

GEOCHEMISTRY, FACTOR ANALYSIS AND CLAY MINERAL DISTRIBUTION OF THE SEDIMENTS AND RELATIONSHIP WITH THE ASSOCIATED FERROMANGANESE NODULES FROM THE SW CARLSBERG RIDGE

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ABSTRACT

Calcareous clay and ooze from the southwestern Carlsberg Ridge show wide range of concentrations in Si (4.62-23.88%), Al (1.63- 7.98%), Ti (0.43-1.70%) and CaCO₃ (8.50-79.75%) and reveal association of the elements with carbonate, and hydrothermal ferromanganese oxide phases. R-mode factor analysis produced 5 major factors - biological, detrital, hydrothermal and authigenic origin for the concerned sediments and further represents a transition from hydrothermal to authigenic origin. Geochemical data indicate that Cu and Ni have resulted from biogenic processes, whereas Fe, Zn and Co are from hydrothermal source. The Mn/Fe ratios (0.08-0.36 for sediments; 0.90-1.48 for nodules) and relationship of Mn with Cu, Ni and Zn for nodules and sediments suggest diagenetic supply of Mn⁺² through pore waters during early diagenesis. Distribution of clay minerals show that illite and smectite are more abundant than kaolinite and chlorite, whereas palygorskite and montmorillonite occur as poorly crystalline phases. Illite and chlorite are derived from Indus source due to dispersion in the southern latitudes whereas smectite and kaolinite are authigenic and point towards the alteration of submarine volcanic rocks.

Key-words : Geochemistry, sediments, clay minerals, ferromanganese nodules, R - mode factor, Carlsberg Ridge.

INTRODUCTION

Earlier studies on the sediments from the Arabian Sea are of regional interest (Stewart, Pilkey and Melson, 1965; Gorbunova, 1966; Goldberg and Griffin, 1970; Böstrom and Fisher, 1971; Kolla, Henderson and Biscaye, 1976; Kolla, Be and Biscaye 1976; Kolla, Ray and Kostecky, 1981; Kolla, Kostecky, Robinson, Biscaye and Ray, 1981; Shankar, Subbarao and Kolla, 1987). Although the association of ferromanganese nodules in the sediments from the Carlsberg Ridge is known, the previous studies do not throw light on the sediment - nodule relationship mainly because the work was done separately on the nodules (Cronan and Tooms, 1967 and 1969) and on the sediments (Gorbunova, 1966; Goldberg and Griffin, 1970; Kolla, Henderson and Biscaye, 1978; Kolla, Be and Biscaye, 1978; Kolla, Ray and Kostecky, 1981; Kolla Kostecky, Robinson, Biscaye and Ray, 1981; and Shankar, Subbarao and Kolla, 1987). The studies on the clay minerals of the sediments from the Carlsberg Ridge were based on limited sampling (Gorbunova, 1966; Goldberg and Griffin, 1970; Kolla, Henderson

and Biscaye, 1976). The present study describes major and minor element geochemistry of the sediments and the distribution of clay minerals. The nature of sediments is determined by carrying out chemical, mineralogical and R-mode factor analysis. The source and origin of the sediments is discussed using factor analysis and clay mineralogy. The chemical data of nodule and sediment are compared to observe the possible relationship, if any.

MATERIAL AND METHODS

Twenty sediment samples were collected by Petterson grab (Fig. 1) during 86th and 87th Cruises of R.V. *Gaveshani* in 1981. The maximum water depth is 5400 m and the sea bottom topography is rugged. Pleistocene sediments are white and dark brown calcareous oozes. Foraminiferal oozes are encountered on the topographic highs and siliceous oozes in the valleys.

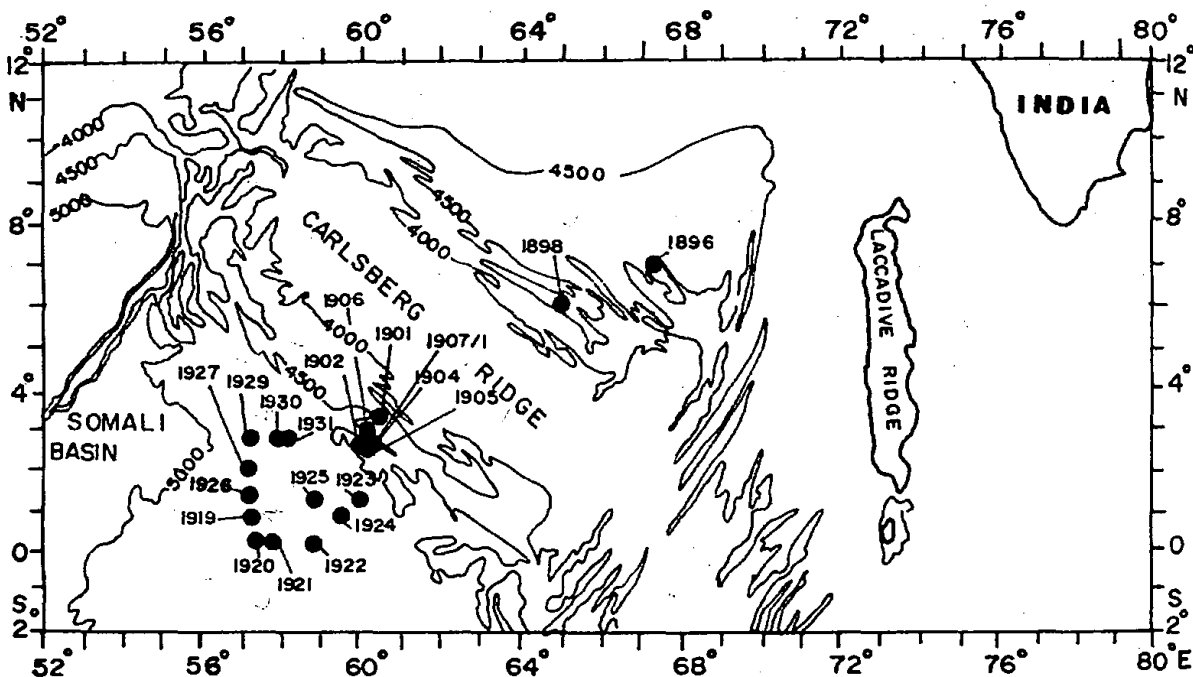


Fig. 1. Sampling locations on the Carlsberg Ridge.

The sediment samples were washed, oven dried, ground and treated with HCl, HF and HNO₃ for total digestion. Sufficient care was taken to make sure that the samples were free of any nodule fragments. Major elements Mn, Fe, Cu, Ni, Co, Zn, K and Mg were measured by atomic absorption spectrophotometry (AAS, PE-5000), whereas Si, Al, Ti, and P were determined on Spectronic 1000 following Shapiro (1975). The CaCO₃ content was determined by 'Karbonate Bombe' (Müller and Gastner, 1971). The analytical precision and accuracy were checked by using the USGS standards and by replicate analyses which was found to be <10%. The clay fraction

(<2 μm) from the samples were rendered CaCO_3 free by treating with 10 ml glacial acetic acid whereas the organic matter was removed by adding 5 ml H_2O_2 . Oriented samples were prepared by pipetting the clay suspension and air dried. Clay mineralogy was determined by running the glycolated samples (at 100°C for 1 hr) on Philips PW 1728 X-ray diffractometer (XRD) from 3° to 30° 2θ using Ni filter and $\text{Cu K}\alpha$ radiation. The clay minerals were identified according to the method of Brindley and Brown (1980) and the weighted proportion of the major clay minerals were estimated (Biscaye, 1965).

The geochemical data were further subjected to varimax rotated R-mode factor analysis using computer programme of Fernandes and Mahadevan (1982). Ferromanganese nodules associated with the sediments were analysed on AAS for Mn, Fe, Cu, Ni, Co and Zn, according to the method given in Valsangkar, Rajamanickam, Karisid-daiah and Siddiquie (1988). The chemical data of the nodules were used to compare with that of the sediments.

RESULTS AND DISCUSSION

The metal content (bulk sediment basis) of the sediments are given in Table I. The analyses in oxide form (not shown) sum to between 99.41 and 100.59. The samples contain variable amounts of moisture, oxide and organic matter. Large variations are seen for SiO_2 (9.89 - 51.10%), Al_2O_3 (3.08 - 15.21%) and CaCO_3 (8.50 - 79.75%) contents. In comparison with pelagic clays the Carlsberg Ridge sediments have an equal concentrations of Ni, Co and Zn, and are depleted in Mn, Fe, Cu and Al. Variation in TiO_2 (0.46 - 2.84%) is significant whereas that of P_2O_5 (0.28 - 0.38%) is not pronounced.

The main reason for subjecting bulk data for factor analyses is to provide room for the factors associated with carbonate fraction of the sediment. The factor loading matrix (on bulk basis) after orthogonal - varimax rotation (R-mode factor, Table II) accounting for 96.9% of the total variance suggests that five factors are responsible for the observed variation in the geochemistry. The factor analysis further represents combination of more than one source and also transition between two processes.

Average chemical composition of the ferromanganese nodules associated with the sediments are given in Table III which includes 53 analyses of 18 specimens from 9 locations in the study area. Seven types of nodules are recognised of which Sr (spheroidal rough) nodules are more abundant than the others. The nodule abundance varies from 1.1 to 5.8 kg/m^2 . Weathered rock fragments commonly occur in the nodule cores and δ - MnO_2 is the common mineral phase in these nodules.

Compared to the Indian Ocean averages (Cronan and Moorby, 1981) Ni, Cu and Zn contents of these nodules are similar whereas those of Mn, Fe and Co concentrations are lower. Spheroidal and ellipsoidal nodules with rough surface texture (Sr and SEr respectively) show a gradual increase of Mn, Fe, Cu, Ni, Co and Zn from core to the outer layer whereas in other types of nodules an inverse trend is noticed.

Table I. Bulk Chemical composition of the sediments from the Carlsberg Ridge.

Stn. No.	Depth in m	Moisture %	Mn %	Fe %	Cu %	Ni %	Co %	Zn %	CaCO ₃ %	Mg %	K %	Al %	Si %	Ti %	Mn/Fe
1896	5100	6.32	0.51	4.05	0.02	0.03	0.03	0.01	08.50	3.68	2.31	7.98	23.88	1.70	0.12
1898	4400	3.24	0.32	2.58	0.01	0.02	0.01	0.01	42.05	2.68	1.52	5.33	14.63	1.45	0.12
1901	4400	3.40	0.46	3.01	0.01	0.02	0.02	0.01	36.72	3.10	1.53	6.20	16.03	0.93	0.15
1902	4400	2.70	0.24	1.61	0.01	0.02	0.00	0.01	62.50	2.05	0.83	3.50	08.71	0.84	0.15
1904	4500	1.74	0.08	0.75	0.01	0.01	0.01	0.00	79.75	1.66	0.41	1.63	04.62	0.43	0.11
1905	4800	6.56	0.62	2.83	0.02	0.04	0.02	0.01	34.90	2.48	1.40	6.05	16.03	0.98	0.22
1906	4500	3.34	0.44	2.44	0.02	0.03	0.01	0.01	45.00	2.41	1.23	5.29	14.09	1.06	0.18
1907/1	4500	3.40	0.44	2.55	0.02	0.02	0.01	0.01	52.87	2.39	1.02	4.31	11.40	0.79	0.20
1919	4700	3.62	0.25	2.19	0.01	0.01	0.00	0.00	60.80	1.87	0.66	2.79	10.10	0.55	0.12
1920	4700	4.64	0.10	1.25	0.02	0.06	0.01	0.03	59.75	1.81	0.64	2.86	10.75	0.43	0.08
1921	4700	2.50	0.10	1.08	0.05	0.04	0.01	0.06	69.37	1.86	0.69	1.95	08.18	0.48	0.09
1922	4600	3.00	0.08	0.86	0.01	0.04	0.01	0.04	70.25	1.71	0.54	2.18	07.74	0.27	0.10
1923	4700	5.38	0.39	2.40	0.02	0.03	0.02	0.01	43.76	2.44	1.17	5.29	13.66	0.84	0.16
1924	5100	7.68	0.40	3.23	0.02	0.03	0.02	0.01	20.52	2.74	1.56	6.82	20.98	1.18	0.13
1925	4000	2.74	0.19	1.36	0.01	0.02	0.00	0.01	65.62	1.91	0.72	3.12	08.07	0.57	0.14
1926	4800	3.96	0.22	1.51	0.03	0.04	0.01	0.04	62.50	1.94	0.76	3.33	08.60	0.68	0.15
1927	4600	4.38	0.13	1.41	0.03	0.03	0.01	0.02	47.88	2.45	1.17	5.26	13.00	0.93	0.09
1929	4600	5.06	0.49	2.29	0.02	0.02	0.02	0.01	41.62	2.31	1.12	4.44	14.42	1.11	0.21
1930	4700	4.84	0.67	1.86	0.02	0.01	0.02	0.01	51.25	2.29	1.01	4.24	12.05	0.93	0.36
1931	5400	7.56	0.27	3.30	0.03	0.01	0.02	0.01	25.75	2.97	1.69	6.78	18.38	1.38	0.08
Av.	4660	4.30	0.32	2.13	0.02	0.03	0.01	0.02	49.07	2.34	1.10	4.47	12.77	0.88	0.15

Table II. Factor loading matrix (after varimax rotation, on bulk basis) for Carlsberg Ridge sediments

Factor	1	2	3	4	5
CaCO ₃	0.96	-0.02	-0.01	-0.14	-0.19
Mn	-0.48	0.11	-0.15	0.09	0.83
Fe	-0.88	0.11	-0.14	0.23	0.26
Cu	-0.09	-0.97	0.13	-0.01	0.00
Ni	0.05	-0.22	0.96	0.07	-0.06
Co	-0.69	-0.13	-0.09	-0.08	0.30
Zn	0.31	-0.68	0.54	0.00	-0.24
Mg	-0.92	0.02	-0.14	0.25	0.08
K	-0.96	0.00	-0.07	0.20	0.11
Al	-0.93	0.06	-0.05	0.24	0.18
Si	-0.95	0.02	0.04	0.17	0.16
Ti	-0.93	0.00	-0.22	-0.02	0.14
P	-0.40	0.00	0.09	0.91	0.02
Sum of the squares of each column of the above factor loading matrix are					
	7.064	1.505	1.370	1.101	1.055

Table III. Average chemical composition of manganese nodules from the Carlsberg Ridge*.

Stn. No.	Mn %	Fe %	Ni %	Cu %	Co %	Zn %	Mn/Fe	Growth ** rate (mm/Ma)
1902	12.75	11.77	0.29	0.10	0.09	0.05	1.13	2.93
1907/1	13.33	10.80	0.43	0.17	0.29	0.09	1.28	3.11
1908/3	11.66	12.93	0.56	0.17	0.15	0.08	0.90	2.74
1925	17.19	11.62	0.52	0.23	0.14	0.08	1.46	3.22
1932/2	15.75	12.27	0.43	0.17	0.16	0.08	1.28	3.03
1933	14.26	13.64	0.53	0.21	0.12	0.10	1.20	2.80
1934	15.38	13.00	0.47	0.18	0.13	0.11	1.18	2.92
1935/1	16.25	11.67	0.23	0.12	0.12	0.09	1.39	3.15
1935/2	16.51	11.52	0.57	0.18	0.12	0.07	1.43	3.20
I.O.A.	15.40	14.80	0.46	0.27	0.22	0.07	1.04	2.75

* - After Valsangkar, Rajamanickam, Karisiddaiah and Siddiquie (1988);

I.O.A. - Indian Ocean Average, from Cronan and Moorby (1981);

** - Sharma and Somayajulu (1987). Ma - Million Yrs.

Factor Analysis

Normally observed very strong positive correlation (matrix) between Cu-Ni-Zn is weakly reflected in the R-mode factor analysis which suggests that the major component CaCO₃ is acting as a diluent on the phases containing other elements. Factor 1 is heavily loaded with CaCO₃ and can be considered to be a biogenic

carbonate factor. This is further supported by negative loading of Al which would be expected to represent in the case of true biogenic factor (Cronan, 1986).

Factor 2 has a high negative loading for Cu, Zn and Ni whereas Mn and Fe show a weak positive loading. Further, some of the samples with high scores on factor 2 (supposed to be authigenic) have high scores on factor 5 (hydrothermal). This factor therefore reflects the transition from hydrothermal to authigenic ferromanganese oxides. Cronan (1980), also observed similar transitional factor for the equatorial NE Pacific Ocean sediments.

Factor 3 has high loading for Ni, moderate for Zn and a weak loading for Cu. This is more likely to be an authigenic (Smectite) factor, since authigenic manganese oxides are characteristically rich in Ni and Cu but poor in Fe and Al (Friedrich, 1976). Cronan (1986) also mentioned enrichment of Ni in authigenic marine ferromanganese oxide factor.

Factor 4 is heavily loaded with P and has comparatively weak loading for Mg, Al, Fe, K and Si. Factor loading matrix calculated on MOC (Moisture, Organic carbon and CaCO₃) free basis (not shown) however does not show high loading for P and a weak loading for other elements as mentioned earlier. It is therefore clear that P is carried by CaCO₃ (authigenic apatite ?) acting as a diluent. Phosphorus can also be removed directly from sea water by bacterial process (O'Brien, Harris, Milnes and Veeh, 1981). Factor 4, therefore, accounts for biological contribution to the sediments and excess of biogenic silica in some of the samples supports this view. Factor 4 is also characterized by scores on Mg, Al, Fe, K and Si which represents transported dust or detrital material. The factor 4, therefore, represents a contribution of both biological and detrital contribution.

Factor 5 has high loading for Mn and a weak loading for Co and Fe. This would suggest a hydrothermal ferromanganese oxide factor because they (hydrothermal factors) are generally depleted in Ni as reported by Cronan (1980).

Clay mineral distribution

Identification of clay minerals shows presence of illite, smectite, chlorite, and kaolinite along with quartz as a common detrital phase. Palygorskite is poorly crystalline and montmorillonite is a non-crystalline phase (Fig. 2). Illite (26.93 - 87.97%) and smectite (13.30 - 43.18%) are dominant clay minerals than kaolinite (3.03 - 11.46%), chlorite (9.63 - 25.62%) and palygorskite (0.68 - 5.92%). Gorbunova (1966) and Kolla, Henderson and Biscaye (1976) reported wide spread occurrence of 10 - 20% montmorillonite along with palygorskite respectively from the present study area, whereas our data shows poor occurrence of palygorskite. If the reported montmorillonite is considered to represent the smectite group (Hathaway, 1979) then, the smectite percentage in the present study should have closely matched with those from Gorbunova (1966) and Kolla, Henderson and Biscaye (1976), which in fact is much

higher (>20% for all the samples, excepting 1898, Table IV). The chlorite content determined in the present study is also much higher (9- 25%, Table IV) than the reported 1% by Gorbunova (1966) and <10% by Kolla, Henderson and Biscaye (1976). The percentages of illite determined matches quite closely with those reported (20% to >50%) by Kolla, Henderson and Biscaye (1976).

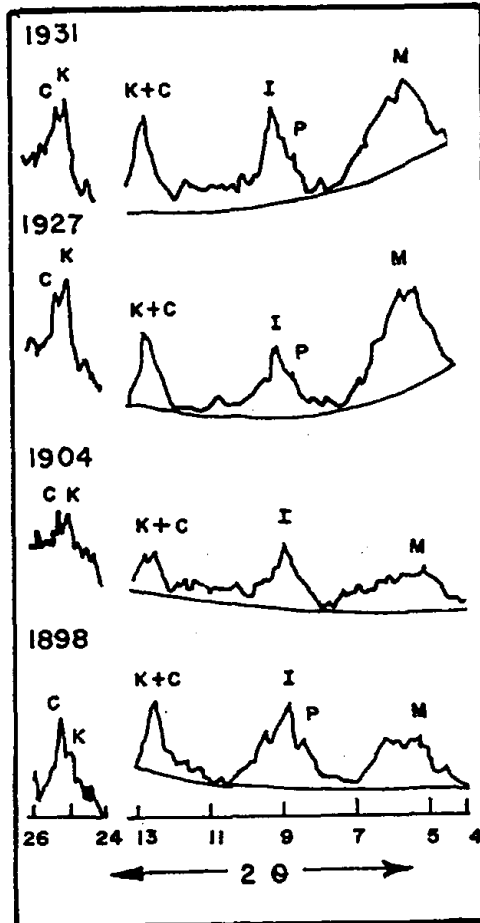


Fig. 2. X-ray diffractograms of glycolated clay samples showing illite (I), montmorillonite (M), smectite (S), kaolinite (K), chlorite (C) and palygorskite (P).

Although deep-sea clay is normally considered of detrital nature, Biscaye (1965) mentioned that the formation of clay minerals *in situ* on ocean bottom in the Indian Ocean is significant. Kolla, Henderson and Biscaye (1976) indicated that the study area is suitable for *in situ* province. The fact that the study area is far away from the continent, the increased abundance of (kaolinite + chlorite) from NW to SW can be attributed to the *in situ* alteration of submarine basalts. *In situ* formation of kaolinite has also been reported by Biscaye (1965), but Hathaway (1979) opined kaolinite as a

Table IV. Distribution of major clay minerals in the sediments from the Carlsberg Ridge

Str. No.	Smectite (%)	Illite (%)	Kaolinite (%)	Chlorite (%)	Palygorskite (%)
1896	21.28	60.50	3.30	15.17	-
1898	13.30	62.08	6.01	19.25	1.33
1902	24.29	52.33	6.48	16.87	-
1904	17.58	67.75	4.88	9.76	-
1905	26.42	54.31	9.63	9.63	-
1906	34.26	41.71	5.35	16.05	2.60
1907/1	41.07	26.93	9.40	22.57	-
1919	32.61	32.61	9.15	25.62	-
1920	24.41	43.89	6.03	24.82	0.82
1921	29.43	44.14	5.90	19.17	1.33
1922	43.18	39.09	4.01	13.03	0.68
1923	39.37	33.16	7.06	18.83	1.55
1924	33.02	44.03	11.46	11.46	-
1925	22.06	57.01	9.30	10.23	1.37
1926	28.96	46.89	7.54	16.58	-
1927	34.56	33.58	10.91	15.01	5.92
1929	26.47	48.52	9.34	14.54	1.10
1930	22.01	55.77	5.27	15.82	1.10
1931	27.31	44.51	11.36	14.76	2.02

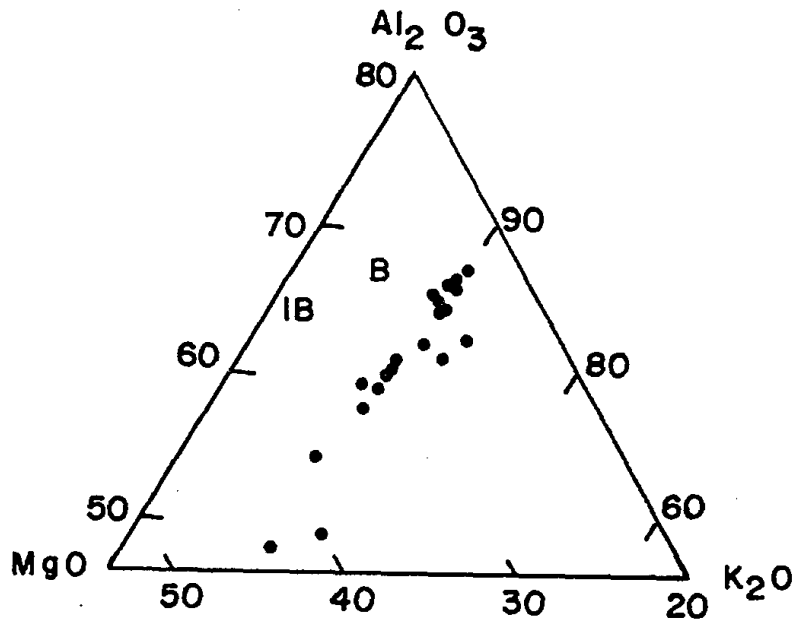


Fig. 3. Al_2O_3 - MgO - K_2O relationship for the sediments and rocks. B-average basalts (Daly, 1933). IB - average oceanic basalts (modern MORB) and Indian Ocean Tholeiites (Glikson, 1983).

product of chemical weathering. Sugisaki and Kinoshita (1980) pointed that mutual relation of Al-Mg-K elements does not change greatly during sedimentation on the ocean floor. Examination of the relations among Al_2O_3 -MgO-K₂O is, therefore, helpful in ascertaining the sediment sources. The compositional points in Fig. 3 are closely associated with the world ocean averages of basalts (Daly, 1933) and modern Mid Ocean Ridge Basalts (Glikson, 1983) suggesting that portion of the sediments are derived from the *in situ* alteration of submarine basalts. Böstrom and Fisher (1971) thought of submarine volcanism responsible for the high Fe values (8-10%) obtained for the sediments in the study area. However, in the present study, majority of the samples have Fe values ranging from 1 to 3% which do not support the view of the major contribution coming from submarine volcanism.

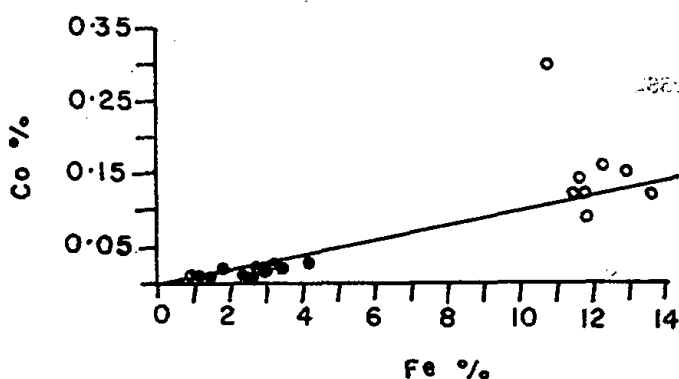


Fig. 4. Relationship between Fe and Co contents.
● - sediments, ○ - nodules.

Palygorskite is absent or rare in the sediments derived from the Indus and Ganges sources (Kolla, Henderson and Biscaye, 1976). Poor occurrence of palygorskite in the illite rich sediments under study, therefore, points towards the Indus source which is about 3000 km away from the area in the northwest. The Indus sediment is considered to be dispersed largely by surface circulation and by turbidity currents during Pleistocene sea level lowering (Kolla, Henderson and Biscaye, 1976).

Gorbunova (1966) also reported minor occurrence of montmorillonite in the western Arabian Sea and suggested their formation due to decomposition of volcanic material. Nearly same concentration of Mn and Fe in the nodules (Table III) suggests that very little Fe was utilized for the formation of montmorillonite and most of Fe was incorporated in the nodules favouring the formation of δ - MnO_2 and not the todorokite.

The formation of smectite is either from *in situ* alteration of submarine basalts, and/or by wind blown volcanic ash, which is also thought to be responsible for the presence of smectite in the sediments from the Central Indian Ocean Basin (CIOB) where according to Kolla, Henderson and Biscaye (1976), the long distance sediment

transport has less affected the sediments. Similar source is also thought for the another *in situ* province in the Indian Ocean where the high smectite in the sediments are associated on aseismic 90°E ridge (Kolla and Rao, 1990).

Sediment - Nodule Relationships

An examination of the relationship between the minor elements in the sediments and nodules show much scatter of the plots and suggest independent sources for these elements.

When authigenic elements Mn, Cu and Ni in the sediment undergo post depositional mobilisation under reducing conditions at deeper levels in the sediments, they migrate and finally precipitate near sediment-water interface under oxidising conditions in the form accessible for the ferromanganese nodules. The nodules growing under such conditions should show an increase in the content of these elements from the innermost layer to the periphery. This hypothesis is already tested for the ferromanganese nodules from the study area where Valsangkar, Rajamanickam, Karisiddaiah and Siddiquie (1988) showed progressive increase in Mn, Cu and Ni from fore to the outer layers. Cu and Ni can replace Mn in δ - MnO₂ structure (mineralogical control) after being scavenged by Mn oxides in the sea water (Burns and Burns, 1977). The source of Cu and Ni is thought to be biological as this is an important factor in the multivariant analyses. Halbach, Hebisch and Scherhag (1981) have also considered biological source for Cu and Ni which were carried along the radiolarian tests in the equatorial Pacific region.

Strong correlation between Fe and Co (Fig. 4) suggests replacement of other elements by Co. Favourable conditions for Co are typically oxidising conditions (such as on seamounts) which should also be reflected from the present study. For this, the support from mineralogy of the nodules can be considered which is discussed by Valsangkar, Rajamanickam, Karisiddaiah and Siddiquie (1988) and showed the presence of highly oxidised δ - MnO₂ phase. According to Craig, Andrews and Meylan (1982) Co⁺³ can substitute Mn⁺⁴ in the δ - MnO₂ matrix. The hydrothermal activity on the spreading Carlsberg Ridge may also be responsible for the enrichment of non-lithogenic base metals such as Co.

Mn/Fe ratios for the associated sediments and nodules from 3 stations (1902, 1907/1 and 1905) vary from 1.13 to 1.48 for nodules (Table III) and 0.08 to 0.36 for the sediments. It is possible that much of the Mn and Fe in the nodules is coming from the sediments which is reflected in the higher values of Mn/Fe ratios; which otherwise would have been similar for both the nodules and sediments, in the absence of interaction between them (Price, 1967). The nodule/sediment enrichment factor shows highest enrichment for Mn (44.56 times) and least for Zn (4.5 times). For Fe, Ni, Cu and Co the enrichment is 5.69, 17.66, 10.5 and 12.0 respectively.

CONCLUSION

Variation in depth (4000 to 5400 m) has reflected very well in the CaCO_3 content of the calcareous sediments from the Carlsberg Ridge area. Calcium carbonate is acting as strong diluent on the phases of other elements. Elemental variations suggest their origin by biological, detrital, hydrothermal and authigenic processes and also transition between hydrothermal to authigenic process. Calcium carbonate, Cu and Ni are formed biogenically whereas Fe, Zn and Co are the result of hydrothermal process. Clay minerals study indicate illite and chlorite as derived from Indus source whereas smectite and kaolinite are authigenic due to *in situ* alteration of rocks. The sediment-nodule relationship supports hydrothermal activity and post-depositional mobilisation of Mn, Cu and Ni.

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