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ORIGINAL ARTICLE

The formation of porphyry copper deposits

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Abstract Copper is a moderately incompatible chalcophile element. Its behavior is strongly controlled by sulfides. The speciation of sulfur is controlled by oxygen fugacity. Therefore, porphyry Cu deposits are usually oxidized (with oxygen fugacities $> \Delta$FMQ $+2$) (Mungall 2002; Sun et al. 2015). The problem is that while most of the magmas at convergent margins are highly oxidized, porphyry Cu deposits are very rare, suggesting that high oxygen fugacity alone is not sufficient. Partial melting of mantle peridotite even at very high oxygen fugacities forms arc magmas with initial Cu contents too low to form porphyry Cu deposits directly (Lee et al. 2012; Wilkinson 2013). Here we show that partial melting of subducted young oceanic slabs at high oxygen fugacity ($>\Delta$FMQ $+2$) may form magmas with initial Cu contents up to >500 ppm, favorable for porphyry mineralization. Pre-enrichment of Cu through sulfide saturation and accumulation is not necessarily beneficial to porphyry Cu mineralization. In contrast, re-melting of porphyritic hydrothermal sulfide associated with iron oxides may have major contributions to porphyry deposits. Thick overriding continental crust reduces the "leakage" of hydrothermal fluids, thereby promoting porphyry mineralization. Nevertheless, it is also more difficult for ore forming fluids to penetrate the thick continental crust to reach the depths of 2–4 km where porphyry deposits form.

Keywords Porphyry Cu deposit · Oxygen fugacity · Slab melting · Sulfide · Sulfate · Solubility

1 Introduction

Most porphyry copper (Cu) deposits are distributed along convergent margins (Fig. 1). A porphyry deposit is a semi-closed system that usually formed at depths of 2–4 km (Sillitoe 2010), but the controlling factors are still a topic of contention amongst scholars (Chiaradia 2014; Chiaradia et al. 2012; Cooke et al. 2005; Lee et al. 2012, 2014; Richards 2013; Sun et al. 2013; Wilkinson 2013). Thick overriding continental crust (Chiaradia 2014; Lee 2014), pre-enrichment of Cu through sulfide saturation (Lee et al. 2012; Wilkinson 2013), partitioning of Cu into fluids from high Sr/Y andesite (Chiaradia et al. 2012), addition of water (Richards 2011), accidental emergence of multiple favorable conditions (Richards 2013), subduction of young oceanic crust, in particular young ridges, flat-slab subduction, crustal thickening (Cooke et al. 2005), or high oxygen fugacities plus adakite (Mungall 2002; Sun et al. 2011, 2013) have all been proposed as potential factors.

In principle, the behavior of Cu is controlled by sulfide, whereas the speciation and behavior of sulfur is controlled by oxygen fugacity. This potentially explains the close association between porphyry Cu deposits and oxidized

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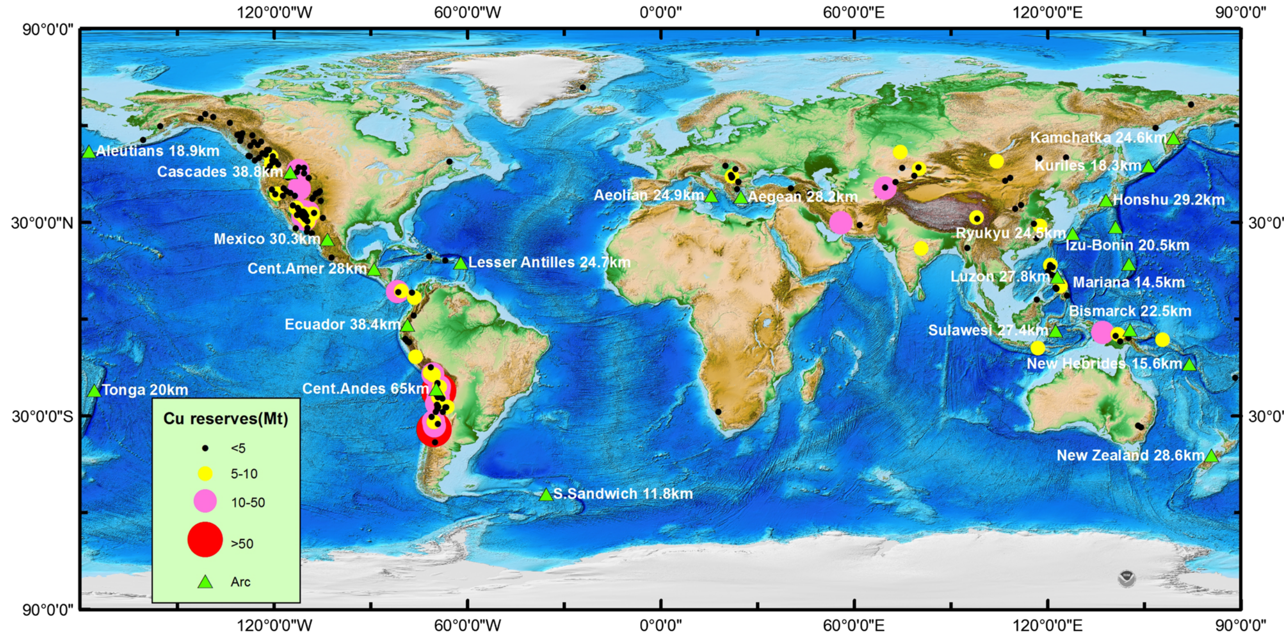

Fig. 1 The distribution of porphyry deposits (Data source USGS) (Sun et al. 2015) and the thickness of major arcs (Chiaradia 2014). Overall, thicker arc crust favors porphyry Cu mineralization, but is not the controlling factor. The west coast of the South America is the favorite place for porphyry Cu deposit, with a thickness of up to ~65 km and a total Cu reserve of over 490 million tons. This is followed by the west coast of North America, with a thickness of ~38 km and a total Cu reserve of ~380 million tons. Both are associated with subduction of young oceanic crust that are subjected to slab melting. Nevertheless, there are many fewer porphyry Cu deposits in the Tibetan Plateau, which is thicker than the Andes. There are essentially no porphyry deposits along the northwestern Pacific margins. The distribution of porphyry Cu deposits along the Pacific margins may be plausibly explained by ages of subducting oceanic plates (Sun et al. 2012, 2013). High oxygen fugacity (ΔFMQ +2–+4) and partial melting of subducted young oceanic slabs are the two key factors that controls porphyry Cu deposits. Subduction of young oceanic ridges satisfies both criteria and thus is the most favorable geologic process for porphyry Cu deposits (Sun et al. 2015)

magmas (Sun et al. 2013, 2015). It is however, still not clear why porphyry deposits favors oxygen fugacities >ΔFMQ +2, but not lower (Mungall 2002; Sun et al. 2015). More importantly, more than half of the magmas at convergent margins are highly oxidized (>ΔFMQ +2) (Bryant et al. 2007; Sun et al. 2012). Nevertheless, porphyry Cu deposits are very rare, which suggests that high oxygen fugacity is not the only controlling factor. Consistently, previous studies have suggested that partial melting of mantle peridotite even at very high oxygen fugacities forms arc magmas with initial Cu contents too low to form porphyry Cu deposits directly (Lee et al. 2012; Wilkinson 2013).

Here we show that partial melting of subducted young oceanic slabs at high oxygen fugacity (>ΔFMQ +2) may form magmas with high initial Cu. High oxygen fugacities + adakite formed by slab melting are the two essential factors that control porphyry mineralization. Pre-enrichment through sulfide saturation and accumulation is not always beneficial to porphyry mineralization, but hydrothermal processes that precipitate sulfides and iron oxides at high oxygen fugacity are favorable.

2 Modeling on partial meltings under different levels of oxygen fugacity

Porphyry Cu deposits are commonly associated with adakites (Mungall 2002; Oyarzun et al. 2001; Sajona and Maury 1998; Thieblemont et al. 1997) that formed through partial melting of subducted oceanic slabs (Sun et al. 2011, 2012). Mid-ocean ridge basalt (MORB) has Cu contents of ~100 ppm (Sun et al. 2003), which is more than three times higher than those of the primitive mantle and the continental crust (McDonough and Sun 1995; Rudnick and Gao 2003). Our modeling shows that partial melting (~10%–20%) of subducted oceanic slab forms adakitic melts with high initial Cu contents. The peak Cu contents increase with increasing oxygen fugacity, and the highest peak Cu content is reached at ΔFMQ +2 (Fig. 2).

Oxygen fugacity of ΔFMQ +2 is a turning point of sulfur (Jugo 2009; Jugo et al. 2005, 2010). At oxygen fugacities >ΔFMQ +2, sulfur is presented as sulfate, which is ~10 times more soluble in magmas. This will eliminate residual sulfur at about 10% partial melting for a slab with 1000 ppm of sulfur. Moreover, Cu is far less compatible in sulfate than in sulfide. Therefore, even if

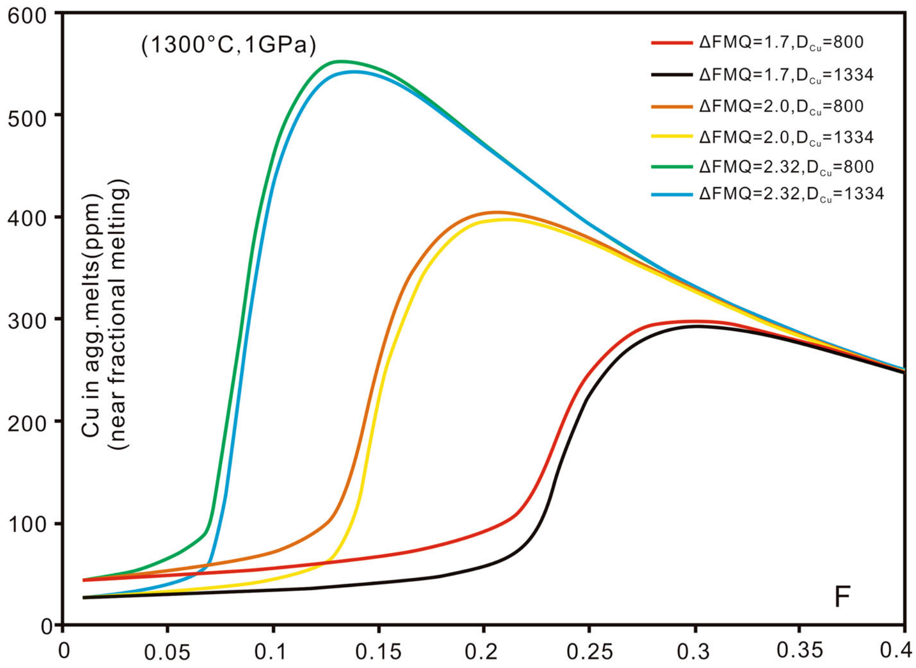

Fig. 2 Modeling results showing that partial melting of subducted oceanic slab under different oxygen fugacities and different Cu partition coefficients between melt and sulfides (1334 and 800) (Lee et al. 2012; Patten et al. 2013). Slab melting may form magmas with Cu contents >400 ppm at >ΔFMQ +2 and <20% of partial melting. The peak Cu contents are controlled by residual sulfides, not by variations of the partition coefficients of Cu between melt and sulfides

there is residual sulfate, most of the Cu is still extracted into the magmas.

According to our modeling results, at ΔFMQ > +2, the Cu content of slab melts reaches a maximum of >500 ppm at ~15% partial melting. Being a moderately incompatible element that is partitioned both into melt and hydrothermal fluid, Cu may get 3–4 times enriched during magma evolution, and an additional 3–4 times enriched during hydrothermal processes. Therefore, slab melts with initial Cu contents of >400 ppm can easily reach the average grade of porphyry Cu deposit, >4000 ppm. This can plausibly explain the close association between porphyry Cu deposits and oxidized adakites (Mungall 2002).

In contrast, mantle peridotite has much lower S and Cu contents (~250 and 30 ppm) than MORB (~1000 and 100 ppm) (Lee et al. 2012; Mavrogenes and O'Neill 1999; McDonough and Sun 1995; Sun et al. 2003, 2015). Partial melts attain maximum Cu contents of ~330 ppm at ~3% melting and oxidized conditions (ΔFMQ > +2). Nevertheless, most arc magmas form at >10% partial melting, corresponding to Cu contents of <200 ppm, which is too low to form porphyry Cu deposits (Lee et al. 2012). As a moderately incompatible element, Cu content increases by ~3 times during magma evolution, which then drops dramatically once magnetite crystallization starts at SiO_2 contents of ~58% (Sun et al. 2004). Such low Cu contents are not enough for porphyry mineralization.

To form porphyry Cu deposits through partial melting of mantle peridotite, i.e. normal arc magmas, it has been proposed that Cu is enriched via segregation of sulfide in magmas in the deep crust, followed by the remelting or dissolution in exsolving volatiles, transferring Cu in localized settings (Lee et al. 2012; Wilkinson 2013). This implies that porphyry Cu deposits may form whenever there is enough pre-enrichment. Such a model, however, is not consistent with the distribution of porphyry Cu deposits worldwide. For example, there are no porphyry deposits in Aleutian, through Japan to Izu-Bonnie–Mariana arcs (Fig. 1). If normal arc magmas may form porphyry Cu deposits through sulfide accumulations, then we should find porphyry deposits in those arcs. Instead, none exist in these areas.

3 Melting of Cu pre-enriched rocks

Our modeling shows that the melting of sulfide-enriched rocks does not necessarily form magmas with high Cu contents. It again depends strongly on oxygen fugacities. As mentioned, Cu is highly chalcophile, therefore small amount of residual sulfide may retain large amount of Cu, keeping Cu contents at low levels in magmas (Fig. 3). Nevertheless, if Cu is concentrated through sulfide saturation and accumulations in earlier magmas, then the later ore-forming pulse of magmas should be dramatically different from earlier ones. For example, later magma should be extremely oxidized and sulfide undersaturated, which requires major changes in the subduction regime. Otherwise, it cannot eliminate residual sulfide from the sulfide cumulates.

Injection of highly oxidized sulfur undersaturated magmas may trigger partial melting of sulfide-rich cumulates. In return, the oxygen fugacity of the melt is lowered during sulfide oxidations. Magmas parental to porphyry Cu

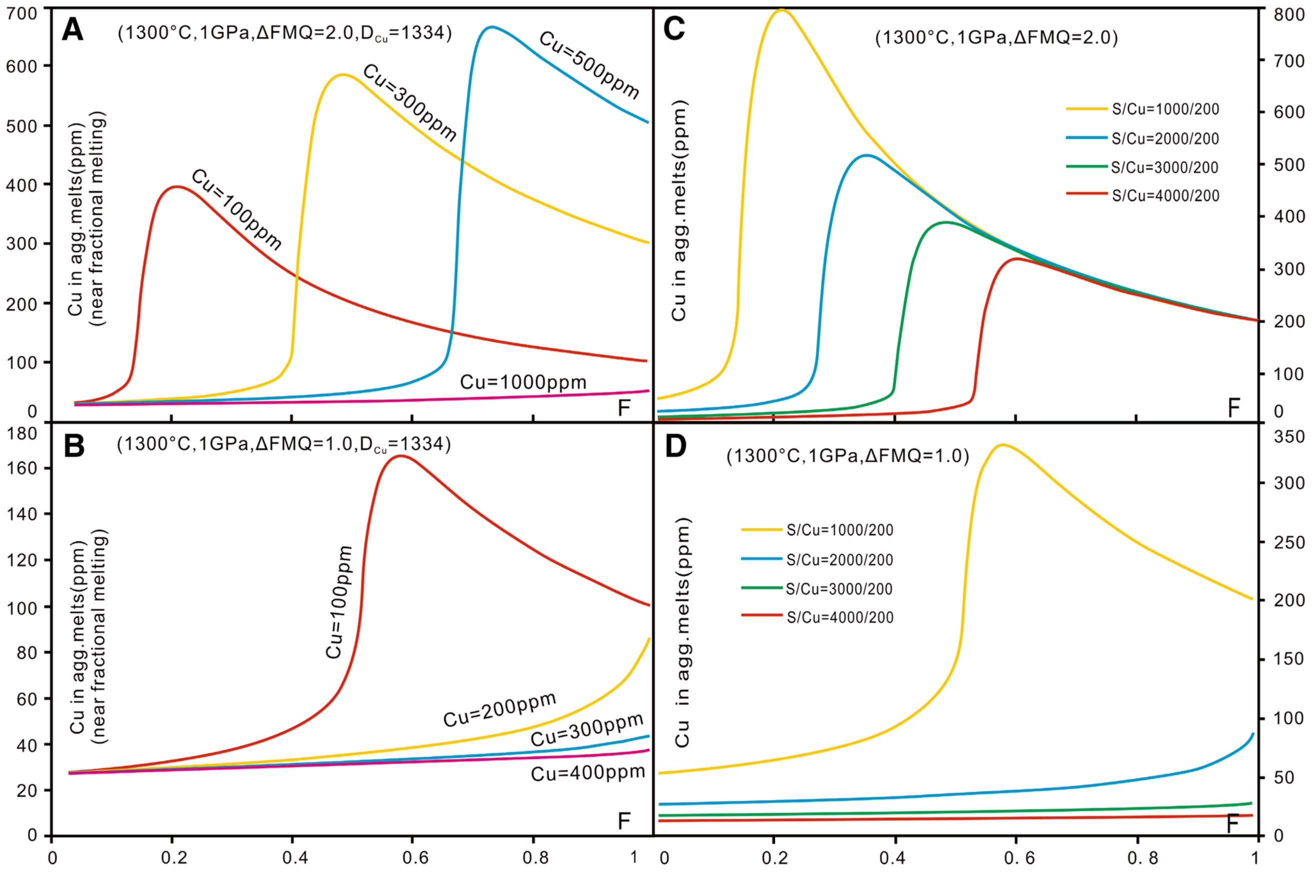

Fig. 3 Melting of sulfide cumulates through melt injection. Copper contents vary with degrees of partial melting at different Cu and S contents in the source region and different oxygen fugacities. The Cu contents may be higher than slab melts at high oxygen fugacities but are much lower at oxygen fugacities lower than ΔFMQ +1. Given that the S/Cu ratio is about 10 in the primitive mantle and MORB, a S/Cu value of 10 was assigned to sulfide cumulates in modeling **A** and **B**. At higher oxygen fugacities, melting of pre-enriched sulfide may form higher Cu peaks, but need larger degrees of partial melting (**A**). The highest Cu contents increase with decreasing S/Cu values at ΔFMQ +2 (**C**). Melts has dramatically low Cu at oxygen fugacities lower than ΔFMQ +1 (**B**, **D**)

deposits are usually highly oxidized (>ΔFMQ +2) and sulfide undersaturated (Sillitoe 2010; Sun et al. 2013). Such magmas is difficult to form oxidized magmas through melt injection induced partial melting of sulfide-rich cumulates, unless the amount of sulfide cumulates is small (Fig. 3).

Arc magmas generally have higher oxygen fugacities compared to MORB (Ballard et al. 2002; Ballhaus 1993; Kelley and Cottrell 2009; Lee et al. 2010; Parkinson and Arculus 1999; Sun et al. 2007b). Assuming the injected mafic magma has 10% of total FeO with a Fe^{3+}/total Fe ratio of 0.4 (corresponding to oxygen fugacity of ~ΔFMQ +3.3), ~2500 ppm of sulfur can be oxidized from sulfide to sulfate when all the ferric iron is used (Eq. 1).

$$7Fe_2O_3 + FeS_2 = 2FeSO_4 + 13FeO \quad (1)$$

In this case, the oxygen fugacity is lower than FMQ, and sulfur is mainly present as sulfides, i.e., not enough ferric iron to oxidize sulfide. The sulfide saturation concentration in magmas is ~1000 ppm, i.e. not much sulfide can be incorporated by magmas with residual sulfides retain Cu.

To keep the oxygen fugacity higher than ΔFMQ +2 (the magic number for porphyry mineralization, corresponding to Fe^{3+}/total Fe ratio of ~0.2), only ~1250 ppm sulfur can be oxidized to sulfate, which match up ~125 ppm of Cu, assuming a S/Cu ratio of 10 in sulfide cumulates. Such small amounts of sulfide cannot dramatically increase the ore forming capacity (Fig. 3). In case there is any residual sulfide, Cu is mostly retained, resulting in low Cu contents in magmas.

In contrast to sulfide cumulates, porphyry mineralization processes may enrich Cu through deposition of sulfide from oxidizing aqueous fluids (Sun et al. 2015). During porphyry mineralization, reduction of sulfate by ferrous iron lowers pH values. This in turn elevates the oxidation potential of sulfate, resulting in the association of sulfide with hematite and specularite (Sillitoe 2010; Sun et al. 2013, 2015; Zhang et al. 2013). Hydrothermal fluids with low pH values then react with wall rocks, which dramatically increase the pH value of the system (Sun et al. 2015). Consequently, the sulfide + hematite mineral association is not stable. Injection of sulfur (sulfide and/or sulfate) undersaturated oxidizing magmas may incorporate these hydrothermal sulfides through oxidation of sulfides by iron oxides, increasing Cu contents in the magmas up to >500 ppm, assuming a S/Cu ratio of 10 (Fig. 3). This is an efficient process that further enriches Cu, potentially forming porphyry deposits. Many superlarge porphyry Cu deposits with long mineralization process (Sun et al. 2015) may have experienced such a process.

We propose that deposition of sulfide together with hematite from oxidizing aqueous fluids is a diagnostic process for porphyry deposits. Peridotite melts have initial Cu contents too low for porphyry deposits even at very high oxygen fugacities (Lee et al. 2012), but may still precipitate some sulfide + hematite from oxidizing fluids. If pre-enrichment is high enough, they may form smaller porphyry deposits with lower grades than those derived from slab melts. At oxygen fugacities <ΔFMQ +2 (e.g., Japan, Izu–Bonin arcs), there is no co-precipitation of sulfide and hematite, and thus no porphyry deposits.

4 Discussions

Statistical results show that magmas of thicker arcs are systematically more calc-alkaline and more depleted in Cu than magmas of thinner arcs. This seemingly explains the preferential association of porphyry copper systems with thicker arcs (Chiaradia 2014; Lee 2014). For example, ~40% of the world's total Cu reserves are found along the west coast of Chile (with an average thickness of ~65 km), whereas there is essentially no porphyry Cu deposits from west Aleutian through Japan to Izu–Bonin and Ryukyu arcs (thinner than 30 km in average) (Fig. 1). This association was attributed to more sulfide cumulates within or at the base of thicker arcs (Chiaradia 2014).

Thick overriding plate is favorable to the formation of intrusive rocks over volcanic rocks. It also favors magma evolution and prevents the leakage of magmatic fluids, both of which promote porphyry deposits. Nevertheless, it is not essential to porphyry Cu deposits.

First of all, thick overriding crust all over the world does not necessarily have more porphyry Cu deposits. For example, there is not many porphyry Cu deposits in the Tibetan Plateau (with crust thicker than 70 km. Additionally, many porphyry Cu deposits in the Tibetan Plateau are older than 65 Ma, i.e., before the collision. Meanwhile, there are abundant porphyry Cu deposits in arcs thinner than 30 km, such as in the Central American and southwest Pacific arcs (Fig. 1). Most parts of the Japan arc are thicker than the Luzon and the Sulawesi arcs. There are, however, abundant porphyry deposits in the Luzon and the Sulawesi arcs, whereas there is essentially no porphyry deposits in the Japan arc (Fig. 1).

Porphyry deposits form at depths of 2–4 km (Sillitoe 2010). Thicker overriding continental crust means longer distance for the porphyry to penetrate. Moreover, the difference between 30–35 km and 45–55 km in thickness has no effect on SO_2 degassing, because high pressure experiments show that SO_2 is not stable under the pressures, temperatures and oxygen fugacity of porphyry deposit (Ni and Keppler 2012). Porphyry should not have SO_2 degassing as occurs in low-pressure epithermal deposits.

Moreover, Cu lost through sulfide saturation and accumulation is usually hosted in sulfides with low oxygen fugacities. As discussed above, sulfides need to be oxidized to sulfate to form oxidizing magmas and porphyry deposits, which cannot be easily achieved through either partial melting or melt injection.

In fact, oxidized arc magmas are not likely to be sulfide saturated because of large proportions of sulfate. Previous study showed that Cu and Au drop suddenly when magnetite crystallization started (Moss et al. 2001; Sun et al. 2004), which has been attributed to sulfate reduction and partition of sulfide into magmatic fluids under relatively constant oxygen fugacities (Sun et al. 2004) (Eq. 2).

$$SO_4^{2-} + 12FeO = 4Fe_3O_4 + S^{2-} \tag{2}$$

This has been supported by later studies, but was re-explained by sulfide accumulations (Jenner et al. 2010). As shown by very low sulfide contents, these arc magmas are not sulfide saturated (Sun et al. 2004). The close association of sulfide globules with fluids inclusions strongly suggests that sulfides went into fluid phases as hydro-sulfide complexes, not sulfide cumulates (Sun et al. 2015). Because of the solubility of water in magmas, such processes usually occur at depths shallower than 10 km (Sun et al. 2007a). Nevertheless, given that there are no major changes in pH values during the reduction of sulfate in magmas, the oxygen fugacity is relatively stable (Sun et al. 2004). Copper scavenged through this process may form hydrothermal ore deposits or dispersed as sulfides in the arc. None of these sulfides can be easily incorporated into porphyry deposits.

5 Conclusions

In summary, partial melting of subducted young oceanic slabs at oxygen fugacities >ΔFMQ +2 forms magmas with high initial Cu contents (>500 ppm), which may easily reach industrial Cu contents for porphyry deposit. Pre-enrichment of Cu through sulfide saturation and accumulation is not beneficial to porphyry Cu mineralization. Re-melting of porphyritic hydrothermal sulfide associated with iron oxides may have major contributions to porphyry deposits. Thick overriding continental crust may indeed reduce the "leakage" of hydrothermal fluids. Meanwhile, it is also more difficult for ore forming fluids to penetrate the thick continental crust to reach the depths of 2–4 km where porphyry deposits form.

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