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Electron microscopy investigation of the genetic link between Fe oxides/ oxyhydroxides and nontronite in submarine hydrothermal fields

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ABSTRACT

The two major components most frequently found in sediments near submarine hydrothermal fields are Fe oxides/oxyhydroxides and nontronite. Physico-chemical conditions in these environments vary from site to site and even within sites as the type and intensity of the hydrothermal activity change. The investigation of Fe oxides/oxyhydroxides and nontronite is helpful to constrain the specific conditions in which they formed or evolved, and thus to recreate the history of hydrothermal vents. Microbial activity is also involved in the formation of these mineral phases, which adds the interest of revealing processes at the interface of biological and inorganic phenomena. We investigated sediments from ten seafloor hydrothermal fields, most of them Recent sediments. They are rich in Fe oxides/oxyhydroxides and/or nontronite and/or Fe-talc. SEM analysis with energy-dispersive X-ray spectroscopy revealed many morphological types, among which vermicular habits are common, with chemical composition ranging frequently between Fe oxides/oxyhydroxides and nontronite. Smooth surfaces of the investigated particles correlates with high Fe/Si ratios, and rough surfaces with low Fe/Si ratios. This feature and the fact that many particles had chemical compositions (Si, Fe, Mn, Mg, Al) in a continuous line linking Fe oxide and nontronite are compatible with the in situ reaction of Fe oxides/oxyhydroxides as a frequent mechanism of nontronite formation. This process involves the reaction of Fe oxides/oxyhydroxides with either silica polymers adsorbed on their surface or Si-Fe amorphous phases precipitated on the same surface. However, it was not possible to establish a pseudomorphic replacement (of Fe oxides by nontronite) as no nontronite grains were found reproducing habits observed in grains with large proportion of Fe oxides/oxyhydroxides. All particle morphologies can have a range of surface roughness and composition. Chemical composition consistent with nontronite was represented mainly by small particles ($< 1 \, \mu m$) and flakes or veil-like habits. The frequent vermicular habit caused by precipitation on Fe-oxidizing bacteria was in line with previous findings indicating this as an important process of precipitating a wide range of Fe-Si phases. Overall, our data suggest that in situ replacement of Fe oxides/oxyhydroxides by nontronite takes place in all types of particles but that the replacement does not preserve the original particle morphology.

1. Introduction

Marine metalliferous sediments of hydrothermal origin commonly contain, as major components, poorly crystalline Si-Fe complexes and nontronite (Fe-rich smectite) of a wide range of crystal order. The microscopic investigation of these components indicates numerous mineral phases ranging from Fe phases such as hematite (both nanocrystalline and well crystallized), goethite, akaganeite (Schwertmann et al., 1998), lepidocrocite (Taitel-Goldman et al., 2009) and disordered Fe oxyhydroxides (Ueshima and Tazaki, 2001), to Si-Fe phases such as ferrihydrite (Schwertmann et al., 1998; Sun et al., 2013), an Si-Fe poorly crystallized mineraloid (Taitel-Goldman et al., 2009), Fe-Si oxide gels (Masuda, 1995; Ueshima and Tazaki, 2001) and nontronite (Murnane and Clague, 1983; Masuda, 1995; Ueshima and Tazaki, 2001; Severmann et al., 2004, among others). For the sake of simplicity, we refer to Fe oxides and oxyhydroxides as *FeO* + *FeOOH*. In addition, Mn

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Table 1 Description of the in	westigated samples. T	The numbers in parenth	eses in the sample labels in	dicate the dep	th (cm) of the	sample in th	le core.				
Sample	Hydrothermal system type	Hydrothermal system subtype	Locality	Latitude	Longitude	Water depth (m)	Sampling device	Sample type	Hand sample description	Reference	Provided by
Va1-52KH (5-10)	Basalt-hosted	Sediment-covered	South-West Basin, Atlantis II Deep, Red Sea	21°20′38″N	38°05′03″E	2181	Heavy kasten corer	Sediment	Red-brown silicate sulfide ooze.		A. Kraetschell, GEOMAR
Va1-52KH	- 3 -	- ,, -	2		- " -	- ** -	- " -	- 8 -	Brown-red silicate ooze.		
Va1-52KH		- 33 -	- ,, -	- " -	- " -		- " -	- 27 -	Brown silicate mud.		- " -
(4/-52) Va3-41 3KH	- " -		- 2 -	21°20.51 N	38°05.45′E	2155		- " -	Red-brown silicate mud.		
(5–10) Va3-413KH	- ,, -	- ,, -	- " -	- " -	- " -	- " -	- " -	- ** -	Red-brown mud.		- ,, -
(179–181) Va22-146KS	- " -	- " -	West Basin. Atlantis II	21°21.59'N	28°03.94'F.	2138	Super heavy	- " -	Red-brown silicate ooze.		- " -
(5–10)			Deep, Red Sea	NI 60.17 17	7 10.00 07	0017	kasten corer		INCH-DIOWII SIIICAIC 005C.		
Va22-146KS (190–195)	- ,, -	- 33 -	- , , -	- * -	- " -	- " -	- " -	- ** -	- 32 -		- ,, -
Va22-146KS	- . .	- ,, -	- 2 -		- " -	- "	- " -	- * -	Brown mud.		, -
(6/01-//01) 7D-8A	- " -	- ** -	Southern Trough of	27°02.7'N	111°22.8W	2000	Dredge	Massive sulfide	Orange-white talc.	Koski et al.	R. Koski, USGS
			Guaymas Basin, Gulf of California							(1985)	
Sea Cliff Dive 308	' 3		Northern Trough of Guaymas Basin, Gulf of California	27°18.3'N	111°30.4'W	2000	Submersible	Fragments of talc ledges	Light gray with rusty stains; light, crumbly, porous fragments.	Lonsdale et al. (1980)	P. Lonsdale, SIO, UCSD.
Cyp-78-08-14B*		Sediment-free	East Pacific Rise	20°54'N	109°01 W	2650	' 3	Sediment	Light brown mud with sub-cm consolidated fragments.	Hekinian et al. (1993)	R. Hekinian, IFREMER
Cyp-78-12-37b	- 22	- 37 -	- ₂₃ -	20°54.25'N	109°02′W	2660	- 33 -	Sediment	Orange-red sediment with small, hard, red fragments	- , , -	
Cyp-78-12-37-21	- ,, -	- 37 -	- .	20°54'N	109°01 W	- " -	- 3 -	Oxyhydroxide mud	Red sediment with	- ,, -	- ,, -
Cyp-78-12-41D	' ''	3 1	3 -	20°54.35'N	- ,, -	2620		Sediment	ychow brown runps. Multicolor fragments: red, pale brown, yellow-	- " -	' 3
Cy 84-31-1	- ,, -	- 	- 39 -	12°42.20'N	103°52.00 ^w	2502	- 33 -	Oxyhydroxide denosit	prown, gray-plack. Pale brown sediment.	- 33 -	- ,, -
Cy 82-14-5	- " -	- ,, -	Southeastern Seamount, East Davific Dies	12°42.09'N	103°52.24'W	2550	- » -	Hydrothermal	Brown to brown-black	- " -	- 23 -
1183-9	י ז י	, , ,	East Pacific Rise	20°48'N	109°22°W	1700	, ,	Solid fragments in the sediment	Orange-brown to black, light chunks: talc (+FeOOH).	Alt et al. (1987)	J. Alt, University of Michigan
1183-15		- ** -	- ,, -	- 3	3 -	*	- 3 -		Dark-green to light-green fragments: nontronite (+MnOOH).	- 	- 3 -
Cy 988-TH05-02	- 33 -	- * -	Teahitia volcano	17°33.20′S	148°49.50′W	1526		Hyaloclastite	Purple red sediment.	Hekinian et al. (1993)	R. Hekinian, IFREMER
St. 631 6	Peridotite-hosted	- 3 -	Logatchev-2, Mid- Atlantic Ridge	14°43.21 N	44°56.27'W	2710	TV-grab	Massive sulfide talus	White to orange-white colloform masses.	Dekov et al. (2011)	M. Davydov, VNIIOkean- geologia

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