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The origin of hydrothermal chlorite- and anhydrite-rich sediments in the middle Okinawa Trough, East China Sea

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ABSTRACT

During the Integrated Ocean Drilling Program (IODP) Expedition 331, five sites were drilled into the Iheya North Knoll hydrothermal system in the Okinawa Trough (OT) — a back-arc basin characterized by thick terrigenous sediment. Following up on the previous study by Shao et al. (2015), we present new mineralogical, geochemical, and Sr-Nd isotope data to investigate the origin of the hydrothermal sediments and characterize the hydrothermal system. The substrate at the Iheya North Knoll is dominated by pumiceous sediment and other volcanoclastic materials interbedded with hemipelagic (terrigenous and biogenous) sediments. Impermeable layers separate the hydrothermal sediments into distinct units with depth that are characterized by various assemblages of alteration materials, including polymetallic sulfides, sulfates, chlorite- and kaolinite-rich sediments. The rare earth elements (REEs) and Nd isotope data suggest that the chlorite-rich and kaolinite-rich layers primarily resulted from the alteration of pumiceous materials in different chemical and physical conditions. Kaolinite-rich sediment likely reflects low pH and low Mg concentration fluids, while chlorite-rich sediment formed from fluids with high pH and increased Mg contents, likely at higher temperatures. The Sr isotopic compositions of subsurface anhydrite reflect high seawater/hydrothermal fluid ratios in the mid-OT hydrothermal area. Compared with chlorite-rich sediments from other sediment-covered or felsic-hosted hydrothermal systems, the chlorite-rich sediments in the mid-OT are characterized by lower concentrations of Al and Fe but much higher Y, Zr, Hf, Th and REEs, indicative of the distinct nature of the precursor rocks in this region.

1. Introduction

Hydrothermal systems primarily occur in three different spreading environments: mid-ocean ridges, intraoceanic back-arc rifts in ocean crust (such as the Manus Basin, N. Fiji Basin and Lau Basin), and intracontinental rifts in continental crust (such as the Okinawa Trough, East China Sea) (Herzig and Hannington, 1995; Herzig and Hannington, 2000; Glasby and Notsu, 2003). The hydrothermal fields in the Okinawa Trough (OT) are characterized by a thick covering of terrigenous sediments that play an important role in determining the nature of hydrothermal deposits (Lee et al., 1980; Letouzey and Kimura, 1986; Kawagucci et al., 2011). In addition, volcanic materials like pumice are widely distributed in the OT and also affect the geochemistry of hydrothermal alteration reactions (Shinjo, 1999; Shinjo et al., 1999; Shinjo and Kato, 2000; Expedition 331 Scientists, 2011).

Hydrothermal alteration of rocks and sediments results in the formation of secondary mineral assemblages of kaolinite, illite, smectite, montmorillonite, chlorite and other mixed layer clays (Alt, 1999; Marumo and Hattori, 1999; Lackschewitz et al., 2000b; Lackschewitz et al., 2004; Dekov et al., 2005; Miyoshi et al., 2013; Beaufort et al., 2015). The Iheya North Knoll hydrothermal field in the mid-OT is characterized by high concentrations of chlorite, reaching up to 70% at Integrated Ocean Drilling Program (IODP) Site C0013 (Expedition 331 Scientists, 2011). Shao et al. (2015) compared the mineralogy and geochemistry of the clay-size sediments in the mid-OT recovered during the IODP Expedition 331, and discriminated the clay origins between hydrothermal and terrigenous sources. However, whether the chlorite-rich sediments result from hydrothermal alteration of terrigenous

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sediments and/or volcanic materials remains to be clarified.

Anhydrite, which exhibits retrograde solubility, is the most important sulfate phase at the mid-OT hydrothermal area (Expedition 331 Scientists, 2011). The precipitation of anhydrite provides the opportunity to investigate the extent of seawater entrainment into the subsurface (Mills and Elderfield, 1995; Humphris, 1998; Teagle et al., 1998a; Teagle et al., 1998b). However, little is known about subsurface circulation within the mid-OT hydrothermal area, where substantial amounts of anhydrite were found during IODP 331 Expedition.

To better understand the hydrothermal environment and deposits in the mid-OT area, we have analyzed the bulk mineralogical, elemental and Sr-Nd isotopic compositions of selected samples from several sediment cores collected during IODP Expedition 331. The main objectives of this study are to: 1) determine the nature of the material hydrothermally altered to produce the chlorite-rich sediment in the mid-OT; 2) use the behavior of Sr and the REEs in anhydrite to establish the role of seawater entrainment and subsurface mixing in the hydrothermal system; 3) develop a schematic model of the hydrothermal system at Iheya North Knoll based on the stratigraphy, mineralogy, and geochemistry.

2. Geological setting and sample selection

The OT is a back arc basin (BAB) extending for about 1200 km between Kyushu Island and Taiwan. The north OT (NOT) is about 230 km wide with a maximum water depth of 200 m, and the south OT (SOT) is about 60–100 km wide with a maximum water depth of 2300 m (Letouzey and Kimura, 1986). The OT has been undergoing rifting since ~2 Ma, and was preceded by an earlier rifting episode during the Miocene (Lee et al., 1980). Seismic reflection data suggest that the mantle (at ~6000 mbsf (meter below seafloor)) is overlain by potentially young basalt (~3000–6000 mbsf), igneous rocks (~1000–3000 mbsf), and sediments up to 1000 m thick (Expedition 331 Scientists, 2011). In 1988, Halbach et al. (1989) reported the occurrence of hydrothermal activity in the mid-OT. Thus far, at least six active hydrothermal fields have been identified: Minani-Ensei Knoll, Iheya North, Iheya Ridge, Izena Hole, Hatma Knoll, and Yonaguni Knoll IV (Takai et al., 2012).

The Iheya North Knoll (27°47′50″N, 126°53′80″E) is located approximately 150 km NNW of Okinawa Island (Fig. 1). Previous seismic and gravity core studies in the central valley of this hydrothermal field (apparent seafloor area as 2000 m × 2000 m), suggested the presence of thick pumice layers with coarse to fine grain sizes, which often contain abundant gas-filled voids accompanied by elemental sulfur and sulfide minerals, probably deposited by gas-rich hydrothermal fluids (Oiwane et al., 2008; Expedition 331 Scientists, 2011; Masaki et al., 2011).

The samples selected for this study include sediments recovered from IODP 331 Sites C0013, C0014, C0015 C0016, C0017, as well as surface sediments and rocks collected in the north OT (NOT) during East China Sea (ECS) Investigation Expedition 2012 (Table 1). More detailed information about the cores is included in the IODP 331 report (Expedition 331 Scientists, 2011). A substantial amount of hydrothermally-formed secondary clay minerals, including chlorite, kaolinite-muscovite (intergrowth) and illite/smectite were recovered during IODP 331 Expedition (Expedition 331 Scientists, 2011; Miyoshi et al., 2015; Shao et al., 2015).

IODP Site C0016 is located at the base of the active (> 310 °C) hydrothermal vent site and sulfide–sulfate mound at the North Big Chimney (NBC). Three cores were collected in Hole C0016B (no recovery in C0016A) over an interval of 0 to 45 mbsf, with a recovery of only 2.1 m (4.7%) of core. Core recovered from the 0–9 mbsf interval consists of massive and semi-massive sulfide underlain by silicified volcanic rock. The second core consists of only three rocks: two pieces of silicified volcaniclastic breccia and one piece of coarse anhydrite. The third core (27–45 mbsf) recovered ~1 m of quartz-chlorite-rich altered volcanic rock with abundant stock-work veining (Expedition 331 Scientists, 2011; Takai et al., 2012). For this study, we selected two samples from the first interval and one sample of white, coarsely crystalline anhydrite from the second interval (Table 1).

At IODP Site C0013, ~100 m east of NBC, four lithologic units were identified (Fig. 2). Unit I (0–4 mbsf): hydrothermally-altered mud containing crystalline pipes of elemental sulfur and sulfide grit; Unit II (4–14 mbsf): hydrothermally-altered mud with some heavily veined intervals and clastic units containing anhydrite breccia and fragments of metalliferous massive sulfide; Unit III (14–26 mbsf): hydrothermally-altered mud with abundant nodular anhydrite occurring in some layers; and Unit IV (26–55 mbsf): volcanic breccia with clasts of various volcanic rocks (Expedition 331 Scientists, 2011) (Fig. 2). Based on XRD data, the hydrothermally-altered mud is chlorite rich, up to 70% by volume (Miyoshi et al., 2015; Shao et al., 2015). For this study, twenty-four samples of the hydrothermally altered mud were selected between 0 and 26 mbsf.

IODP Site C0014 is located ~450 m east of NBC and is a region of relatively low surficial heat flow compared with Site C0013 (Fig. 2). The drilled sequence at this site represents deposition of hemipelagic and volcanoclastic materials that have been subjected to hydrothermal alteration to different extents. The upper part (Unit I: 0 to < 18 mbsf) shows little evidence of hydrothermal alteration and consists of a succession of hemipelagic oozes and coarse pumice gravel. The degree of hydrothermal alteration increases downward, showing partially consolidated hydrothermally-altered mud and coarse angular pumice gravel in Unit II (~12 to ~30 mbsf), and consolidated and often well-cemented volcanic sediments interbedded with hydrothermally-altered mud from ~30 to ~128 mbsf (Unit III). This site has much lower abundance of anhydrite than Site C0013, implying less direct seawater input into the system at this site or lower temperature (< 150 °C) (Humphris, 1998). The lower part of Unit III exhibits lower contents of chlorite but higher amounts of quartz and muscovite. In this study, we consider this part as Unit IV — a separate unit from Unit III, although they are classified as one unit in the IODP report (Expedition 331 Scientists, 2011). We analyzed 8 samples from Unit I, 8 from Unit II, 8 from Unit III, and 7 from Unit IV.

IODP Site C0017, located ~1550 m east of NBC, is inferred to be the recharge zone because of its low heat flow and low surficial thermal gradient (Fig.1; Expedition 331 Scientists, 2011). It mainly contains units of homogeneous hemipelagic mud, volcanoclastic–pumiceous breccia and mixed sand. No significant hydrothermal alteration was observed at this site, with the exception of weak alteration to clay within the deepest part (~150 mbsf) (Shao et al., 2015). The upper Unit I (0–28 mbsf) contains abundant foraminifera and coccoliths, suggesting a normal hemipelagic depositional environment (Expedition 331 Scientists, 2011; Shao et al., 2015). Pumiceous gravels dominate C0017-Unit II with interbedded detrital sediments, which have similar mineralogical characteristics to pumiceous sediments encountered at Site C0015. Because of the similarity in sediment types, we combined Unit II and Unit III as described in the IODP report (Expedition 331 Scientists, 2011; Shao et al., 2015) into C0017-Unit II. The C0017-Unit III is dominated by hemipelagic mud, which corresponds to the Unit IV of IODP report (Expedition 331 Scientists, 2011; Shao et al., 2015).

IODP Site C0015 is located on a hill ~600 m northwest of NBC. Drilling reached only 9.4 mbsf and only two cores were recovered. They contain coarse pumiceous gravel and grit, siliciclastic sand, hemipelagic mud, bioclastic gravel, and foraminifera sediments. Pelagic sedimentation dominates at this site, but redeposited volcanic clasts comprise the bulk of the sediment (Expedition 331 Scientists, 2011). The C0015-Unit I mainly consists of hemipelagic mud, bioclastic and pumiceous sediment, while pumice gravels predominate C0015-Unit II. Overall, this site lacks obvious hydrothermal alteration.

The northern Okinawa Trough (NOT) has very thick sediments (up to 8 km thick) due to the supply of terrigenous material from the