of 0.5 -1.0 cm has also been observed within the lignite seam.

Sulphides are also characteristic of authigenetic mineral of coal with marine influence and they are commonly enriched in Fe sulphides as pyrite (Vassilev and Vassileva, 1996) (Figure 3a, d). According to Singh *et al*. (2019) presence of framboidal pyrite indicates these lignites influenced by the marine incursion (Figure 3d).

Pyrites occur as infilling in cell cavities and cell lumens in fusinites and semifusinite and mainly in vitrinite patches (Singh and Singh, 2000) (Figure 3b, c). Clay minerals occur in the pores, cell cavity and oxidation cracks of the maceral (Dai *et al*., 2015, 2012; Singh and Singh, 1995) indicative of authigeneic and syn-depositional origin (Figure 3b). Clay minerals, being clastic in nature, also suggest these lignites derived from the detrital origin (Singh *et al*., 2019).

XRD and XRF

XRD is the most widely used tool for identification and some extent characterization of minerals present in coals. Previous studies (Ward, 1989; Ward and Taylor, 1996) reported the kaolinite is a very common constituent of various coal seams; although in some cases have only clay minerals (Speight, 2005).

Important elements reported in the lignite samples such as Si, Al, and Ti associated with SiO_2, Al_2O_3 and TiO_2 respectively. The concentration of major oxides in lignites of Ihioma reveals the predominance of SiO_2 (26.23%) and Al_2O_3 (18.73%) which indicate more clastics derived by high energy depositional condition (Ayinla *et al*., 2017). In addition, a positive correlation has been recorded between TiO_2 – SiO_2, TiO_2 – Al_2O_3 and TiO_2 – K_2O (Figure 5) in studied lignites (Singh *et al*., 2019).

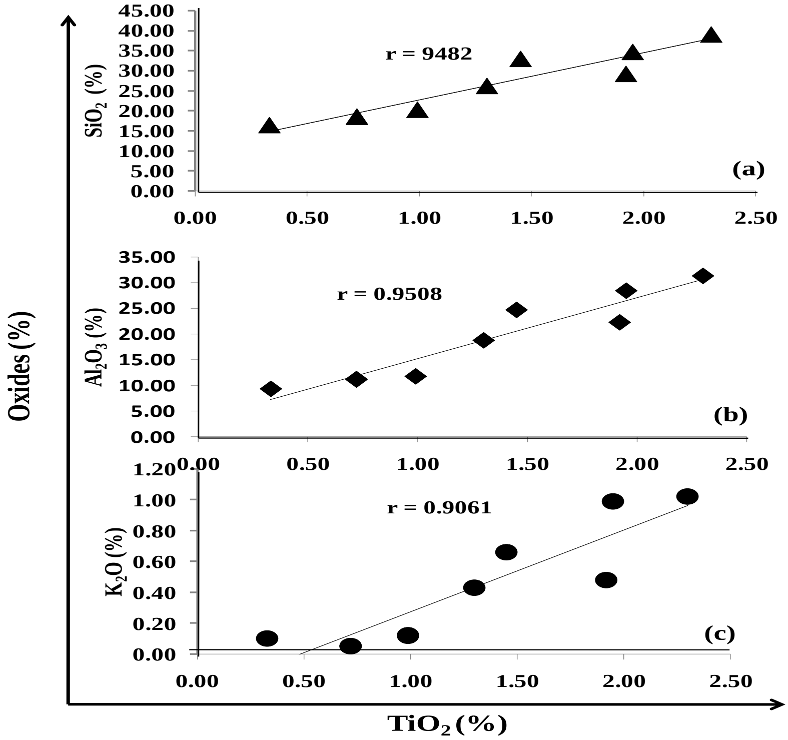


Figure 5: Correlation between major elements oxides for Ihioma lignites (a) SiO_2 versus TiO_2 (b) Al_2O_3 versus TiO_2 (c) K_2O versus TiO_2 ((adapted after Singh *et al.*, 2019).

The concentration of trace elements such as Sr, Rb, and V has been used to discriminate between seawater and freshwater condition (Makeen *et al.*, 2015) while redox sensitive elements, such as V, Ni, Cu, and Cr have been used to understand the paleodepositional setting (Armstrong-Altrin and Machain-Castillo, 2016). The concentrations of trace elements in ppm show dominance of Rb, Sr, Zn, and Zr, followed by V, Cu, Cr, Ni, Pb, and Co. Vanadium (V) and nickel (Ni) are indicators of redox conditions during sedimentation process (Galarraga *et al.*, 2008; Rimmer, 2004). The plot between V and Ni (Figure 6) indicates that the deposition of organic matter took place in terrestrial oxic to marine and terrestrial oxic to the sub-oxic condition.

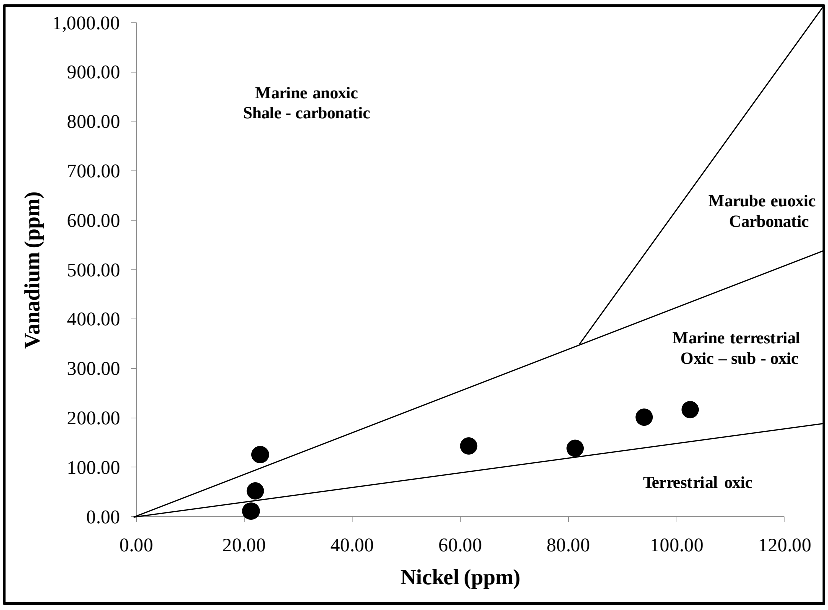


Figure 6: Plot vanadium versus nickel in ppm showing the depositional condition for the Ihioma lignite (adapted after Galarrga *et al*., 2008).

CONCLUSION

This study indicates a variety of minerals in the lignite samples. Pyrite band is also identified at the macroscopic scale. Dominance of carbonates, sulphides and clay minerals are reported at the microscopic scale in the lignites of the study area. The presence of these minerals and their mode of occrrence suggest that Ihioma lignites have undergone a series of syngenetic and authigenic mineralization process. XRD based mineral study reveals that quartz mineral is dominant along with occurrence of muscovite, biotite, sedirite, talc, coesite, aragonite, anorthite, albite, calcite, and rutile. The major oxides in lignites of Ihioma are predominated by SiO_2 and Al_2O_3 indicating a high energy deposition. Trace elements concentration suggests deposition under terrestrial oxic to marine and terrestrial oxic to the sub-oxic condition.

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