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Research paper

Smectite, illite, and early diagenesis in South Pacific Gyre subseafloor sediment

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ABSTRACT

Subseafloor sediment and basalt rock samples at seven sites in the South Pacific Gyre (SPG) were recovered by Integrated Ocean Drilling Program Expedition 329 (2010.10.8–2010.12.13). Microscopic and spectroscopic measurements on the structural Fe-redox states and the elemental composition of smectite, and polytypes of illite in the sediment at two sites (U1365 and U1369) were performed to understand the origin/formation mechanism of clay minerals in the oligotrophic open ocean. The dominant phases of clay minerals found in the present study were smectite and illite polytypes. Suggestive of non-uniform early diagenetic processes in the expansive SPG seafloor, higher ordering of illite polytypes (1M, 2M₁, and 3T) were identified at site U1369 while disordered 1M_d illite were found at U1365. Smectites of hydrothermal origin (Al-rich beidellite, and saponite) were observed at U1369. Fe-rich montmorillonite minerals that are likely associated with the terrigenous input, were dominant at U1365. Nontronite (Fe-rich smectite) was detected at both sites. Red-brown to yellow-brown semiopaque oxide minerals (RSO) were widely distributed with Fe-rich smectite near the basaltic crust at U1365. Lower observed heat flow at U1365 relative to U1369 provides a possible explanation for the observed variability in clay mineral speciation between these two sites. The presence of K-nontronite at the basalt/sediment interface at both sites indicates an oxidative basalt alteration; however variations in the oxidation states of structural Fe in nontronite measured by EELS indicate that reductive environment persists locally at the basalt/sediment interface.

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1. Introduction

Integrated Ocean Drilling Program (IODP) Expedition 329 recovered subseafloor sediment and basalt rock samples across a range of crustal ages and tectonic spreading rates in the South Pacific Gyre (SPG). The SPG is a unique depositional environment where the sedimentation rates are among the lowest (<1.6 to 1.9 m/Myr) of any seafloor setting on Earth, and sediments and basaltic basement have been continuously exposed to oxic conditions for their entire sedimentation history (D'Hondt et al., 2009, 2015). The low sedimentation rates are due to extremely low influxes of terrigenous matters and low bioproduction in the euphotic zone (Anderson, 1986; Glasby, 1991; Claustre and Maritorena, 2003; D'Hondt et al., 2009). Given these unique conditions, the formation of subseafloor sediments, particularly the composition

and structure of clay minerals and their formation mechanism supposed to be different from those in the anoxic conditions of equatorial upwelling regions and high accumulation rates at the continental margins (D'Hondt et al., 2015). IODP Expedition 329, for the first time, allows investigation of a vast range of mineralogical data throughout the entire sedimentary column of the SPG.

Smectites in subseafloor sediment can originate from aerial or fluvial influx of terrigenous material, or hydrothermal alteration of basalt (Biscaye, 1965; Chamley, 1989; Weaver, 1989; Fagel et al., 2001). Variations of chemistry and crystalline structure within smectites reflect their origin and formation mechanisms (Wilson, 1999). Trioctahedral smectite, mainly occurring as the mineral saponite, likely forms through the hydrothermal alteration of basaltic basement igneous rocks (Zierenberg and Shanks, 1983; Cole, 1988; Cuadros et al., 2008, 2013; Dekov et al., 2008). Dioctahedral Al-rich smectite (beidellite) appears to form via interactions among hydrothermal solutions and sediment at high temperatures (~160 to 200 °C) (Haggerty and Baker, 1967; McMurtry et al., 1983; Parra et al., 1985, 1986; Fagel et al., 2001). Fe-rich smectite, known as the

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mineral nontronite, is quite frequent in hydrothermally altered basalt (Zierenberg and Shanks, 1983; Cuadros et al., 2008, 2013; Dekov et al., 2008), although Pacific pelagic clays also possess nontronite that possibly forms through low temperature reactions within seafloor sediment (Cuadros et al., 2011).

Fe-rich smectite, particularly nontronite has variable crystal structure and chemistry. Variation of Fe(II)/Fe(III) in smectite structure is likely attributable to redox changes that may involve oxidation of organic matter (Eslinger et al., 1979), or local microbial Fe-respiration (Gates et al., 1998; Kostka et al., 1996, 1999a, 1999b; Stucki, 1998; Kim et al., 2003, 2004; Zhang et al., 2007a, 2007b; Jaisi et al., 2005, 2007a, 2007b, 2011; Seabaugh et al., 2006; Stucki and Kostka, 2006; Dong et al., 2009; Kim and Dong, 2011). In contrast, the formation of K-smectite indicates an oxidative weathering process of seafloor sediment in the presence of seawater (Hart et al., 1974; Clayton and Pearce, 2000; Zhou et al., 2001). Variations in mica polytypes associated with endogenous heat flow (Nespolo, 2001) are common in sediments. In seafloor hydrothermal deposits, micas develop the stacking order with a sequence of $1M_2$ – $1M$ – $2M_1$ depending on the hydrothermal activity and reaction time (Yoder, 1959; Velde, 1965). The structural assessments by Selected Area Electron Diffraction (SAED) pattern allows to detect the evolution of mica polytypes in hydrothermal regions (Inoue et al., 1988; DiMarco et al., 1989). Pelagic red clays in the Pacific Ocean constitute 49% of seafloor sediment (Anderson, 1986). They are especially abundant in the southwestern Pacific Basin (Piper et al., 1985). Fe-rich smectite and Fe/Mn-oxyhydroxides are the dominant mineral phases in Pacific pelagic clays (Singer et al., 1984; Cuadros et al., 2011).

This study aims to investigate the structure and mineral composition of smectite phases and illite polytypes in the seafloor sedimentary sequence in the oligotrophic open ocean of the SPG. Specific objectives of this research include (1) investigating the diversity of smectite phases, (2) determining the redox states of structural Fe in Fe-rich smectite, and (3) defining the elemental composition and formation mechanisms of clay minerals and polytypes of illite in the sediment of two oligotrophic sites (IODP Expedition 329 sites U1365 and U1369) that experienced long-term, low rates of sediment accumulation and

persistent oxic conditions. Therefore, the present study provides insight into the processes of mineral formation and early diagenesis.

2. Samples and methods

2.1. Sampling locations

IODP Expedition 329 recovered seafloor sediment from seven sampling sites along two transects spanning the western half of the SPG. One transect (sites U1365–U1368) crosses the center of the SPG at 26°S from the upper slope of the East Pacific Rise (EPR) to the western abyssal plain with increasing basement age from 13.5 Ma to 84–120 Ma (Gradstein et al., 2004) (Fig. 1, Table 1). The entire sediment column at four sites and the upper 100 m of basement at three sites were cored along this transect. Water depths at these sites varied from 3740 meters below sea level (mbsl) to 5695 mbsl (Table 1). The other transect (sites U1369–U1371) crosses the southern edge of the gyre at approximately 42°S and also sampled the entire sediment column at each site. Crustal ages is 75 Ma and water depths increase from 5075 m to 5305 m. Sediment cores were recovered at three sites where basement rocks were deliberately not cored. The thickness of the sediment column in the SPG seafloor increases with the basement age and is greatest at site U1371 (131 m), a location that is influenced by upwelling along the southwestern edge of the SPG (D'Hondt et al., 2009). Expedition 329 shipboard scientists characterized the overall stratigraphy and preliminary mineralogy for all recovered cores (Expedition 329 Scientists, 2011). Based on those results and to more fully characterize differences among the sites with respect to crustal age and position within the SPG, sites U1365 and U1369 were selected for further study. Sites U1367 and U1368 were excluded from further consideration because the dominant lithology at those sites is calcareous nannofossil ooze; site U1371 was excluded because its cores are composed primarily of siliceous ooze. Site U1365 is located 4500 km west of the EPR. It represents the location furthest from the ridge and resides on the oldest crust available within the SPG, dated at 124.6 Ma (Gradstein et al., 2004). Sediment recovered at site U1369 resides upon crust dated at 58 Ma, approximately 2000 km west of the EPR.

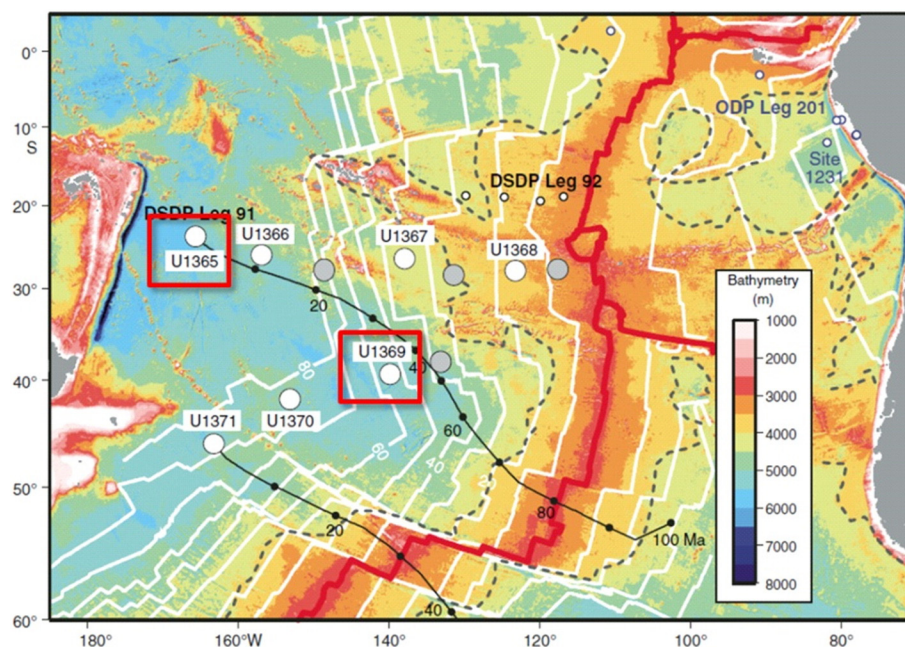


Fig. 1. Bathymetry map of the SPG showing drilling sites by IODP Expedition 329 (U1365–U1371). Sediments cored at sites U1365 and U1369 (outlined in red) were used for the present study. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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