

# Sulphide mineral evolution and metal mobility during alteration of the oceanic crust: Insights from ODP Hole 1256D

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## Abstract

Fluxes of metals during the hydrothermal alteration of the oceanic crust have far reaching effects including buffering of the compositions of the ocean and lithosphere, supporting microbial life and the formation of sulphide ore deposits. The mechanisms responsible for metal mobilisation during the evolution of the oceanic crust are complex and are neither fully constrained nor quantified. Investigations into the mineral reactions that release metals, such as sulphide leaching, would generate better understanding of the controls on metal mobility in the oceanic crust. We investigate the sulphide and oxide mineral paragenesis and the extent to which these minerals control the metal budget in samples from Ocean Drilling Program (ODP) Hole 1256D. The ODP Hole 1256D drill core provides a unique sample suite representative of a complete section of a fast-spreading oceanic crust from the volcanic section down to the plutonic complex. The sulphide population at Hole 1256D is divided into five groups based on mineralogical assemblage, lithological location and texture: the magmatic, metasomatised, high temperature hydrothermal, low temperature and patchy sulphides. The initiation of hydrothermal alteration by downward flow of moderate temperature (250–350 °C) hydrothermal fluids under oxidising conditions leads to metasomatism of the magmatic sulphides in the sheeted dyke and plutonic complexes. Subsequent increase in the degree of hydrothermal alteration at temperatures >350 °C under reducing conditions then leads to the leaching of the metasomatised sulphides by rising hydrothermal fluids. Mass balance calculations show that the mobility of Cu, Se and Au occurs through sulphide leaching during high temperature hydrothermal alteration and that the mobility of Zn, As, Sb and Pb is controlled by silicate rather than sulphide alteration. Sulphide leaching is not complete at Hole 1256D and more advanced alteration would mobilise greater masses of metals. Alteration of oxide minerals does not release significant quantities of metal into the hydrothermal fluid at Hole 1256D. Mixing of rising high temperature fluids with low temperature fluids, either in the upper sheeted dyke section or in the transitional zone, triggers local high temperature hydrothermal sulphide precipitation and trapping of Co, Ni, Cu, Zn, As, Ag, Sb, Se, Te, Au, Hg and Pb. In the volcanic section, low temperature fluid circulation (<150 °C) leads to low temperature sulphide precipitation in the form of pyrite fronts that have high As concentrations due to uptake from the circulating fluids. Deep late low temperature circulation in the sheeted dyke and the plutonic complexes results in local precipitation of patchy sulphides and local metal remobilisation. Control of sulphides over Au, Se and Cu throughout fast-spreading mid-oceanic crust history implies that the generation of hydrothermal fluids enriched in these metals, which can eventually form VMS deposits, is strongly controlled by sulphide leaching.

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## 1. INTRODUCTION

The cycling of metals in the oceanic crust is an important process which buffers the composition of the lithosphere and oceans, contributes to development of microbial life, and leads to the formation of hydrothermal ore deposits such as volcanogenic massive sulphide (VMS) deposits. Investigations from modern-day oceanic crust (Nesbitt et al., 1987; Alt et al., 1989, 1996, 2010; Alt, 1995; Gillis et al., 2001; Bach et al., 2003; Teagle et al., 2006; Heft et al., 2008; Coogan and Dosso, 2012; Patten et al., 2015) and ophiolites (e.g. Richardson et al., 1987; Schiffman et al., 1987; Jowitt et al., 2012) suggest that mobilisation of metals from the lower sheeted dyke complex during on-axis high temperature hydrothermal alteration of the oceanic crust is a systematic phenomenon. The high temperature (>350 °C) hydrothermal fluids that leach metals from the lower sheeted dyke section rise buoyantly through upflow zones towards the seafloor where venting occurs (Alt et al., 1989, 2010; Hannington, 2014). During this ascent metals can be lost to mineralised horizons within the upper oceanic crust (Honnorez et al., 1985; Alt et al., 1998, 2010; Bach et al., 2003; Hannington, 2013), trapped within VMS deposits (e.g. Fouquet et al., 1988; Herzig and Hannington, 1995; Hannington et al., 1998; Wohlgemuth-Ueberwasser et al., 2015) or vented in hydrothermal plumes and associated sediments (e.g. Feely et al., 1994a,b; Hannington, 2013). Mobilisation of metals in the lower sheeted dykes occurs at high temperature and under reduced conditions (>350 °C; e.g. Alt et al., 2010) when solubility of metals is optimal (Seewald and Seyfried, 1990). It is generally assumed that this mobilisation is partly achieved through leaching of magmatic sulphides by hydrothermal fluids (e.g. Keays, 1987; Alt, 1995; Jowitt et al., 2012). The extent, however, to which this mineral reaction controls the release of base metals and other trace metals such as Au, As, Sb, Se and Te into the hydrothermal fluids is neither constrained nor quantified. Reactions involving oxide and silicate minerals have also been shown to mobilise metals including Zn (Doe, 1994; Jowitt et al., 2012). The mineral reactions that produce metal-rich fluids in the modern-day oceanic crust are not well constrained and systematic investigation of the sulphide population evolution during hydrothermal alteration would greatly improve our understanding of metal mobilisation in the oceanic crust.

Located in the Cocos Plate, Pacific Ocean, ODP (Ocean Drilling Program) Hole 1256D is an ideal location to investigate the mobility of metals in a fast-spreading mid-oceanic ridge (MOR) crust. The 1256D drill core recovers a complete section of the oceanic crust down to the plutonic complex (Wilson et al., 2003; Teagle et al., 2006; Wilson et al., 2006). The hydrothermal system in the crust at Hole 1256D and the resulting alteration are well defined (Teagle et al., 2006; Alt et al., 2010; Alt and Shanks, 2011; Violay et al., 2012), providing a solid framework for investigation of mineral reactions leading to metal mobility. Recent investigations on Hole 1256D have shown depletions in whole rock of Cu, Zn and Pb and also of Au, As, and Se concentrations in the deeper parts of the core (Alt et al., 2010;

Patten et al., 2015). These metals are depleted at different crustal levels ranging from the upper sheeted dykes down to the plutonic section implying that multiple mineral reactions may control metal mobility.

This investigation aims to determine the paragenesis and metal contents of the sulphide and oxide minerals in the ODP Hole 1256D crust in order to identify the mineral reactions that release metals into the hydrothermal fluids. We employ in situ analytical methods including electron-probe microanalysis (EPMA) and laser ablation-inductively coupled plasma mass spectrometer (LA-ICP-MS) analysis to enable quantification of major, minor and trace elements in sulphide and oxide minerals present in the Hole 1256D drill core. We compare our data with published whole-rock concentrations for the same samples from Hole 1256D (Patten et al., 2015) in order to identify the host minerals for this suite of metals and the mineral reactions that liberate metals into hydrothermal fluids during alteration.

## 2. GEOLOGICAL SETTING

### 2.1. ODP Hole 1256D lithologic units

The oceanic crust at drilling site Hole 1256D is a 15 Myr old crust generated from a superfast spreading ridge (~200 mm/yr) and is located in the Cocos Plate (6.736°N, 91.934°W; Wilson et al., 2003). The oceanic crust can be divided into four main lithological units: the volcanic section, the transitional zone, the sheeted dyke complex and the plutonic complex (Fig. 1). The volcanic section is capped by a lava pond on the first 100 m that was formed during off-axis volcanism events (Wilson et al., 2003) underlined by phyrlic to aphyric sheeted flows, aphyric massive units, pillow lavas and hyaloclastites (Wilson et al., 2003; Teagle et al., 2006). The transitional zone is mainly composed of aphyric sheeted flows with presence of breccias characterised by angular aphyric cryptocrystalline basaltic clasts cemented by chalcedony, saponite, carbonate, albite, anhydrite and sulphides (Teagle et al., 2006). The sheeted dyke complex comprises massive aphyric basalts with common sub-vertical intrusive contacts (Teagle et al., 2006). Due to intrusion of underlying gabbros, high-temperature recrystallisation occurs in the lower part of the sheeted dyke complex giving a granoblastic texture (Teagle et al., 2006). The plutonic complex is characterised by two gabbro intrusive bodies (Teagle et al., 2006; Koepke et al., 2008; Wilson et al., 2006). The contacts between the gabbro bodies and the dykes are intrusive, with dyke fragments occurring in the margin of the lower gabbro body (Alt et al., 2010).

### 2.2. Hydrothermal system and metal mobility

The oceanic hydrothermal system at Hole 1256D can be divided into two simplified domains: an upper low temperature domain and a deeper high temperature domain (Teagle et al., 2006; Alt et al., 2010). The low temperature domain occurs mostly in the volcanic section (250–1004 mbsf) and is characterised by alteration formed from circulation of low-temperature (50–185 °C) oxidised

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