

Variation in size, morphology and chemical composition of polymetallic nodules from the Central Indian Ocean Basin

A.B. VALSANGKAR, S.M. KARISIDDAIAH and G. PARTHIBAN

National Institute of Oceanography, Dona Paula, Goa - 403 004, India.

Abstract - Chemical composition of 613 polymetallic nodules from 150 stations in the Central Indian Ocean Basin (CIOB) are determined and variations in Mn, Fe, Cu, Ni, Co, Zn and moisture content are studied with respect to their size and surface texture. Using these parameters nodule forming processes governing the size are discussed and three models for the formation of nodules are suggested.

In CIOB, the water depth varies from 4700 to 5550 m and the topography is mainly uneven. About 25 nodule types have been recognised of which spheroidal and ellipsoidal nodules of small to medium size with rough surface texture (Ssr and Ser) have 70% of occurrence. X-ray diffractograms of 79 samples showed todorokite peak in small size nodules. The peak area of todorokite decreases from small to larger size nodules.

At every station, the average concentration of Mn, Cu, Ni, Co and Zn decreases whereas that of Fe and moisture content increases from medium (2-4 cm) to very large size (> 10 cm) nodules. Fe and moisture content is maximum in larger nodules; Mn, Cu, Ni and Zn in smaller nodules and Co is maximum in smaller as well as bigger (6-8 cm) size nodules. Mn shows strong positive correlation with Cu, Ni, Zn and Mn/Fe whereas Mn and Fe are negatively correlated in all size nodules. Fe shows strong association with Co and moisture content.

The average Mn/Fe (3.69) is higher than that of the Pacific Ocean nodules (2.76). The large variation in Mn/Fe (2.97 to 5.55) indicate multiple source of supply for the elements and suggest different accretionary processes for nodule formation. Nodules of specific size group have common morphological, mineralogical and chemical properties and their variations are best explained by proposing three models. The smaller nodules are formed diagenetically in the presence of pore water supply whereas the larger nodules indicate hydrogenous precipitation.

INTRODUCTION

Polymetallic nodules are spread over all ocean basins which vary in composition and abundance. The nodules cover larger area in the Pacific than in the Indian and Atlantic Oceans. The study related to the Pacific Ocean nodules is extensive and several papers appeared which discussed numerous fundamental problems such as the ones by HORN *et al.* (1972); GLASBY (1977); BURNS and BURNS (1977). Limited published literature exists on the Indian Ocean nodules (BEZRUKOV and ANDRUSHCHENKO, 1974; CRONAN, 1977; SIDDIQUIE *et al.*, 1978; FRAZER and WILSON, 1980; CRONAN and MOORBY, 1981) before the National Institute of Oceanography (NIO), Goa, undertook several expeditions since 1982 to the central Indian Ocean Basin (CIOB). So far more than 60 research papers have been published from the NIO, wherein they discussed biology (PARULEKAR *et al.*, 1982), reserves (SIDDIQUIE *et al.*, 1984), morphology (MUKHOPADHYAY, 1987), mineralogy (RAO, 1987), topography (PATTAN and KODAGALI, 1988), and geochemistry and formation of different sized CIOB nodules (VALSANGKAR and KHADGE, 1989).

Although there has been quite good amount of contribution towards the problems posed to nodule composition and origin, the mystery of formation of deep-sea nodules

is an unsolved scientific problem at present. This has stimulated the present paper where correlation of nodule types and composition has been established for over 600 nodules covering a very large area (28938 sq.km) in the CIOB where first generation mine site has been indicated. Formation of different sized nodules due to variation in accretionary processes are discussed and three conceptual models are proposed to explain the observed variations in the composition with respect to nodule size.

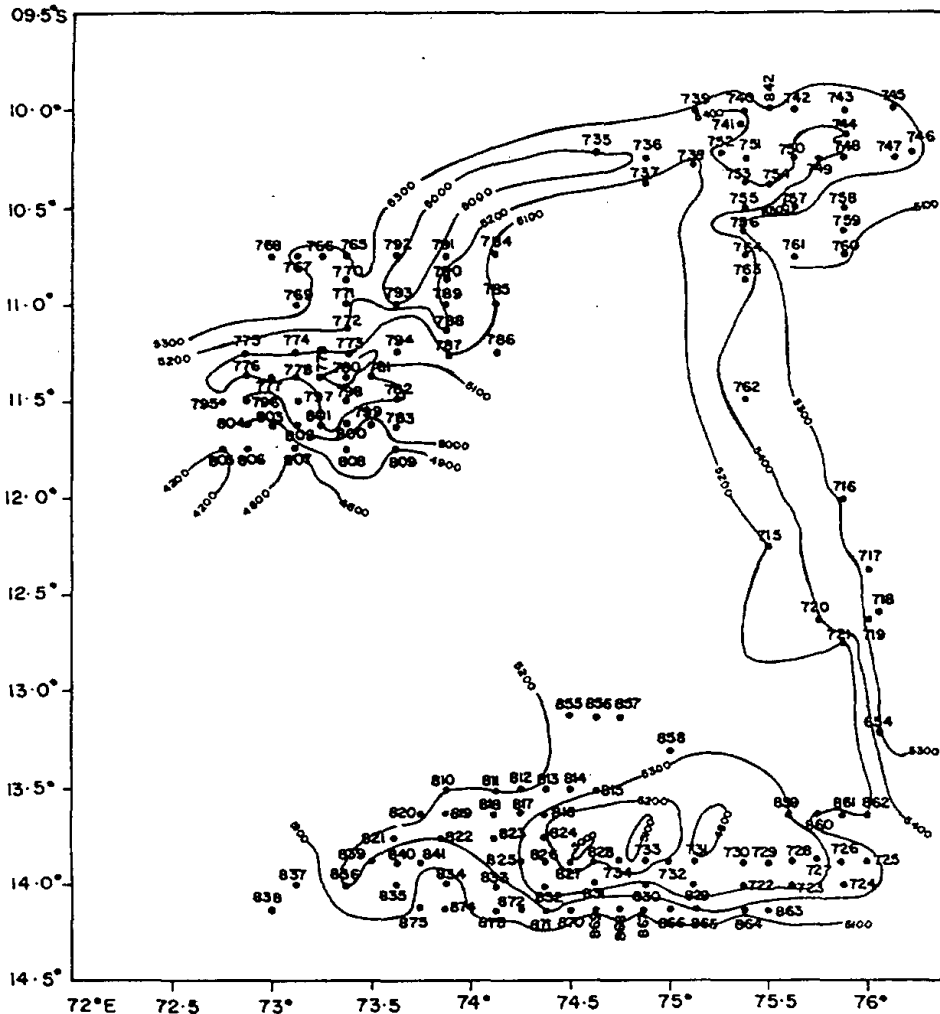


Fig. 1. Sample locations in the Central Indian Ocean Basin. o - Sample number. Contours are in meters (in thousands).

METHODS

Sampling was done by boomerang grabs during cruises of *M.V. Skandi Surveyor* in 1987-88 (SS-10; SS-14 to SS-17). From each station (Fig.1) different size nodules were selected for the study which totalled 4 to 6 depending on availability. Shape, size and surface texture of each nodule was described before it was crushed for bulk analyses. Accurately weighed 0.1 gm nodule powder was dissolved in 10 ml 1:1 HCl, 5 ml HClO₄ and 5 ml HF by digesting on sand bath and slow heating. Elemental composition of Mn, Fe, Cu, Ni, Co and Zn was determined on atomic absorption spectrophotometer (AAS), double beam, model PE-5000; and the moisture content was calculated by deter-

mining dry weight at 110°C. X-ray diffractograms (XRD) of 79 samples were obtained on Phillips diffractometer model PW 1729.

Table 1. Distribution of different size nodules in CIOB

Size Range (cm)	Small < 2	Medium 2-4	Inter-mediate 4-6	Big 6-8	Large 8-10	Very large >10	Whole Data < 2 to > 10
Nodule Surface Texture (%)							
Rough	87.50	68.62	68.64	69.23	100.00	66.67	69.09
Smooth	12.50	30.00	26.69	26.92	-	33.33	28.04
Rough to Smooth	-	1.38	4.66	3.85	-	-	2.87
Nodule Types (%)							
Spheroidal	4.50	51.35	32.05	13.33	-	-	40.95
Ellipsoidal	4.50	22.97	38.46	13.33	-	-	28.33
Discoidal	-	5.86	1.92	6.67	75.00	-	4.52
Elongated	-	1.80	2.56	33.33	-	100.00	5.00
Irregular	-	18.02	25.00	33.33	-	-	21.19
Poly-nucleate	-	17.84	25.08	25.71	25.00	-	21.38

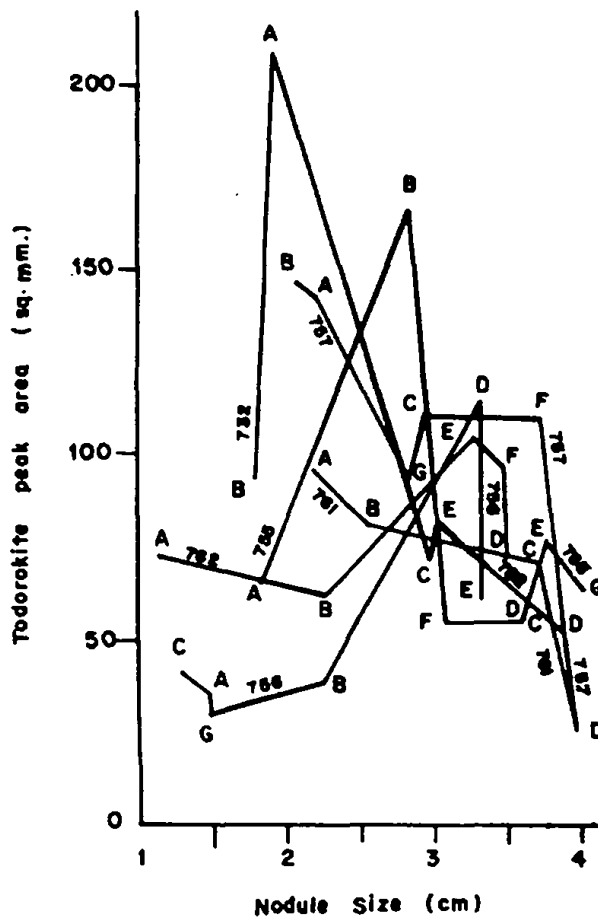


Fig. 2. Relation between todorokite peak area and nodule size, sample numbers are in figures and alphabets denotes subsamples.

Table 2. Range of chemical concentration in CIOB nodules

	Small 2cm n=8	Medium 2-4cm n=309	Intermediate 4-6cm n=237	Big 6-8cm n=48	Large 8-10cm n=5	Very large > 10cm n=6	Whole Data n=613	Average NE Pacific nodules*
Moisture (%)	9.25-20.94	1.94-23.54	4.46-23.40	7.70-23.30	5.30-22.04	13.86-19.04	1.94-23.54	NA
Mn (%)	6.87-29.26	1.43-48.58	0.18-33.10	13.21-42.04	14.96-26.38	19.22-20.30	0.18-48.58	0.04-50.30
Fe (%)	2.97-12.90	0.30-14.81	0.68-20.51	0.00-14.70	0.61-12.83	0.52-11.77	0.00-20.51	0.03-50.00
Cu (%)	0.26- 1.69	0.07- 2.73	0.06- 2.64	0.16- 2.43	0.56- 1.33	0.46- 1.06	0.06- 2.73	0.00- 1.90
Ni (%)	0.39- 1.69	0.10- 2.02	0.10- 1.88	0.34- 1.61	0.69- 1.89	0.63- 1.26	0.10- 2.21	0.08- 2.48
Co (%)	0.03- 0.30	0.03- 1.08	0.03- 0.22	0.04- 0.81	0.05- 0.15	0.08- 0.11	0.03- 1.08	0.01- 2.53
Zn (%)	0.04- 0.21	0.01- 1.14	0.02- 0.87	0.05- 0.14	0.07- 0.11	0.05- 0.10	0.01- 1.14	0.01- 9.00

* - VOLKOV (1979), NA - Data not available

Table 3. Average chemical concentrations of CIOB nodules

Size	Small	Medium	Inter- mediate	Big	Large	Very large	Whole Data	Average NE Pacific nodules*
Range (cm)	<2	2-4	4-6	6-8	8-10	>10		
No. of nodules	8	309	237	48	5	6	613	
Moisture %	14.18	16.36	17.06	16.47	15.35	17.37	16.61	NA
SD	3.84	3.73	3.58	4.18	5.52	1.78	3.74	NA
Mn %	20.07	24.29	22.95	21.82	20.72	19.37	23.48	22.40
SD	8.04	5.12	4.46	4.46	4.50	3.50	5.01	NA
Fe %	6.18	6.09	6.62	7.03	6.12	7.76	6.39	8.10
SD	3.28	2.86	3.25	3.41	4.56	2.12	3.10	NA
Cu %	0.80	1.14	1.03	0.98	0.80	0.84	1.08	1.02
SD	0.44	0.50	0.45	0.49	0.29	0.50	0.49	NA
Ni %	0.94	1.20	1.16	1.07	1.10	1.03	1.17	1.16
SD	0.46	0.35	0.35	0.32	0.42	0.50	0.35	NA
Co %	0.12	0.10	0.10	0.12	0.09	0.10	0.10	0.25
SD	0.08	0.07	0.03	0.11	0.04	0.03	0.06	NA
Zn %	0.12	0.13	0.11	0.09	0.08	0.08	0.12	NA
SD	0.05	0.09	0.06	0.02	0.01	0.02	0.08	NA
Cu+Ni+Co %	1.86	2.44	2.29	2.16	1.99	1.97	2.35	2.43
Mn/Fe	4.46	5.55	4.95	3.71	3.86	2.97	3.69	2.76

SD-Standard Deviation; * -GLASBY et al.(1986); NA-Data not available

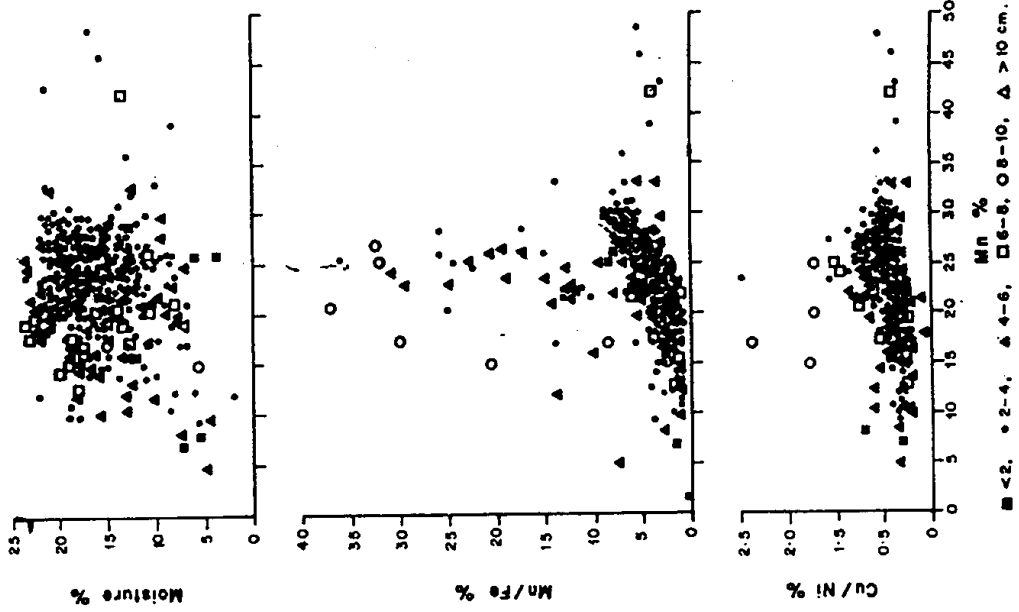


Fig. 3b. Variation in Mn/Fe, Cu/Ni and moisture content with Mn.

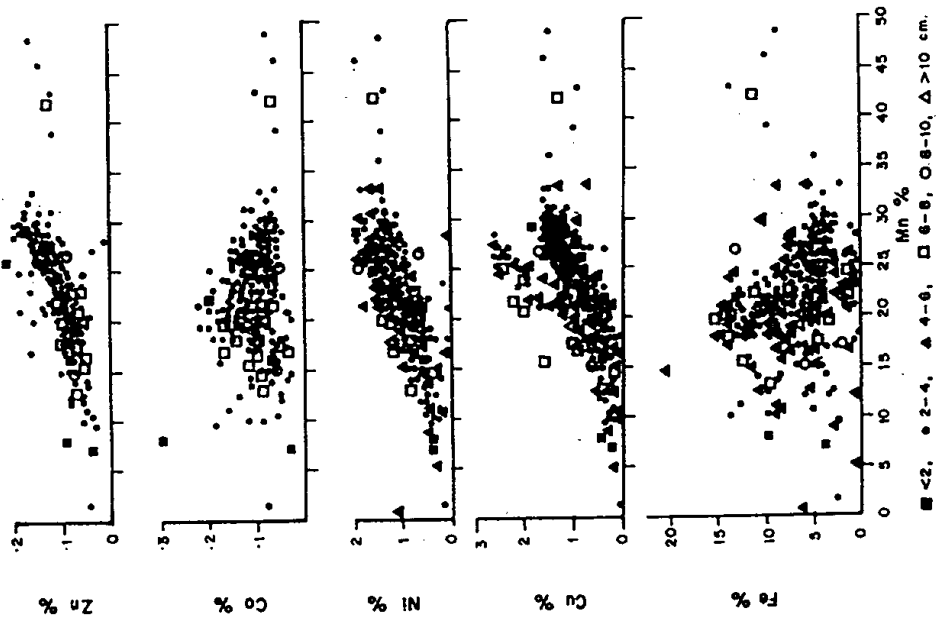


Fig. 3a. Variation in Fe, Cu, Ni, Co and Zn with Mn.

RESULTS

Morphology

Morphological study shows large variations in size, shape and surface characteristics in the CIOB nodules. In all 25 different nodule types are recognised. The nodule distribution show that the nodules of intermediate size (4-6 cm) consists maximum number of types (21) whereas that of large (8-10 cm) and very large (> 10 cm) size consist only two nodule types. Number of nodule types also vary with the nodule size which is 5 in small (< 2 cm), 18 in medium (2-4 cm) and 12 in bigger (6-8 cm) size nodules. Nodule distribution with respect to size (Table 1) further show that in small, medium and intermediate size nodules, occurrence of spheroidal and ellipsoidal types are above 70%. It is also noticed that, from small to large size nodules the percentage of occurrence for rough surface nodules decreases (87 to 66%) whereas that of smooth surface nodules increases (12 to 33%).

Mineralogy

X-ray study of 79 samples showed small and broad peaks of todorokite in small size nodules. Among the others, quartz and δ -MnO₂ (vernadite) occur as an important accessory minerals. Occurrence of phillipsite and feldspars is poor and that of birnessite is rare. It is interesting to note that peak area of todorokite decreases from small to larger size nodules (Fig. 2).

Table 4A. Correlation matrix for small size (< 2 cm) nodules

	Moisture	Mn	Fe	Cu	Ni	Co
Mn	0.27					
Fe	0.50	-0.31				
Cu	-0.01	0.79	-0.56			
Ni	0.02	0.91	-0.61	0.96		
Co	-0.07	-0.29	0.69	-0.24	-0.38	
Zn	-0.23	0.77	-0.48	0.84	0.84	-0.17

($r = 0.63$ for 95%, $n=8$, $df=n-2$)

Table 4B. Correlation matrix for medium size (2-4 cm) nodules

	Moisture	Mn	Fe	Cu	Ni	Co
Mn	0.23					
Fe	0.31	-0.31				
Cu	-0.02	0.66	-0.60			
Ni	-0.02	0.72	-0.59	0.75		
Co	0.14	-0.13	0.37	-0.25	-0.24	
Zn	-0.03	0.27	-0.22	0.36	0.21	-0.06

($r = 0.32$ for 99.9%, $n=309$, $df=2$)

Table 4C. Correlation matrix for intermediate size (4-6 cm) nodules

	Moisture	Mn	Fe	Cu	Ni	Co
Mn	0.23					
Fe	0.38	-0.34				
Cu	-0.16	0.64	-0.66			
Ni	-0.12	0.68	-0.60	0.77		
Co	0.42	-0.19	0.79	-0.54	-0.50	
Zn	0.10	0.39	-0.11	0.29	0.27	0.07

($r = 0.32$ for 99.9%; $n=237$, $df=2$)

Table 4D. Correlation matrix for big size (6-8 cm) nodules

	Moisture	Mn	Fe	Cu	Ni	Co
Mn	-0.05					
Fe	0.49	-0.19				
Cu	-0.33	0.49	-0.60			
Ni	-0.45	0.59	-0.70	0.72		
Co	0.02	-0.15	0.08	-0.17	-0.14	
Zn	-0.04	0.74	-0.31	0.54	0.52	-0.13

(r = 0.45 for 99.9%; n=48, df=2)

Table 4E. Correlation matrix for large size (8-10 cm) nodules

	Moisture	Mn	Fe	Cu	Ni	Co
Mn	0.61					
Fe	0.26	0.30				
Cu	0.20	0.32	-0.81			
Ni	0.14	0.27	-0.81	0.97		
Co	0.54	0.49	0.95	-0.61	-0.63	
Zn	0.38	0.71	-0.36	0.79	0.84	-0.15

(r=0.75 for 95%; n=5, df=2)

Table 4F. Correlation matrix for very large size (10cm) nodules

	Moisture	Mn	Fe	Cu	Ni	Co
Mn	0.44					
Fe	0.19	-0.03				
Cu	0.38	0.23	0.70			
Ni	0.34	0.20	0.69	0.99		
Co	0.24	0.08	0.94	0.89	0.88	
Zn	0.54	0.11	0.87	0.90	0.89	0.94

(r = 0.71 for 95%; n=6, df=2)

Table 4 G. Correlation matrix for CIOB nodules - Whole Data

	Moisture	Mn	Fe	Cu	Ni	Co
Mn	0.19					
Fe	0.36	-0.27				
Cu	-0.09	0.58	-0.61			
Ni	-0.08	0.64	-0.59	0.76		
Co	0.17	-0.12	0.41	-0.28	-0.27	
Zn	0.00	0.29	-0.19	0.35	0.25	-0.06

(r= 0.32 for 99.9%, n=613, df=n-2)

Geochemistry

As large number of samples have been analysed it is difficult to present all the analyses. Range and averages are given in Tables 2 and 3 respectively.

Average chemical composition (Table 2) show good correlation with nodule size. In fact, nodules of specific size group have common chemical properties. Medium size (2-4 cm) nodules have high Mn (24.29%), Cu (1.14%), Ni (1.20%) and Zn (0.13%), whereas very large size (> 10 cm) nodules have high Fe (7.76%) and moisture (17.37%). Similarly, medium size nodules have high Cu+Ni+Co (2.44%) and high Mn/Fe ratio (5.55). The Cu+Ni+Co content and Mn/Fe ratio decreases whereas Fe and moisture content increases with increase in nodule size. Average values of Co and Zn are nearly

constant. Variation in elements (Fe, Cu, Ni, Co, Zn, moisture and Mn/Fe ratios) with respect to Mn for 613 analyses is shown in Fig. 3a and 3b..

Table 4 (A to G) shows correlation matrix for different size nodules in the CIOB. It is observed that Mn correlates very strongly with Cu, Ni and Zn in all size nodules except for very large nodules of > 10 cm (Table 4F). Similarly, Fe correlation with Co is strong in all size nodules except for big nodules of 6-8 cm size (Table 4D). Excellent correlation is seen between Cu, Ni and Zn in all size nodules. Also, good correlation between Ni and Zn can be seen for all the nodules except for medium and intermediate size. Very large nodules of > 10 cm size also show strong correlation of Co with Fe, Cu, Ni and Zn which is not duplicated in other nodules. It is also interesting to note that moisture content show good correlation with Fe for medium (2-4 cm, Table 4B), intermediate (4-6 cm, Table 4C), big size nodules (6-8 cm, Table 4D) and also for the whole data (Table 4G).

DISCUSSION

Morphology

Although 25 different nodule types have been recognised, there can be many more in other parts of the CIOB. The major morphological types are spheroidal and ellipsoidal with rough surface texture which has 70% of occurrence. These results are comparable with those from the SW Pacific. GLASBY *et al.* (1986) reported 72 to 89% of occurrence for small (<3 cm), 10 to 26% for medium (3-6cm) and 1 to 4% for large (> 6cm) nodules from 3 basins in the SW Pacific where spheroidal and ellipsoidal nodules together have 50 to 88% of occurrence. Higher occurrence of small to medium size nodules of spheroidal and ellipsoidal shape in both the Indian and Pacific Oceans therefore suggest their formation more or less under similar conditions with faster growth rates in comparison with the other nodule types.

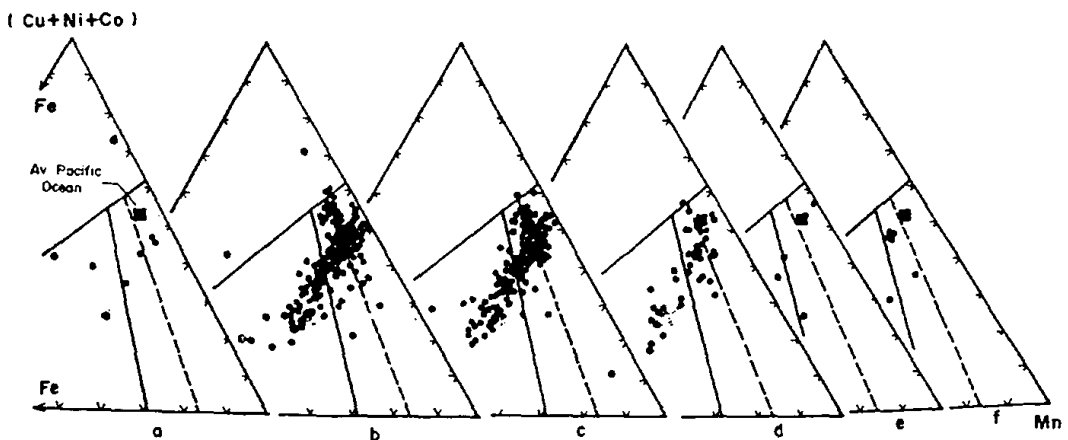


Fig. 4. Triplot of Mn-Fe-(Cu+Ni+Co)X10 for (a) < 2 cm, (b) 2-4 cm, (c) 4-6 cm, (d) 6-8 cm, (e) 8-10 cm and (f) > 10 cm size nodules. ■ - Average Pacific Ocean, GLASBY *et al.* (1986)

Geochemistry

The chemical composition of CIOB nodules vary with respect to size and type of nodules. GLASBY *et al.* (1982) also reported similar compositional variations with respect

to morphology for three different nodule types in the Pacific. An inverse correlation between nodule size and major ore elements (Table 3) suggest formation of different size nodules at different locations. Higher concentrations of Mn, Cu, Ni and Zn only in the case of medium (2-4 cm) and intermediate (4-6 cm) size nodules clearly indicate that these nodules were formed during diagenesis and were favoured due to additional supply of source through pore waters. The smaller size is due to faster growth and the resulted rough surface texture is because of longer contact with sediment-water interface as noted by USUI *et al.* (1987). This is further supported by their high Mn/Fe ratios in medium and in intermediate size nodules which are 5.55 and 4.95 respectively. Medium and intermediate size nodules have more abundance of todorokite which is revealed from the higher peak area (Fig. 2). Todorokite mineral being the main carrier of Mn, Cu and Ni elements, the higher percentages of Mn, Cu and Ni are expected only in the case of medium and intermediate size nodules owing to the depletion of todorokite abundance in the larger nodules, in the present study.

On the other hand, the larger nodules which have lower concentrations of Mn, Cu, Ni and Zn suggest their formation by hydrogenesis thereby allowing enrichment in Fe and moisture content. The larger nodules therefore have lower Mn/Fe ratios. Changes in the rate of sedimentation is inferred from such large variations in Mn/Fe ratios (PIPER and WILLIAMSON, 1977). A triplot of Mn-Fe-(Cu+Ni+Co)x10 for different size group of the CIOB nodules (Figs. 4 a to f) show a spread between oxic diagenetic and hydrogenous accretionary processes. Large variations in Mn/Fe ratio (2.97-5.55) according to (HALBACH *et al.*, 1981) indicate early diagenesis and multiple accretionary processes (VALSANGKAR *et al.*, 1988) supporting the above view.

The CIOB nodules can be grouped into six different sizes. Nodules of each size group have nearly constant chemical, mineralogical and morphological characteristics. Different size nodules therefore indicate their formation under different accretionary processes which were active at different locations with varying intensity.

The sampling area consists mainly of siliceous ooze (SUDHAKAR, 1989). VALSANGKAR and KHADGE (1989) considered high biological productivity in the CIOB to explain the high Mn/Fe ratios and high Cu+Ni content in nodules. Such favourable conditions are expected near carbonate compensation depth (CCD). The high grade nodules in the NE Pacific Ocean are also thought to be controlled by high biological productivity and CCD (EXON, 1983; CRONAN, 1987).

Following geochemical models are suggested for the observed variation in morphology and chemical composition of CIOB nodules with respect to size :

i) *CCD Model* : According to BELYAEVA and BURMISTROVA (1985), the average CCD in the CIOB is about 4735 m. In the regions of high silica formation in surface water, the siliceous- diatomaceous organisms after their decomposition (*i.e.* CCD) on the seafloor, release Mn, Cu and Ni and thereby allow silica enrichment in the adjacent sediments. The elements thus released act as an additional source which are absorbed in the nodules through pore waters during diagenesis. The nodules grow rapidly but to smaller size with rough surface texture. Such nodules typically show enrichment in Mn, Cu and Ni; high Mn/Fe ratios and have todorokite as principal mineral phase.

ii) *Deep-sea model* : Maximum water depth in the study area is 5500 m which is deeper than CCD and therefore indicate lack of biological productivity. The nodules therefore grow slowly by hydrogenesis in the absence of additional source. Thus, larger size nodules are formed which show enrichment in Fe and moisture content. They predominantly have smooth surface texture, low Mn/Fe ratios and δ -MnO₂ (vernadite) as major mineral phase.

iii) *Mixing model* : Diagenetically formed smaller nodules at lower depths near CCD are moved towards deeper water regions due to bottom currents where they mix with the larger nodules formed by hydrogenous precipitations.

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