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Weidong Sun [a] [b], Hong Zhang [a] [c], Ming-Xing Ling [a] [c], Xing Ding [a] [c], Sun-Lin Chung [d], Jibin Zhou [a], Xiao-Yong Yang [b] & Weiming Fan [a]

[a] Key Laboratory of Isotope Geochronology and Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, China

[b] Research Centre for Mineral Resources, School of Earth and Space Sciences, University of Science and Technology of China, Hefei, China

[c] Graduate University of Chinese Academy of Sciences, Beijing, China

[d] Department of Geosciences, National Taiwan University, Taipei, ROC

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The genetic association of adakites and Cu–Au ore deposits

Weidong Sun[a,b]*, Hong Zhang[a,c], Ming-Xing Ling[a,c], Xing Ding[a,c], Sun-Lin Chung[d], Jibin Zhou[a], Xiao-Yong Yang[b] and Weiming Fan[a]

[a]*Key Laboratory of Isotope Geochronology and Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, China;* [b]*Research Centre for Mineral Resources, School of Earth and Space Sciences, University of Science and Technology of China, Hefei, China;* [c]*Graduate University of Chinese Academy of Sciences, Beijing, China;* [d]*Department of Geosciences, National Taiwan University, Taipei, ROC*

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Adakites may form by partial melting of either the subducting oceanic lithosphere or the lower part of the continental crust. These two magma types can be discriminated geochemically using a combination of La/Yb, Sr/Y ratios, MgO and Na_2O contents, and Sr–Nd isotopes. Given that the basaltic crust has Cu concentrations more than two times higher than the lower continental crust and the mantle wedge, 'primitive' adakites produced by oceanic slab melting should contain significantly higher Cu contents than adakites derived from the continental crust, as well as normal arc andesites. A globally compiled dataset shows that Cu concentrations in adakites are generally lower than that in normal arc rocks. We attribute this low copper content to loss of magmatic fluids as a result of sulphate reduction during adakitic magma differentiation, in turn induced by the crystallization of Fe–Ti oxides, essential to mineralization. Therefore, the underflow of oceanic-slab-derived adakites that can release larger amounts of Cu (presumably Au as well) by crystal fractionation leads to higher potential for Cu–Au mineralization along convergent margins, usually associated with ridge subduction. Such basaltic slab melts initially have considerably higher Cu contents and thus play a crucial role particularly in the relatively closed magma system responsible for generating porphyry Cu deposits.

Keywords: adakite; Cu–Au ore deposit; slab melting; subduction

Introduction

Adakite is a rare rock type in the modern arc system. It was originally named to represent magmas with components derived from partial melting of subducted oceanic slab (Defant and Drummond 1990). Later on, it was believed that adakite could also be formed by partial melting of thickened lower crust or fractional crystallization (Defant *et al.* 2002; Kay and Kay 2002; Chung *et al.* 2003; Castillo 2006; Wen *et al.* 2008; Goss and Kay 2009). A close relationship between adakites and epithermal/porphyry ore deposits (Au, Ag, Cu, Mo) was proposed by previous authors (e.g. Thieblemont *et al.* 1997; Zhang *et al.* 2001a), who argued that most of the deposits they studied worldwide were closely associated with, and often hosted by, adakites. This notion has been supported by numerous later studies, such as those on porphyry Cu and epithermal Au deposits in the Philippines (Sajona and

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*Corresponding author. Email: weidongsun@gig.ac.cn

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Maury 1998), epithermal Au deposits in Ecuador (Beate *et al.* 2001), porphyry Cu deposit in Mongolia (Morozumi 2003) and Chile (Kay *et al.* 1999; Oyarzun *et al.* 2001, 2002; Kay and Mpodozis 2002; Reich *et al.* 2003), porphyry Cu–Au deposits in China (Hou *et al.* 2004, 2007b; Wang *et al.* 2004, 2006a, 2006b, 2007b; Xie *et al.* 2008, 2009), and Au deposits in Mexico (Gonzalez-Partida *et al.* 2003; Levresse and Gonzalez-Partida 2003; Levresse *et al.* 2004). The close association between adakites and porphyry deposits was attributed to high oxygen fugacity induced by adakitic magmas (Mungall 2002). Some researchers even proposed that the occurrence of adakites may be a useful indicator for Cu deposits (Zhang *et al.* 2004). The proposed connection between adakite and Cu deposits has also been seriously criticized by Rabbia *et al.* (2002), Richards (2002), and Richards and Kerrich (2007), who argued that the geochemical signatures of adakites can be generated in normal asthenosphere-derived tholeiitic to calc-alkaline arc magmas by common crustal interaction and fractionation processes (Richards 2002; Richards and Kerrich 2007) or by melting of thickened crust (Rabbia *et al.* 2002), and do not require slab melting. These arguments, however, do not have any constraints on the relationship between adakite and mineralization.

In fact, adakites and Cu (Au) ore deposits are not always bounded together. For example, some adakitic rocks do not have any deposits (Chiaradia *et al.* 2004; Huang *et al.* 2008), whereas some rocks without clear adakitic geochemical features are ore-bearing, for example some porphyry Cu and epithermal Au deposits in Ecuador (Chiaradia *et al.* 2004) and small porphyry Cu deposits in the western Luzon (Imai 2002); in both cases calc-alkaline andesites are the host rocks. Therefore, it has been argued that high oxygen fugacity, rather than adakitic magma composition, is essential to the formation of porphyry Cu deposits (Imai 2002; Bissig *et al.* 2003).

Adakite is defined by geochemical characteristics (e.g. $SiO_2 \geq 56$ wt.%, $Al_2O_3 \geq 15$ wt.%, $Y \leq 18$ ppm, $Yb \leq 1.9$ ppm and $Sr \geq 400$ ppm; Defant and Drummond 1990). Given that the geochemical characteristics of adakite can seemingly be produced by many geological processes with the presence of garnet (low Y) and absence of plagioclase (high Sr and Sr/Y), several other mechanisms have been proposed in addition to the original slab melting model, for example partial melting of either thickened crust (Petford and Atherton 1996; Zhang *et al.* 2001b), forearc crust carried down by subduction–erosion (Kay and Kay 2002), delaminated lower continental crust (e.g. Xu *et al.* 2002; Gao *et al.* 2004), high-pressure fractional crystallization of mineral assemblages with garnet (Castillo 2006; Macpherson *et al.* 2006), or even pure amphibole of normal arc magmas (Richards and Kerrich 2007); and polybaric fractional crystallization from exceptionally water-rich parent magmas (Rodriguez *et al.* 2007). Consequently, crustal processes have been proposed to play a key role in the metal enrichments of some porphyry Cu deposits (Richards and Kerrich 2007).

In this article, we used compiled data from the GEOROC dataset to conduct geochemical modelling that enables us to evaluate the above debates with emphasis on the relationship between adakites and Cu (Au) ore deposits.

Copper in adakites

Compiled GEOROC dataset

The compiled GEOROC dataset for Cu and SiO_2 contents of arc volcanic rocks are plotted in Figure 1. Given the fact that high-quality Au data are rare and that Au and Cu behave similarly during magma differentiation for arc volcanic rocks (Sun *et al.* 2004),

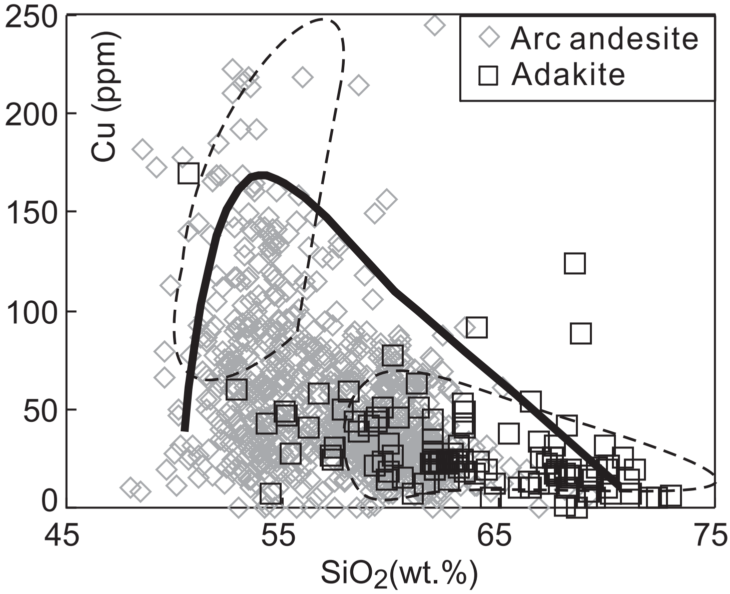

Figure 1. Diagram of Cu versus SiO_2 for adakites and normal arc rocks indicating Cu loss in evolved magmas. Data source: GEOROC. The two zones confined by dashed lines are modified after Sun *et al*. (2004), representing values of eastern Manus basin volcanic glasses for comparison. Cu concentration increases at the early stage of magma fractionation and then drops suddenly as a result of magnetite crystallization, which reduces sulphate, scavenging Cu, Au out of the magmas in the form of hydrosulphide complexes (Sun *et al.* 2004).

we will focus on Cu in the following discussion. We did not change the classification of the GEOROC dataset, except to exclude data published before 1991, as the term 'adakite' was first proposed in 1990 (Defant and Drummond 1990). Some rocks that were classified as adakites by the original authors were excluded from the compiled normal arc andesite dataset.

Compared to normal arc volcanic rocks, adakites have systematically lower Cu concentration (Figure 1). To most people, this phenomenon does not lend any support to a genetic link of Cu (Au) deposits with adakites. It is probably one of the main reasons many geologists do not believe the association between Cu deposits and adakites. Figure 1, however, cannot be used to rule out a possible association. This is because Cu and Au concentrations may drop quickly when magmas evolve to higher SiO_2 contents ($\sim$58 wt.%) because of the oxygen fugacity fluctuation induced by crystallization of Fe–Ti oxides and subsequent sulphate reduction that scavenges Cu, Au into magmatic fluids (Sun *et al.* 2003a, 2004; Liang *et al.* 2006, 2009). These Cu-rich fluids/gases released during magma evolution are important for the transportation and mineralization of Cu (Heinrich *et al.* 1999, 2004; Seedorff *et al.* 2005).

Although the compiled Cu data for normal arc rocks do not drop abruptly as SiO_2 increases, they show a Cu peak at SiO_2 of $\sim$55–60 wt.%. This is likely because the dataset is not representative of samples from a single magma chamber, but a large collection from the convergent margins worldwide. Nevertheless, the Cu peak is consistent with the notion that there is a major change in Cu behaviour during magma differentiation (Sun *et al.* 2004). For adakites, in contrast, Cu concentrations drop continuously with increasing

SiO_2 contents without any changes in Cu behaviour (Figure 1). This is probably because Fe–Ti oxides may start crystallizing at the very beginning stage of the magma evolution of adakites that have relatively higher SiO_2 (Defant and Drummond 1990) and arguably higher oxygen fugacity (Mungall 2002) than normal arc lavas, and thus may have released more Cu for mineralization.

The speculation is supported by plots of FeO and TiO_2 versus SiO_2 (Figure 2), in which FeO and TiO_2 of normal arc rocks are both peaked at SiO_2 of ∼55–60 wt.%, whereas those of adakites decrease continuously. The message of these diagrams (Figures 1 and 2) is that crystallization of Fe–Ti oxides indeed removes Cu dramatically from adakites and also some evolved normal arc rocks, most likely by sulphate reduction as previously proposed (Sun *et al.* 2004). Therefore, Cu concentrations cannot be used directly as a geochemical parameter to prove or disapprove the link between adakites and Cu (Au) deposits. Also, these diagrams tell us that adakites have given away Cu during magmatic processes, most likely to fluids (Sun *et al.* 2004). Therefore, in the case that adakites originally contain higher Cu concentrations than normal arc rocks, they have a higher potential of causing Cu mineralization. An immediate question, then, is whether ‘primitive’ adakitic magmas have high Cu contents.

Modelling results

Copper concentration in ‘primitive’ adakites depends heavily on the source composition and oxygen fugacity. As stated above, there are three types of petrogenetic models proposed

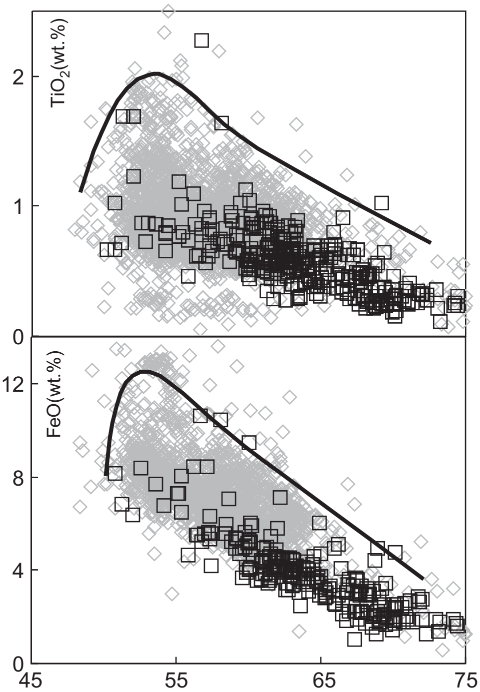

Figure 2. Diagrams of FeO and TiO_2 versus SiO_2 for adakites and normal arc rocks. Symbols are the same as Figure 1. FeO, TiO_2 losses are in phase with Cu loss in Figure 1. Data source: GEOROC.

for adakite formation: slab melting, thickened lower continental crust melting, and fractional crystallization. The continental crust and lower continental crust have Cu abundance estimated as low as 26–27 ppm (Rudnick and Gao 2003), much lower than the oceanic crust represented by MORB that contains 60–120 ppm (e.g. 74.4 ppm in average) of Cu (Hofmann 1988; Sun *et al.* 2003b).

The partition coefficients for Cu vary dramatically as shown by experimental data and natural samples (GERM 2009), possibly because of its chalcophile characteristics. Nevertheless, it is moderately incompatible in natural samples ranging from MORB to arc volcanic rocks, similar to Re (Sun *et al.* 2003a, 2003b, . 2004). Therefore, partial melting of either the lower continental crust or subducted slab would produce magmas with Cu considerably higher than the corresponding sources. The partition coefficients chosen in our modelling are 0.05 for amphibole, 0.5 for plagioclase (Dostal *et al.* 1983), 0.5 for garnet, 0.2 for plagioclase and 1 for rutile. The partition coefficients for Sr, Y are from Rollinson (1993) for plagioclase and Xiong *et al*. (2006) for other minerals (Table 1).

In a Sr/Y versus Cu diagram (Figure 3), melts modelled for partial melting of MORB have Cu (114–245 ppm, using an average Cu of 74 ppm) levels much higher than that of the lower crust (40–85 ppm, using an abundance of 27 ppm). The modelling results indicate that, while the Cu-enriched slab melts are likely to be closely associated with Cu ore formation, melts of the lower continental crust that show significantly lower Cu contents may have no connection to Cu mineralization at all. This explains the positive correlation between tonnages of Cu ores and the mantle components identified in large Chinese porphyry deposits (Hou *et al.* 2007a), because mantle components identified by

Table 1. Modelling results of slab melts and melts derived from the lower continental crust.

			A	B	C	D
Bulk Partition Coefficients		Sr	0.335	0.210	0.051	0.030
		Y	1.57	3.77	6.70	6.69
		Cu	0.226	0.309	0.459	0.503
Lower continental crust	10%	Sr	866	1205	2387	2733
		Y	10.6	4.58	2.61	2.61
		Cu	85.7	68.8	50.7	47.0
	20%	Sr	743	946	1446	1551
		Y	11.0	4.97	2.88	2.88
		Cu	68.3	58.2	45.9	43.2
	30%	Sr	651	779	1037	1083
		Y	11.5	5.44	3.21	3.21
		Cu	56.7	50.4	41.9	39.9
MORB	10%	Sr	444	618	1224	1401
		Y	20.9	9.02	5.15	5.15
		Cu	245	197	145	135
	20%	Sr	381	485	741	796
		Y	21.7	9.80	5.67	5.68
		Cu	195	167	131	124
	30%	Sr	334	399	532	556
		Y	22.6	10.7	6.32	6.33
		Cu	162	144	120	114

Note: Mineral compositions are A: amphibole 55%, garnet 4.3%, clinopyroxene 30%, plagioclase 10%, rutile 0.7%; B: amphibole 40%, garnet 19.3%, clinopyroxene 35%, plagioclase 5%, rutile 0.7%; C: amphibole 10%, garnet 39.3%, clinopyroxene 50%, plagioclase 0%, rutile 0.7%; D: amphibole 0%, garnet 39.3%, clinopyroxene 60%, plagioclase 0%, rutile 0.7%. The compositions of lower crust were from Rudnick and Gao (2003), whereas those of MORB are from Hofmann (1988) for Cu and the average values of Sun *et al.* (2008) for other elements.

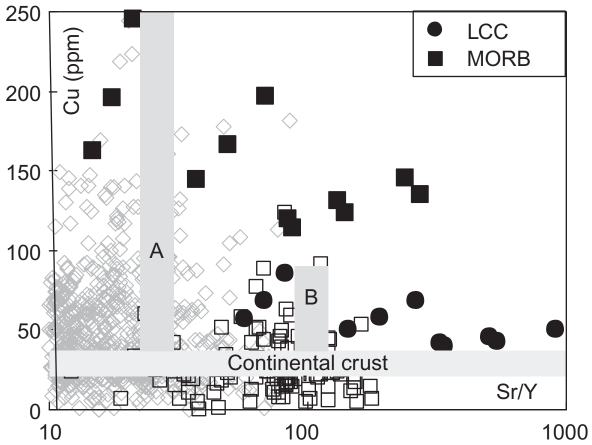

Figure 3. Diagram of Cu versus Sr/Y showing calculated partial melts of MORB having systematically higher Cu than those of the lower continental crust melts and normal arc rocks (Table 1). Backgrounds are normal arc (open light grey diamonds) and adakites (open grey squares) from GEOROC. Continental crust value from Rudnick and Gao (2003). Note: Both Cu and Sr/Y change dramatically during magma fractionation. Bars A and B represent the maximum amounts of Cu that can be released for mineralization from slab melts and lower crust melts, respectively.

isotopic composition are likely related to slab melts, which usually have isotopic compositions identical to mantle rocks (e.g. Sun and McDonough 1989; Hacker 1991). From this point of view, the association of adakitic rocks with Cu ore deposits might be indicative of a magma origin by slab melting.

Discussion

Slab melting versus lower continental crust melting

Adakite was originally defined as rocks with slab melts (Defant and Drummond 1990). The geochemical characteristics of adakites, however, can be created through three ways: slab melting, melting of thickened crust, and fractional crystallization (Defant *et al.* 2002; Kay and Kay 2002). Both melting of thickened crust and fractional crystallization have been proposed as key factors that control Cu mineralization (Bissig *et al.* 2003; Hollings *et al.* 2005). Our modelling results, however, show that adakites formed by slab melting tend to have higher initial Cu concentrations that could facilitate Cu mineralization. On the other hand, those formed by partial melting of the lower continental crust have considerably lower Cu, and thus poor opportunities in ore formation because of the lower Cu in the source and lower oxygen fugacity. Therefore, in terms of Cu deposit exploration, it is important to distinguish slab melts from lower crust melts using certain petrogenetic indicators.

Isotope is arguably the most powerful discrimination parameter. In general, slab melting produces magmas with isotope compositions close to MORB values, which are usually similar to that of the depleted mantle whereas partial melting of the lower continental crust usually forms magmas with enriched isotope signatures (Wang *et al.* 2006a, 2007b; Huang *et al.* 2008; Wen *et al.* 2008). Therefore, isotopes are often used to constrain the sources of adakites (e.g. Dreher *et al.* 2005; Macpherson *et al.* 2006; Wang *et al.* 2007a). However, the isotope ratios of adakites may be modified by magmatic processes, for example crust contamination (Davidson and Desilva 1995; Mori *et al.* 2007; Ling *et al.* 2009) and sediment contributions (Kay *et al.* 1978; Sajona *et al.* 2000). Therefore, it could be problematic to rely solely on isotopic constraints.

La/Y, Sr/Y ratios are another useful parameter. The lower continental crust is more enriched in La, Sr and depleted in Y, Yb than average MORB (Hofmann 1988; Sun and McDonough 1989; Rudnick and Gao 2003); thus, lower continental crust melts should contain higher La/Yb, Sr/Y at given Y, Yb than slab melts. The La/Yb, Sr/Y ratios of adakites, however, are highly varied (Defant and Drummond 1990), depending heavily on the partial melting conditions. In the case that the lower continental crust was melted in the presence of plagioclase and/or absence of garnet, the resultant magmas may have Sr/Y ratios comparable to slab melts. Therefore, Sr/Y values are not always conclusive, either. La/Yb is less affected by plagioclase, such that lower continental crust melts may have distinctively higher La/Yb than slab melts.

MgO and Mg# may also be different in slab and lower continental crust melts. In general, melts from a subducting slab would interact with the overlying mantle wedge during magma ascent and thus gain considerable amounts of MgO that raise Mg# numbers (Kilian and Stern 2002; Xiong *et al.* 2006; Gomez-Tuena *et al.* 2008). In contrast, lower continental crust melts presumably stay mainly in the crust and have lower MgO. These scenarios, however, are not always right, either. For example, flat subduction may squeeze or erase the mantle wedge, forming low-Mg adakites by slab melting. In addition, when the lower continental crust is melted through the addition of upwelling mantle materials, for example asthenosphere or mantle plume, the MgO contents can be elevated.

Na_2O of adakites produced by slab melting are systematically lower than that of experimental results, a feature that has also been attributed to mantle interaction of slab melts (Xiong *et al.* 2006). In contrast, melts from the lower continental crust have higher Na_2O (Xiong *et al.* 2001). The systematically lower Na_2O contents in slab melts compared to lower continental crust melts may also be ascribed to the presence of omphacite in the slab melting residue, as omphacite is a Na-clinopyroxene that could hold back a large portion of Na. Nevertheless, Na_2O contents of adakites may be significantly changed during magma differentiation, such that using only this constraint would not be conclusive either.

Consequently, there seems to be no easy solution for discriminating oceanic slab melts from lower continental crust melts. Even so, the more the above criteria match, the better one may constrain the source and origin of adakites. It is also worth mentioning that slab melts may well be contaminated by the lower continental crust through assimilation, especially in places where thick crust exists (Ling *et al.* 2009). Moreover, there is nearly no magma that can be well preserved from fractional crystallization and assimilation, both of which can dramatically change the composition of the magma. Many adakitic magmas can easily change to no-adakitic characteristics after plagioclase crystallization. In this case, the association of adakites with or without Cu (Au) deposits may provide an additional constraint on the tectonic setting and/or petrogenesis.

Tectonic settings are probably more important than geochemical characteristics for identifying slab melts. Ridge subduction and flat subduction are the most favourable tectonic settings for slab melting. In fact, most of the large porphyry Cu deposits in Chile and Peru are spatially associated with ridge subduction (Cooke *et al.* 2005; Sun *et al.* 2010). This strongly supports our model because ridge subduction is the most favourable process for the formation of adakite.

Ore formation related to normal arc rocks

To confirm the close association between slab-derived adakites and Cu (Au) ore deposits, we take a look at normal arc rocks in terms of Cu concentrations. Copper is a moderately incompatible element in the presence of sulphur (Sun *et al.* 2003a, 2004). The mantle wedge is fairly depleted in incompatible elements, so its Cu abundance is mainly controlled by the addition of Cu from the subducting slab. It has been suggested that aqueous fluids liberated by the subducting slab at the blueschist to eclogite facies transition are dilute, containing only moderate amounts of large-ion lithophile elements, Sr, and Pb and do not transport significant amounts of key elements (Hermann *et al.* 2006). If this is true for Cu, the mantle wedge should have Cu lower than the primitive mantle (30 ppm) (McDonough and Sun 1995), and therefore normal arc magmas should contain Cu much lower than slab melts.

In Figure 1, normal arc rocks from the compiled data of GEOROC appear to have fairly high Cu concentration. Many of these arc rocks, however, have high enough Sr/Y ratios that they can safely be classified as adakitic rocks; in particular, those samples with Cu >150 ppm are actually adakites as constrained by their very high Sr/Y ratios (Figure 3). Moreover, nearly all the arc rocks, including those that are actually adakites, experienced different degrees of plagioclase crystallization; therefore, some of the arc rocks that have high Cu concentrations (Figures 1 and 3) might be originally of adakitic compositions. On the other hand, it has been proposed that an adakite-type slab melt component may be present in the magmatic source throughout the arc system (Yogodzinski and Kelemen 1998). In that case, slab-released fluids cannot transport much Cu, so the proportion of the slab melt component may determine the Cu concentration in arc magmas.

As shown in Figures 1 and 3, normal arc rocks also lose significant amounts of Cu during magma differentiation, and thus likely contributed to Cu mineralization at the convergent margins, in particular for ore formation in epithermal deposit systems. Precipitation of metals depends on many factors, including temperature, acidity, and iron and sulphide availability (Seedorff *et al.* 2005; Liang *et al.* 2009). For a closed magma system (e.g. porphyry), instead specific processes are required to elevate the Cu concentration from 4000 ppm. It is much easier to accomplish this elevation by slab melts that originally contain higher Cu. This, if true, provides a plausible explanation for the observed association between Cu (Au) deposits and slab-derived adakites (e.g. Thieblemont *et al.* 1997; Sajona and Maury 1998; Wang *et al.* 2006a, 2006b).

Conclusions

Primitive adakites derived by partial melting of oceanic lithosphere should have systematically higher Cu contents than those from the lower parts of thickened continental crust because Cu concentrations in the former are much higher than those in the latter. The incompatible characteristics of Cu suggests that concentration of this element in the mantle wedge is also likely to be lower than that in the oceanic crust, unless subduction-released fluids have a very high capacity of transporting Cu to the mantle wedge.

Both adakites and normal arc rocks evidently release Cu and presumably also Au during magma differentiation, so they both may contribute to ore mineralization, especially epithermal deposits. The higher Cu abundance in primitive adakites formed by oceanic slab melting implies that more Cu can be released from such magmas, which are thus favourable for ore mineralization, supporting the close relationship of slab-derived adakites with Cu ± Au ore deposits. For a closed magma system, adakites generated from melting of the oceanic lithosphere have a better chance at mineralization because of their higher Cu concentrations.

Adakites from the lower continental crust apparently possess lower initial Cu contents, thus offering fewer prospects for extensive mineralization. This type of adakite may be discriminated from basaltic slab melts using a combination of Sr/Y and La/Yb ratios, MgO and Na_2O contents, and Sr–Nd isotopes. For magmas consisting of both slab and lower continental crust components, the level of Cu mineralization may provide an additional constraint on their origin. These are supported by the close association of ridge subduction with large Cu deposits, reflecting the most favourable tectonic setting for slab melting.

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