

## A hydrothermal deposit from the floor of the Gulf of Aden

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**SUMMARY.** Fragments of a hydrothermal deposit, partly moulded on pieces of basalt lava, were dredged from the edge of the median valley in the centre of the Gulf of Aden. The deposit consists of two main components, manganese oxide in the form of spongy brown to hard black lumps and coatings, and friable green massive smectite. There are smaller amounts of an iron-oxide component, and mixed manganese-oxide-smectite material. Contents of the less mobile trace elements in the smectite are too low for it to have formed by alteration of basalt glass, and it is interpreted as a direct precipitate from hydrothermal solution. None of the components is enriched in Cu or Zn, or contains appreciable amounts of sulphur. If there are sulphides in this deposit, they must lie beneath the parts sampled.

STATION 6243 of *RRS Discovery* was a dredge haul made between 12° 33'1" N, 47° 39'2" E (2550m) and 12° 35'0" N, 47° 39'9" E (2260m) in the Gulf of Aden. This position is across the north edge of the small median valley developed there, in a region where it is broken up into short lengths by repeated small transform faults. The bathymetry of the one-degree square around this area is shown as Fig. 1 in Laughton *et al.* (1970). The dredge haul brought up thin fragments of ropy basalt lava, described by Cann (1970a), as well as lumps of brown ferromanganese oxide (not originally recognized as such) and very friable lumps of yellow to green crumbly material. On the under surfaces of the lavas were masses of spongy, friable ferromanganese oxide, which was described by Glasby *et al.* (1971) and compared with ferromanganese coatings of the usual kind found on weathered basalt dredged some way from the median valley.

It now appears that the ferromanganese coatings, the lumps of ferromanganese oxide, and the yellow to green crumbly material found at station 6243 are different components of a hydrothermal deposit formed in and among lavas and sediments on the floor of the Gulf of Aden. This paper gives an initial description of the deposit.

*Description.* From their appearance, the rocks in the dredge haul were divided into six classes: (a) basalt lava, (b) hard ferromanganese oxide, (c) soft ferromanganese oxide, (d) yellow to green friable material, (e) orange powdery iron oxide, (f) soft chocolate brown material. The last class later proved to be a mixture of (c) and (d). Representative specimens of each class were selected for analysis.

(a) *The basalt lava* has been described before (Cann, 1970a). Briefly it is a nearly aphyric plagioclase olivine basalt, with a very fine-grained to glassy ground mass. Its colour is dark grey and it shows no signs of having been altered by weathering or by hydrothermal solutions. The original way up of the flow fragments can be seen from their surface structure (see Fig. 3 in Cann, 1970a), and crumbly ferromanganese oxide has accumulated under the flow fragments, occasionally spilling around to the top of the fragments, where it develops a hard and smooth black surface. Two major element analyses were reported in Cann (1970a), and are very similar

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to those of other basalts dredged from the same area. Trace elements are also well within the range of other ocean-floor basalts, except for Cu, which is anomalously high. This property is shared, though, with other basalts dredged from the same area that are not related to hydrothermal deposits, so it seems to have some other cause.

(b) and (c) *Ferromanganese oxide*. Although the hard ferromanganese oxide looked very different from the soft material, there turned out to be no sharp break in chemical or physical properties, and they are best treated together. In appearance, at one extreme is very hard, dark brown material, with a bluish sheen on broken surfaces. This compact material is well layered, and is best developed in samples that were clearly just at the water-rock interface at the ocean floor, as in the places where oxide has deposited around the edges of the thin flow fragments. Some rather hard material, of relatively high reflectivity, does occur as bands in lumps of very friable oxide. At the other extreme in appearance is a less dark brown, very soft, spongy, and friable ferromanganese oxide, occurring most characteristically beneath the thin lava fragments or as discrete lumps, perhaps broken off during dredging.

X-ray diffraction shows that all of these materials are poorly crystalline. One sample (1; Table I) shows only lines of the 10 Å phase todorokite, at 9.6, 4.7, 3.31, and 1.80 Å. Another sample (6) shows only lines of the 7 Å phase birnessite, at 7.0 and 3.54 Å. Of the other samples analysed, to judge from the relative areas of the 10 and 7 Å peaks, samples 2 and 5 are predominantly composed of todorokite, with minor birnessite, while samples 3 and 4 contain roughly equal amounts of the two phases. There seems to be no correlation of this mineralogy with the hardness or chemical composition of the samples.

Major and trace-element analyses of the six selected ferromanganese oxide samples are given in Table I. The major elements from Si to K were determined by atomic absorption spectroscopy after a lithium metaborate fusion followed by HCl extraction.  $\text{H}_2\text{O}^+$  and  $\text{CO}_2$  were determined on a Hewlett Packard CHN analyser. The trace elements were determined by XRF analysis of pressed powder samples. These six analyses are in addition to the four of Glasby *et al.* (1971), which were made by different methods for a rather different range of elements, and on samples scraped from the underside of lava flows. The samples used for the present work were all from more massive material.

The major element analyses show that the oxides are very rich in Mn in comparison with analyses of typical oceanic Mn nodules. All of the samples have an Mn/Fe ratio greater than 5, and it rises to greater than 50 in sample 5. In oceanic Mn nodules this ratio is close to 1. Bonatti (1975) has shown that such high ratios are characteristic of Mn-rich components of submarine hydrothermal deposits. Comment on the other major elements is not easy in the absence of much comparative data. They do not seem to vary in any way with the trace-element changes, and may represent contamination by other components. However, limits can be set to some contamination. For example, the Si and Fe contents limit the possible contamination by smectite (see below) to 5–10 %, so that only a small part of the Na and K content of the Mn oxide can be from this source.

The samples from station 6243 analysed by Glasby *et al.* (1971) were shown to be depleted by an order of magnitude in Fe, Co, and Pb relative to a normal manganese crust at station 6224, on a weathered basalt about 30 km from the spreading axis. Since this crust was of similar composition to other Mn nodules, these depletions also apply to global means of nodule composition. The new analyses confirm and amplify this observation.

The samples in Table I can be divided into two groups on the basis of their trace-element composition. One group (1, 2, 3, and 6) is enriched in Ti, Co, Ni, Cu, Ga, Y, and Zr relative to the other group (4 and 5). The samples analysed by Glasby *et al.* (1971) fall, as comparison of the elements analysed in common shows, in the more enriched group. The composition of the

TABLE I. *Analyses of ferromanganese oxides*TABLE II. *Analyses of smectite (d) and of mixtures of smectite, ferromanganese oxide, and iron oxide (e and f)*

TABLE I

	1	2	3	4	5	6
	2c(b)	2c(c)	3B(c)	3M(c)	3T(c)	5(c)
Si %	4.79	5.38	4.04	2.32	3.38	1.97
Al	1.11	1.11	0.84	0.06	0.69	0.34
Fe	6.39	1.31	1.53	4.52	0.76	1.51
Mn	34.22	34.89	37.33	38.83	39.82	42.45
Mg	1.09	2.21	2.01	1.60	1.50	2.17
Ca	1.41	1.74	1.49	1.50	1.55	1.52
Na	3.53	2.57	2.75	3.04	2.90	3.07
K	1.13	1.54	1.57	1.28	1.21	1.62
H <sub>2</sub> O <sup>+</sup>	9.16	8.13	9.90	10.97	9.78	10.11
CO <sub>2</sub>	1.30	1.74	0.99	0.26	0.52	0.73
S p.p.m.	1650	1100	1800	1050	900	1450
Ti	2000	1600	1100	200	700	800
Co	37	75	28	13	2	25
Ni	390	900	670	14	22	375
Cu	77	110	203	9	8	83
Zn	215	445	780	18	22	385
Ga	3	5	4	0	0	2
Ge	2	2	1	2	2	1
Rb	12	19	12	6	11	8
Sr	375	350	325	340	215	335
Y	6	11	11	1	2	7
Zr	47	63	42	15	18	28
Ba	1650	2000	1100	1000	530	660

TABLE II

	1	2	3	4	5	6
	10(d)	12(d)	24(d)	31(d)	31(e)	32(f)
Si %	22.12	22.14	21.94	21.76	10.21	14.93
Al	0.33	0.10	0.10	0.10	0.28	0.09
Fe	22.75	19.14	22.89	24.17	23.49	16.16
Mn	<0.1	<0.1	<0.1	<0.1	10.38	10.94
Mg	1.80	1.67	1.94	1.89	1.11	1.75
Ca	0.55	0.46	0.47	0.51	1.82	0.56
Na	1.24	1.02	1.27	1.45	2.37	2.00
K	3.41	3.05	3.57	2.73	1.08	2.39
H <sub>2</sub> O <sup>+</sup>	8.50	6.54	8.32	7.62	13.86	8.24
CO <sub>2</sub>	1.24	0.66	2.65	0.92	2.12	0.44
S p.p.m.	400	700	450	700	1550	900
Ti	250	—	200	200	620	—
Co	12	2	1	3	7	—
Ni	6	4	3	1	35	100
Cu	0	0	0	0	2	17
Zn	2	0	2	2	47	96
Ga	0	0	0	0	1	0
Ge	0	0	1	0	1	1
Rb	73	79	80	59	9	41
Sr	47	32	40	55	413	147
Y	2	1	2	2	6	2
Zr	7	2	9	7	27	13
Nb	2	1	1	1	0	0
Ba	120	40	65	70	400	420

Atoms per 22 oxygens, excluding H<sub>2</sub>O and CO<sub>2</sub>

Si	7.33	7.75	7.32	7.22
Al	0.11	0.04	0.03	0.03
Fe	3.79	3.37	3.84	4.03
Mg	0.69	0.68	0.75	0.72
Ca	0.13	0.11	0.11	0.12
Na	0.50	0.44	0.52	0.59
K	0.81	0.77	0.86	0.65
H <sub>2</sub> O	4.39	3.57	4.33	3.94
CO <sub>2</sub>	0.26	0.15	0.56	0.19

depleted group emphasizes the differences between this type of Mn deposit and average oceanic Mn nodules. It is depleted relative to these other materials by as much as 200 to 300 times in Co, Ni, Zn, and Zr. Similar extreme depletions in this group of elements have been reported by Bonatti *et al.* (1972) for an Mn–Ba-rich component of a hydrothermal deposit in Afar, and by Scott *et al.* (1974) for a rapidly deposited manganese coating from the Mid-Atlantic Ridge.

(d) *Smectite*. The yellow to green friable and crumbly material was described by Cann (1970a) as 'lumps of soft, crumbling hyaloclastite . . .' but the term 'hyaloclastite' now seems wrong as the material seems rather to be a smectite precipitated out of hydrothermal solution.

Thin sections of the smectite are difficult to make, and show only a structureless isotropic green material. X-ray powder diffraction work gives patterns corresponding to a very poorly crystalline smectite. The broad 14 Å basal spacing moves to 17 Å on treatment with glycerol.

No lines are seen from any other phases, and the material is treated here as composed of a single phase. Scanning electron microscope photographs of fractured surfaces show a rather botryoidal-seeming structure with included foraminifera and coccoliths. At magnifications greater than a few thousand the material is seen to be composed of rather undulating wavy flakes, the 'cornflakes' texture characteristic of montmorillonite group minerals (Tovey, 1971). Here the flakes are generally 1–5  $\mu\text{m}$  across, though occasional flakes up to 25  $\mu\text{m}$  are seen.

Major and trace-element analyses of the smectite are given in Table II, nos. 1 to 4. These analyses were made by the same techniques as those of the ferromanganese oxides. With the absence of Mn from these samples, iron oxidation ratios could be measured by conventional wet-chemical means. Results are as follows (quoted as  $\text{Fe}^{3+}/\text{Fe}^{2+}$ ): no. 1, 110; no. 2, 90; no. 3, 160; no. 4, 150. In all cases, the iron is almost entirely ferric; indeed the ferrous iron contents are so low as to suggest that this amount might be produced by reduction during analysis.

The analyses show clearly that the clay mineral is essentially an alkali ferric-iron hydrated silicate. Other components are at very low levels, a trait that is shared by most of the trace elements too. Perhaps most strikingly, Al is at a very low level for clay minerals. This is probably important in discussing the genesis of this smectite.

Recalculation of the smectite analysis (Table II) shows  $(\text{Si} + \text{Al})$  is always less than the tetrahedral occupancy of 8.0 corresponding to 22 oxygens, while  $(\text{Fe}^{3+} + \text{Mg})$  is always rather greater than the dioctahedral occupancy of 4.0. However, taken together the sum of  $(\text{Si} + \text{Al} + \text{Fe}^{3+} + \text{Mg})$  is always very close to 12.0. Most probably some  $\text{Fe}^{3+}$  is substituting into the tetrahedral sites, as Weaver *et al.* (1967) demonstrated by Mössbauer spectroscopy for an Al-poor nontronite.

The sum of  $(\text{Na} + \text{K} + \text{Ca})$  is about 1.4, balanced by divalent Mg in octahedral sites and trivalent Fe and Al in tetrahedral sites in about equal amounts. Though the rest of the cell contents are very close to an Al-poor nontronite, this content of alkalis is about twice that of an ideal nontronite. A typical structural formula for the analyses of Table II might be:  $(\text{K}_{0.8}\text{Na}_{0.5}\text{Ca}_{0.1})(\text{Fe}_{8.3}^{3+}\text{Mg}_{0.7})(\text{Si}_{7.3}\text{Al}_{0.1}\text{Fe}_{0.6}^{3+})\text{O}_{20}(\text{OH})_4 \cdot 2\text{H}_2\text{O}$ . Because of the poor crystallinity of the mineral it is not clear whether it is a nontronite with rather more than usual alkalis, or whether the differences are more profound than this. Further work will be directed to finding this out.

The trace elements are almost all at very low levels. Rb is high, presumably substituting in the alkalis site, as may the Sr and Ba. The S may be present as sulphide, but the low levels of Cu and Zn and the higher content of Ba and Sr suggest that it is more likely to be present as sulphate. The low levels of such elements as Ti, Y, and Zr, as well as the very low level of the major element Al, is of some interest. If this clay had formed by the action of hot water on basalt glass, then these elements, which are all relatively immobile during secondary alteration of basalt (Cann, 1970b), would have been expected to be close to their values in fresh basalt. As it is, they are 20–30 times lower than in fresh basalt nearby (Cann, 1970a). Such a degree of depletion has not been seen in even very severely altered basalts—in fact very severe alteration tends to increase the contents of these elements because of the leaching of all others. On the other hand, if the clay was formed by direct precipitation from hydrothermal solutions, then its lack of these immobile, and hence insoluble, elements is not surprising. The poverty in Al of the source is also indicated by the presence of  $\text{Fe}^{3+}$  in the tetrahedral sites, a state of affairs that would seem, from its rareness, to be energetically unfavourable when normal amounts of Al are available.

(c) *Iron oxide.* Some of the lumps of smectite, and some of the mixed smectite–Mn-oxide (see below) contained veins and coatings of bright orange powdery Fe oxide. Very little of this material was available. A fairly pure concentrate was scraped out of one vein for X-ray diffraction, but the material was X-ray amorphous: no diffraction lines could be seen under the most

favourable circumstances. Hand picking a sample for chemical analysis was more difficult. Although the sample looked fairly clean, Table II shows that it consists of about 30 % of Mn oxide, 40 % of smectite, and only 30 % of hydrated Fe oxide (calculated by assuming all Mn as Mn oxide, residual Si as smectite, and subtracting these components to leave essentially Fe and H<sub>2</sub>O in approximate proportions Fe:H<sub>2</sub>O~1:1.5). Because of the impurity of the sample, little further can be added.

(f) *Mixed smectite—Mn oxide*. This formed small soft lumps up to 10 cm across of a light chocolate brown colour. X-ray diffraction and chemical analysis (Table II, 6) show it to be composed of a mixture of smectite and Mn oxide (about 60 % smectite, 40 % Mn oxide, to judge from the analysis). Its properties are those expected of a mixture of the two phases.

Its main interest is that it is an exception to the otherwise very efficient separation of Mn from Fe in the components of this dredge haul. Most of the Mn oxide contains no smectite detectable by X-ray diffraction, and gives Mn/Fe ratios between 5 and 50. The lumps of pure smectite show the reverse, with Mn/Fe ratios less than 0.005. Thus, for most of the material in this dredge haul, there is not only a separation of Mn and Fe into different phases, but a physical separation of Mn and Fe-rich phases from one another. Such a separation is common in hydrothermal deposits generally (e.g. Bonatti *et al.*, 1972), but it is clearly not always entirely efficient.

*Discussion*. There is a growing consensus among theoreticians (Wolery and Sleep, 1976; Lister, 1972), marine geophysicists (Williams *et al.*, 1974), and geologists working on ophiolite complexes (Spooner and Fyfe, 1973; Constantinou and Govett, 1973) that penetrative convection of sea water through newly formed hot oceanic crust at mid-ocean ridge crests does occur, and is responsible for the removal of large quantities of heat in the axial zone of the ridges, the introduction of water to form ocean-floor metamorphic rocks, the leaching of material from the rocks followed by its precipitation to form hydrothermal deposits as the hot water emerges on the ocean floor, and the removal of some components from sea water (especially Mg) and addition of others to sea water to maintain the composition of the oceans. Such a process is clearly of great potential in affecting several aspects of global geological cycling.

Direct observation of the hydrothermal products of such a process on the ocean floor has been slow in emerging. There are good reasons for this, in that the deposits probably form very close to the spreading axis, and are usually covered by subsequent lava flows (as indicated by the deposits in ophiolite complexes), and that they make up, even near the axis, a very small proportion of the ocean floor. However, direct empirical observation of ocean-floor deposits would clearly be of some interest, and investigation of the properties of material from such deposits should help to set limits to the nature of the processes occurring.

Apart from the metalliferous sediments cored from sediments immediately overlying oceanic crust on mid-ocean ridges, and at the base of sediments on older crust drilled by the Deep Sea Drilling Project, the direct observation of hydrothermal deposits seems limited to a few places. The deposits beneath the hot brine pools in the Red Sea (Degens and Ross, 1969) are the classic example. Bonatti and Joensuu (1966) reported an iron-rich deposit from the crest of the East Pacific Rise. Bonatti *et al.* (1972) described a subaerial deposit apparently formed in a shallow submarine environment at the spreading centre in Afar. Scott *et al.* (1974) dredged lumps of Mn-rich oxide from the TAG hydrothermal area 5 km from the axis of the Mid-Atlantic Ridge at 26° N. A similar deposit to the present one was discovered by submersible and sampled in the FAMOUS area at 37° N on the Mid-Atlantic Ridge (Arcyana, 1975). This deposit from the Gulf of Aden seems to be the sixth known.

Evidence for its hydrothermal nature is principally from the rate of growth of the Mn oxide. The dredge haul 6243 was made on the edge of the small median valley in the Gulf of Aden, about 3 km from the spreading axis. Given a spreading rate in this region of about 1 cm per

year (Laughton *et al.*, 1970), the maximum age of the surface flows here is about  $0.3 \times 10^6$  yr. However, because lava flows must run away from the spreading centre for a kilometre or so, this is an upper limit, and the age of the surface flows may be as low as  $0.1-0.2 \times 10^6$  yr. The morphological evidence (above) shows that the formation of the deposit post-dates these flows. Some of the fragments of Mn oxide are more than 10 cm thick, suggesting a rate of deposition of Mn oxide greater than 50–100 cm per Myr. If this is compared with the rates of a few millimetres per Myr measured on normal Mn nodules (Ku and Broecker, 1969), then it is clear that exceptional, and probably hydrothermal, conditions have existed at station 6243.

Compared with the other hydrothermal deposits, that from the Gulf of Aden shows some interesting features. The nontronite-like smectite component has only been described from the Red Sea deposits, of those found in the oceans, though similar material has been described overlying sulphide deposits in Cyprus (Searle, 1972). It appears to be present in other deposits, too, where it is usually, as on Cyprus, described as an altered tuff. However, the trace-element geochemistry of the dredged smectite from station 6243 suggests strongly an origin as a direct precipitate from a hydrothermal solution and it is likely that other smectite components in hydrothermal deposits may have the same origin.

In the Red Sea, a similar association of Mn-rich sediment with Fe-rich smectite overlies a massive sulphide deposit on the floor of the Atlantis Deep (Degens and Ross, 1969). Though no sulphide was dredged from the Gulf of Aden, it would be interesting to know whether there was any beneath the part of the deposit that was dredged. Evidence relating to this is conflicting. In the Red Sea the smectite facies is very enriched in Zn, in the form of scattered globules of ZnS (Degens and Ross, 1969), and background levels of transition elements throughout the deposit are uniformly high, suggesting that the absence of such enrichment in the Gulf of Aden deposit may preclude the existence of an underlying sulphide deposit. It might be suggested that the presence of evaporites, and thus of hypersaline brines, was necessary to mobilize sulphides. On the other hand, Spooner and Bray (1976) show from fluid inclusion studies the marine salinity of Cypriot hydrothermal fluids, and experimental studies (Hajash, 1975) demonstrate that water of marine salinity can mobilize sulphides. Possibly the exceptional character of the Red Sea deposits lies in the mobilization of metals in considerable excess over sulphur, with the corresponding production of metal-rich brines as an end product. Answers to these problems will have to await further investigation.

Finally, it is interesting to ask what light this deposit throws on the nature of the hydrothermal solutions involved. Further work is proceeding on this, but it seems possible to make some initial deductions. Both major components presumably formed from the same hydrothermal solution, precipitating as it emerged at the ocean floor. Since the Mn oxide is the only component to show morphological evidence of having lain at the water-rock interface, it seems reasonable to suppose that the Mn oxide forms a carapace to the deposit, overlying a zone of iron-rich smectite, rather as Bonatti *et al.* (1972) suggest for the deposit in Afar, except that there the iron-rich component is goethite. Thus the smectite presumably crystallized first out of the ascending solution and was followed and covered by the Mn oxide. If any sulphide was present, this was presumably precipitated at some earlier stage.

What is the reason for such a sequential precipitation? Did the ascending solution cool as it neared the ocean floor? Or did it entrain and mix with progressively larger amounts of sea water, producing a kind of natural titration? The second possibility seems most likely. The rate of conductive cooling of the ocean floor would, if the first case operated, have to exceed the upward rate of heat transfer by flowing hydrothermal water. This could only be true over a very thin boundary layer if the rates of water flow envisaged by the theoreticians occurred. Not only that, but the very oxidized character of the deposits would not be expected if they

were precipitated only by cooling. The hydrated and metamorphosed rocks of the ocean crust are generally very reduced, and experimental interaction of sea water with basalt produces reduction of sulphate in the sea water to sulphide (Hajash, 1975). The fact that the Fe in the hydrothermal deposit is mostly in the ferric form and the Mn must be also highly oxidized suggests that the control on precipitation may have been the rate of influx of oxygenated water, oxidizing the reduced forms in the hydrothermal fluid to less soluble valence states. The reason for the sequential precipitation of iron and manganese could thus be progressive oxidation, first taking Fe to the trivalent state, and then Mn to the tetravalent state.

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