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Alteration of Basaltic Glasses from the Central Indian Ocean

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Abstract: Textural, mineralogical and compositional characteristics of basaltic glasses from the Central Indian Ocean show them to be altered to varying extents through their interaction with the seawater, resulting in the formation of palagonite. The major chemical changes are the loss of Si, Mg and Ca and a gain of Na and K whilst, Fe and Ti remained immobile. It reflects that the basaltic glasses have undergone initial to intermediate stages of palagonitisation under low temperature oxidative alterations. Subsequent accretion of ferromanganese oxides over the glasses have halted the progress of alteration.

Keywords: Geochemistry, Basalts, Palagonitisation, Central Indian Ocean.

INTRODUCTION

Investigations till date have revealed that ocean floor volcanics show degrees of weathering, either as low, moderate or high, which result from their interaction with the seawater. Weathering of an outcrop on the seafloor depends on factors such as the presence of joints, fractures and fissures on its surface and also on its mineralogy and age. A number of physico-chemical changes may occur during the weathering process. The main processes involved in the alteration of the oceanic crust are related to weathering by seawater, metamorphism, hydrothermal circulation and deuteric compositional changes, brought about during and after the formation of the rocks. The purpose of studying altered materials from the seafloor is to understand the chemical budget of seawater, the circulation of fluid in the crust, the type of tectonic activity involved, the nature of transport and re-deposition of material extracted from the basement rocks and formation of hot springs and ore deposits on the seafloor.

Reviews of alteration of dredged and drilled basalts in the world oceans include Honnorez (1981) and Thompson (1991). According to Thompson (1991) the effects of seawater-rock interactions can generally be considered under the term "metamorphisms" which are the result of two processes: low temperature ("weathering" or halmyrolysis) and high temperature alteration. The former which occurs at <70°C, is pervasive, ubiquitous and continues for long periods of time (>106 years). The latter process is hydrothermal and occurs between 70-400°C or higher and is generally restricted to sites near accreting plate margins or where localised mid-plate volcanism may be present and is relatively of short interval (102 to 104 years). In either case, the precursor rock would exhibit changes in its physical, chemical and mineralogical properties.

Basaltic pillow lavas erupted on the seafloor undergo quenching resulting in the formation of glass which preserves the nearly pristine magma chemistry. The zone below the glassy top being in a closed chemical environment develops into a holocrystalline interior. It has been reported that the alteration of basaltic glasses results in a greater degree of compositional variations than the holocrystalline interior of the rock (e.g., Melson, 1973; Thompson, 1973). Investigations of altered basalts by Hart (1970), Matthews (1971) and Melson (1973), among others, led to the conclusion that low temperature weathering (halmyrolysis) rather than high temperature alteration was the predominant mechanism and that the glass was more highly affected than the

interior, while the other highly contrasting conclusion is that the pillow glassy rims are fresher than pillow interiors (Corliss, 1971; Macdougall, 1973). But the fact remains that in course of time the pillow basalt would be altered to different extents under various physico-chemical conditions. One notable alteration of glass is to palagonite but the pathways of formation is not fully understood because it involves multiple processes such as hydration of the glass, exchange of certain elements with seawater and progressive devitrification and crystallisation of the resultant product (Moore, 1966).

Three stages of palagonitisation of basaltic glass have been recognized by Honnorez (1978). The initial stage is characterised by the coexistence of fresh glass relicts with residual altered glass ("palagonite") and alteration minerals, that are essentially a Ca-poor Na≥K phillipsite and K- and Mg-rich smectites (probably saponite). This leads to an enrichment in alkalis (mainly K) and Mg and loss of Ca while the concentration of the other elements is unchanged. During the mature stage, the fresh glass is fully altered and the residual glass begins to be replaced in situ by zeolites and smectites. In the last stage, the residual glass is replaced completely by an aggregate of authigenic minerals. Further, the process of alteration leading to palagonitisation of glass has been suggested to occur almost instantaneously at high temperature during eruption (Bonatti, 1965). Alternatively, it has been suggested that palagonitisation occurs gradually after the emplacement of lava flows in a submarine environment (Moore, 1966), a view which is more popular. Hekinian and Hoffert (1975). studying the basalts from the rift valley in the Atlantic Ocean, reported a linear relation between the thickness of manganese and palagonite encrustations on the rocks that help to infer their relative rate of accumulation.

Deep-sea basalts subjected to alteration undergo a number of chemical changes and such rocks may either act as source or sink for various elements, relative to the seawater. In spite of the fact that there is no unanimity about the behaviour of many of the major elements, few broad generalizations have been reported (Honnorez, 1981; Thompson, 1991). An altered basalt acts as a source for Si, Mg, and Ca and as a sink for Ti, K, P, Mn, total Fe and Na. Additionally, ferric iron replaces ferrous iron while Al shows no definite exchange with the seawater (Hart, 1970). K and water are the most effective and noticeable parameters of alteration. Ca is depleted while K is enriched and an inverse relation is noted between them. Further, a close association between the immobile nature of Fe and Ti have been reported (Scarfe and Smith, 1977; Andrews, 1977).

In this background of contrasted views regarding alteration of ocean floor basalts, Iyer (1995) noted the different degrees of alteration of the Central Indian Ocean (CIO) basalts. In this communication the textural, mineralogical and compositional changes in the CIO basalts are presented with a view to understanding the controls of the nature and extent of the alterations.

MATERIALS AND METHODS

Seven dredged basalts (# 81, 6, 14, 24, 26, 85 and 87) recovered from various sites in the CIO (Fig. 1) were examined for the presence of glass and their state of alteration. Thin section of the selected samples were made for petrographical examination. The veneer of glass present on the samples was pressed out using a stainless steel knife and was finely ground and aliquots prepared for ICP-AES analysis. Water content and loss on ignition (LOI) were determined at 100°C and 1000°C, respectively. X-ray diffractometry (XRD) of few samples was also carried out.

GEOLOGICAL SETTING

The general bathymetry of the CIO shows the seafloor relief to change from high to medium, as one goes from west to east of the basin, and with an abyssal plain at the centre

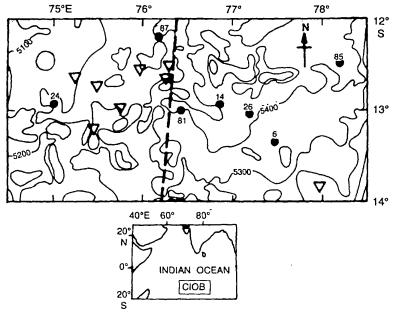


Fig.1. Bathymetry of the study area and sample locations (solid dots). Triangles represent seamounts and the dashed line the trace of a fracture zone. Contour interval = 100 m.

(Kodagali et al. 1992). A large part of the CIO has been mapped using the hydrosweep system which revealed the occurrence of numerous seamounts and abyssal hills and the presence of fracture zones (Kamesh Raju, 1993). Samples 81 and 87 are about 15 km from the 76°30' fracture zone. These two samples and also #24 are located ~30 km from the seamounts (Fig. 1). The remaining samples were recovered from the abyssal plain. In general, the area from where the basalts were recovered is one of a rolling bathymetry. From earlier studies of the CIO basalts (Mukhopadhyay et al. 1995; Mukherjee and Iyer, 1999) it was suggested that the N-MORBs erupted ~60 Ma ago from near-axis originated seamounts.

PETROCHEMISTRY OF THE CIO BASALTS

The basalts occur as fragments of various sizes with glassy top and are non-uniformly

altered to yellowish-red palagonitic materials and encrusted by ferromanganese (FeMn) oxides. The interior zone, below the glass, is partly altered. The location and petrographical description of the samples are shown in Table 1 and representative photomicrographs are presented in Fig.2. XRD shows the presence of phillipsite and plagioclase.

The major, minor and trace element concentrations in the CIO samples are tabulated (Table 2) and compared to reported analyses of altered basalts and palagonite (Table 3). SiO₂ varies between 37.10 to 43.64% in the CIO samples. TiO₂ lies between 0.68 to 1.79%, with an exception for #6 which has the highest FeO* and TiO₂ (13.85 and 2.02%, respectively). A positive relation is seen between FeO* and TiO₂ (Fig.3a) for all the samples depicted in Tables 2 and 3. The whole rock analyses of the interior of the other fragments collected with #6 represent a ferrobasaltic composition, details

Table 1. Locations and petrological details of the Central Indian Ocean altered basalts

Sample	Location (S/E)	Depth (m)	Petrological features				
81	13°00'/76°29'	5346	Altered glass				
6	13°21'/77°30'	5270	0.5 cm glass rust coloured stains. 3 distinct zones: altered glass (A), glass+interior (B) and interior (C) Zone B has submicroscopically intergrown plagical and few pyroxene. Zone C is partly altered and has plagioclase microphenocrysts. Zeolites, limonite an smectite occur.				
14	12°28'/76°52'	NA	2 layers of glass. The upper unaltered glass has olivines and is separated from the lower layer by a band of greenish-yellow palagonite. Between the bands of palagonite, zeolites occur.				
24	12°57'/75°00'	5348	Highly altered, 0.5 cm glass, rust coloured stains. Interior is buff to light gray. 2 glassy layers separated by palagonite. Uppermost glass hosts well-developed and fresh plagioclase. 2nd glassy zone is oxidised at has couple of fresh plagioclase. The interior zone shiplagioclase and glass to form intersertal texture. RFA also present.				
26	12°33'/77°12'	5200	Glass of 0.5 cm with a veneer of sediment and FeMn oxides. Plagioclases form trachytic texture. Few olivines present.				
85	12°30′/78°12′	5280	Altered glass, patches of FeMn oxides. Abundant vesicles and sheaves of plagioclase, RFA and few altered olivines.				
87	12°13'/76°11'	. 5100	Altered glass, patches of FeMn oxides in a matrix of fibrous palagonite. Abundant plagioclase microlites as sheaves and in trachytic form in a glassy ground-mass. Few fresh euhedral plagioclase phenocrysts. Altered and cracked euhedral olivines. Sub-ophitic and RFA textures				

NA- Not available; RFA = Red feathery alteration texture (cf. Baragar et al. 1977)

of which are given elsewhere (Iyer et al., 1999). MgO is uniformly low and CaO has been drastically leached out in almost all the samples. The alkali content is variable and in most cases Na₂O dominates over K_2O . MnO is variable (0.25 % to 7.29 %) due to the presence of FeMn oxide encrustations over these samples.

To understand the alteration process which involves the removal or addition of certain elements, binary plots were made. On a plot of K₂O vs CaO+MgO (Fig.3b, after Matthews, 1971) the reverse trend is consistent with the alteration of plagioclase and olivine and formation of smectite. Plots of K₂O, CaO and MgO vs LOI depict that K₂O increases linearly

with increasing alteration except in 2 samples (85 and 87; Fig. 4) which have higher MnO (6.75 and 7.29 %, respectively). CaO displays a strong negative trend with LOI (Fig. 4) indicating leaching of lime with increasing alteration. MgO, though appears to be scattered but shows higher content for less altered samples; further, for a given LOI content, the variation in MgO is small (Fig. 4). On the basis of the FeMn oxides, the altered basalts fall in two groups: the group I (#81, 26, 85 and 87) has >2% MnO and group II (#6, 14 and 24) has <2% MnO. A plot of K₂O vs CaO+MgO+MnO (Fig. 5) depicts these two groups. Group I displays higher K₂O which

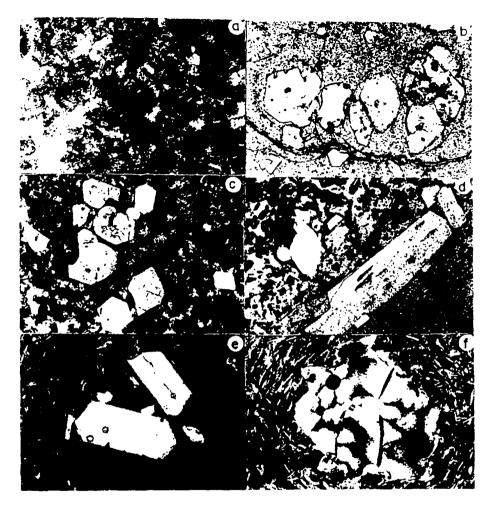


Fig. 2. Representative photoimicrographs of the altered basalts (x40) are eard funder plain light and bid under crossed medis). a. Altered basalt with submicroscopic sheaves of plagioclase. Observe the small elliptical shaped blobs around nucleus of plagioclase microlites. This texture resembles the fred feathery alteration of Baragar et al. (1977). b. Phenocrysts of olivine in a clear glassy groundmass. The glass is minimally altered ic. Phenocrysts of olivines in a zone of palagoritised glass. Observe the well defined shapes of the olivines. The size of the olivines are 0.11 to 1.00 mm long and 0.08 to 0.6 mm wide. One squarish crystal measured 0.29x0.29 mm. Few of the olivines are altered. d. The transition from clear glass to variolitic texture with varioles around nucleus of plagioclase microlities. Couple of well formed plagioclase crystals can be noted. e. A patch of unaftered glass with couple of fresh eithedral plagioclase phenocrysts. f. Interior zone of the basalt with pilotaxitic texture formed around a plagioclase phenocryst, a part of which was lost during polishing.

Table 2. Major (in wt.%) and trace elements (in ppm) composition of the Central Indian Ocean altered basalts. nd = not determined; LOI = Loss on ignition

Oxides	81	6	14	24	26	85	87
SiO,	43.64	39.59	41.72	41.18	38.28	37.28	37.10
TiO,	0.68	2.02	1.09	1.63	1.79	1.31	1.41
Al ₂ O ₃	14.80	14.29	15.34	17.84	13.16	13.86	14.08
Fe,O,	7.58	15.39	13.93	13.35	14.68	11.71	12.44
MgO	0.97	3.41	4.81	1.84	1.49	1.52	1.58
CaO	1.56	3.66	3.08	1.34	1.53	1.19	1.22
Na,O	5.42	2.58	2.77	2.59	2.92	3.18	3.60
K,Ô	3.74	1.97	1.91	2.82	3.18	3.01	2.94
P,O,	0.39	0.10	nd	nd	0.10	nd	nd
MnO	3.71	1.71	0.30	0.25	3.89	6.75	≱ 7.29
LOI	15.79	12.39	12.14	15.41	17.34	17.98	16.97
Ba	784	190	83	81	237	604	547
Co	160	146	90	67	486	459	429
Cu	1203	442	302	321	408	1406	1290
Cr	32	151	519	373	84	92	83
Ni	1519	588	217	101	900	2470	2224
V	191	190	220	100	182	154	123
Y	96	33	23	22	54	60	72
Zn	342	295	270	310	375	420	390
Zr	334	206	128	186	275	240	272

decreases (from 3.18 to 2.94) with increasing CaO+MgO+MnO. This probably suggests that once the hydrogenous precipitation of FeMn oxides begins, K,O tends to be preserved.

Based on their compositional similarities

(Tables 2 and 3), the CIO samples are comparable to the altered basalts and palagonite reported from other oceanic areas. This is also corroborated by the various plots in which all these samples cluster in close proximity

Table 3. Chemical composition of palagonite (1 to 5), most altered basalts (6) and red feathery alteration (7) from the world oceans. n = number of samples; na = not available

Oxides n	4	6	3	4	5	6	7	6
			3	6		1		
SiO,	42.05	53.42	44.19	41.80	37.88	41.96	47.11	47.65
TiO,	2.13	2.38	2.81	1.90	1.21	1.63	1.55	0.42
Al,Ö,	11.43	15.60	15.42	12.15	15.38	16.06	18.37	19.45
FeO*	19.31	20.58	15.72	19.96	11.97	9.39	8.91	9.80
MgO	3.95	5.15	2.82	4.06	7.56	3.69	1.84	3.44
CaO	1.48	1.11	0.60	0.72	5.44	1.49	2.34	11.03
Na,O	0.78	0.14	2.14	0.99	2.12	1.82	2.41	3.48
K,O	2.73	3.49	3.23	3.72	0.50	3.48	3.48	0.39
P,O,	na	na	na	na	na	0.01	0.22	na
MnÖ	па	0.06	na	0.02	0.21	0.08	0.14	0.08
LOI	na	na	na	na	10.99	13.99	8.52	na

Source: 1 = DSDP Leg 37 (Scarfe and Smith, 1977); 2 = DSDP Leg 37 (Andrews, 1977); 3 - Melson (1973); 4 - Baragar et al. (1977); 5 - Furnes (1978); 6,7 - Swallow Bank (Matthews, 1971); 8 = DSDP sites 332 and 335, MAR basalts (Baragar et al. 1977)

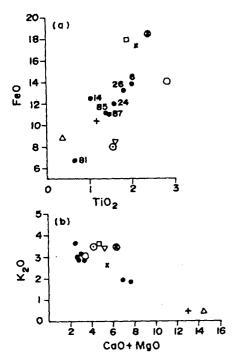


Fig.3. Plot of (a) FeO vs TiO₃ and (b) K₃O vs CaO + MgO for palagonite and altered basalts. Palagonites:-Solid dots = Present study; Cross = Scarfe and Smith (1977); Circle with cross = Andrews (1977); Open circle = Melson (1973); Square and Plus sign = Baragar et al. (1977); Inverted triangle = Matthews (1971). Altered basalts:- Circle with dot = Melson (1973); Triangle = Baragar et al. (1977).

(Figs. 3 and 4). The present study suggests that the CIO and the other oceanic basalts suffered similar alteration processes.

The concentrations of the trace elements (in ppm) are shown in Table 2. The group I samples show high Ba, Co, Cu, Ni, Y, Zn, Zr and low Cr while group II shows a trend opposite to the group I samples. Ba and Y correlate quite well with MnO (Table 1). Staudigel et al. (1980) reported that Ba is generally positively correlated with increasing K_2O , H_2O and Fe_2O_3/FeO suggestive of increasing alteration. However, Ba in the CIO samples seem to be more closely associated with MnO rather than

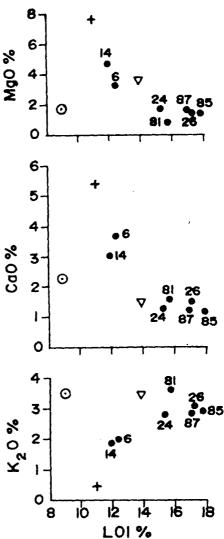


Fig.4. Plot of MgO, CaO and K₁O vs LOI for the altered basaltic glass of the CIO and other oceanic occurrences. Symbols as in Fig. 3.

with K₂O and H₂O. Such an enrichment of Ba with MnO could suggest its association with the biogenic debris or aluminosilicates (Cronan, 1980). Similarly, Co, Cu, V, Ni and Zn contents are higher in Mn-enriched samples, a feature

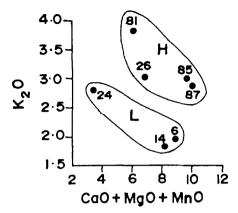


Fig.5. Plot of K.O vs CaO+MgO+MnO shows the group I and II samples as discussed in the text. L and II represent samples with <2% and >2% MnO, respectively.

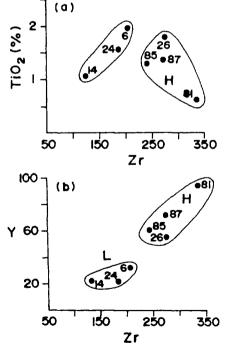


Fig.6. Plot of Zr vs Y and Zr vs TiO, for the altered basaltic glass of the CIO.

common in FeMn oxides (Cronan, 1980). Cr correlates with MgO but for an enrichment in sample #24, which may be attributed to sea water as a source (cf. Donnelly et al., 1980) or its release during the alteration of the basalts.

Since the incompatible elements Y and Zr, are not susceptible to weathering (Thompson, 1973) and Ti is rather immobile (Honnorez, 1981), plots of Zr vs Y and Zr vs TiO, are made (Fig. 6). The samples fall in two clusters depending on the MnO content. The relation between Y and Zr indicates the presence of spinel or rutile. Zr vs TiO, indicates a positive trend for group I in contrast to the group II samples. This suggests that although Zr and Ti were immobile elements during alteration, but some amount of Zr probably admixed with the phosphatic phase (fish debris?), has been removed during the leaching of P. Additionally, plots of Co, V and Zn vs CaO+MgO+MnO+FeO* were also made (Fig. 7). The proximity of samples #26, 85 and 87 which have high Co contents, point to the scavenging of Co and Zn during the accretion of the FeMn oxides. Further, the antithetical relation of V in these samples could be related to the opaque minerals of the host basalt than to the FeMn oxides.

DISCUSSION

Study of alteration of oceanic basalts has been made by many researchers using colour variations, susceptibility of glass to alteration, mineralogical and compositional changes and extent of palagonitisation. Further, the degree and kind of alteration may vary within a single pillow basalt or morphological unit, depending on the porosity, diffusion front and the pathway of the water (Thompson, 1991). In general, the reaction of basalt with seawater at low temperatures result in palagonitisation of the glass, oxidation of Fe+2 to Fe+3 and hydration and the alteration of the groundmass minerals. The most common alteration products of ocean floor basalts are palagonite, clay minerals, zeolites, montmorillonite, iron oxides and

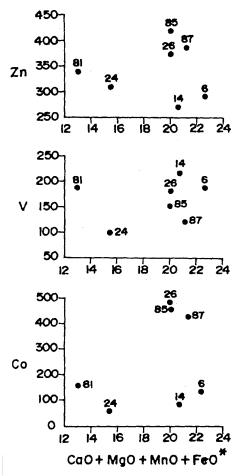


Fig.7. Plot of Co, V and Zn vs CaO+MgO+MnO+FeO* to depict their hydrogenous source and interrelationships.

hydroxides. Studies have shown that the basaltic glass is most readily replaced with smectite aggregates (Kharin, 1979). Montmorillonite is present in all size fractions of the CIO sediments whereas Fe-rich montmorillonite occurs only in <1 μ m and <1-2 μ m of siliceous and <1 μ m of pelagic clays (Rao and Nath, 1988). They opined that montmorillonite is a product of the weathering of ridge rocks whilst the Fe-rich montmorillonite is due to early diagenetic

processes in the basin. But considering the vast occurrence of volcanics in the CIO (Iyer, 1995) and the association of palagonite, montmorillonite and zeolites (Iyer and Sudhakar, 1993), it is more likely that montmorillonite is of local origin and derived from the alteration of the basaltic glasses.

Petrographic examination of the CIO samples shows the alteration of glass to palagonite, plagioclase to sericite, olivine to iddingsite and the presence of limonite (Table 1). XRD of few samples reveal the occurrence of phillipsite. The formation of phillipsite during the halmyrolysis of glass has been well documented (e.g., Bonatti, 1965, Melson and Thompson, 1973, Honnorez, 1981). In the CIO, slabs of zeolities have been recorded which were suggested to have formed by the transformation of basaltic glass to zeolites through an intermediate stage of palagonite formation (Iyer and Sudhakar, 1993).

Further, Mn-poor CIO samples exhibit significant loss in Si, Mg and Ca and variable enrichment in Na and K (Table 2). Such a severe leaching out of Si, Mg and Ca can be attributed to a process of halmyrolysis of the basalts prior to hydrogenous deposition of Fe-Mn oxides. The decrease of CaO and MgO and increase of K₂O with increasing LOI (Fig. 4) in Mn-poor samples, could represent a nearly isomolar exchange during alteration of the basaltic glass to palagonite, an observation that is in accordance with the studies of Staudigel and Hart (1983).

Present study reveals that the CIO basaltic glasses have not been fully replaced by authigenic minerals. The enrichment in Na and K could lead to the formation of Na ≥ K-phillipsite. This is consistent with enrichment of alkali metals in the basaltic glass. Further, the non-mobile nature of Fe and Ti (Fig. 3a), and their local accumulation (cf. Staudigel and Hart, 1983) together with a loss of Si, Mg, Ca and gain of Na and K, all correspond to the initial stage of palagonitisation (Honnorez, 1978; Zhou and Fyfe, 1989). Such a phase of palagonitisation, could occur under low

temperature oxidative alteration conditions (Thompson, 1991). It is apparent that these ancient CIO basaltic glasses responded in a manner similar to the described Mid-Atlantic Ridge pillow basalts (Shido et al. 1974), where the highly weathered pillows have a highly altered exterior and a less altered interior but fresh pillows have a fresh glass exterior and an altered interior. Over a period of several millions of years, the glass entirely alters to palagonite in contrast to the interior, that remains still relatively fresh (cf. Scott and Hajash, 1976).

It is generally agreed that the magnitude of changes in the trace elements is always greatest in the altered glass. The present investigation shows that the levels of the trace elements in the CIO are within the reported values for altered basalts (cf. Thompson, 1991 and references therein). For instance, Ba is enriched (Furnes, 1978) and Zn is quite constant (Donnelly et al., 1980) during the palagonitisation process. The enrichment in certain trace elements in the CIO basalts, notably Co, Cu and Ni, may be related to hydrogenous nature of the FeMn encrustations over these basalts (Fig. 7).

The following process is envisaged for the alteration leading to palagonitisation of the CIO basaltic glasses. The basaltic lava, that erupted ~60 Ma ago, chilled to form pillows with quenched glassy exterior and holocrystalline interior. In course of time there was a gradual breakdown of the glass, and depending on nature of encrustation and local environment it responded differentially to alteration. The glasses suffered a wider range of elemental change than the interior which is a holocrystalline polyphase media. This led the glasses to be altered in a continuous process while the interior was subjected to a stepwise process. The continued interaction between the basalts with the seawater and local biogenic debris brought about mineralogical and compositional changes in the former. The

mineralogical changes are noted by the occurrence of palagonite, iron oxides and phillipsite. Compositionally, there has been removal of Si, Mg and Ca, incorporation of Na and K, whilst Fe and Ti remained immobile. All these characteristics typify initial to intermediate stages of palagonitisation of the CIO basaltic glasses. Subsequent hydrogenous precipitation of FeMn oxides formed a protective veneer over the basalts, which could arrest the progress of alteration.

CONCLUSION *

The alteration of basalts, recovered from various locations in the CIO, show their glassy rims to be extensively altered resulting in the formation of palagonite and in some cases zeolites. The basalts were subjected to low temperature oxidative condition which is reflected by the mineralogical compositional changes. At a later time there has been local hydrogenous deposition and accretion of FeMn oxides over the altered basaltic substrate, and this influenced the subsequent alteration pattern as well as the trace element concentration. Due to a carapace of the FeMn oxides, the high-Mn basalts have been less altered. An increase in the number of observations, in the future, will elucidate the complex interaction between the CIO basalts and the seawater, and of the resultant products.

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620 S. D. 1YER

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