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Native gold and native copper grains enclosed by olivine phenocrysts in a picrite lava of the Emeishan large igneous province, SW China

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Abstract

A native gold bleb found in an olivine phenocryst in a picrite lava from the Emeishan Large Igneous Province (ELIP) may be the first documented case of the transport of gold as a distinct precious metal phase in a mantle-derived magma. Four picrite layers have been recognized in the lower part of the volcanic succession in the Lijiang area, in the western part of the ELIP. The native gold bleb was found enclosed in an olivine phenocryst in the second picritic layer of a basalt-picrite succession in the ELIP. The gold bleb is spheroidal, about 30 μm in diameter, consists of pure gold, and does not contain any other elements. In addition, native copper grains were also discovered in the serpentinized olivine phenocrysts, and native zinc and moissanite (SiC) were separated from an ~20 kg sample of picrite. The paragenesis of these minerals suggests that the primary magmas were S-unsaturated. The native gold and native copper grains are considered to be xenocrysts from the mantle, transported to shallow depths by a rising plume, and then captured by the picritic melts. The discovery of native gold and native copper grains provides direct evidence that the gold in the hydrothermal gold deposits and the native copper deposits in the ELIP lavas ultimately may be derived from a mantle plume.

Keywords: Native gold, native copper, moissanite, mantle plume, olivine phenocryst, picrite, Emeishan large igneous province

Introduction

The production of continental flood basalts (CFBs) has been attributed to the arrival of plume heads from a boundary layer deep within the earth (e.g., Richards et al. 1989; Hill 1991). This has been suggested to be either the 670 km discontinuity or the 2900 km D" core-mantle boundary layer (Allegre and Turcotte 1985; Hofmann 1997). However, the chemical composition and mineralogy of mantle plumes is poorly known because of the lack of mantle xenoliths in the lavas from large igneous provinces (LIPs). Highly magnesian, near-primitive picritic lavas in LIPs have been used to infer the characteristics of a mantle plume (e.g., Brugmann et al. 1987; Puchtel and Humayun 2000). This is possible because of the specific conditions of picrite formation, including high-temperature partial melting, consistent with formation of picritic magmas from mantle plumes (e.g., Campbell et al. 1989; Storey et al. 1991), sulfur undersaturation, rapid adiabatic ascent, and eruption at temperatures close to the liquidus. These conditions ensure that little fractionation occurs prior to eruption (e.g., Arndt et al. 1977; Herzberg 1995). Thus, the picrites should preserve some signatures of materials derived from a mantle plume.

A bleb of native gold and two grains of native copper were discovered in the olivine phenocrysts from the same picrite sample from the Late Permian Emeishan large igneous province (ELIP). This is the first documented case of the occurrence of native gold and native copper in the olivine phenocrysts of picrites. We suggest that it may reflect the signature of plume-derived materials.

Geology and petrography of picrites

The ELIP is a Late Permian continental flood basalt sequence covering large areas in the provinces of Sichuan, Yunnan, and Guizhou along from the eastern margin of the Tibetan Plateau to the western margin of the Yangtze Block in southwestern China (Fig. 1). The ELIP covers an area of at least 250 000 km^2 (Chung et al. 1998; Chung and Jahn 1995; but according to Xiao et al. 2003, this is a conservative estimate) in SW China. The western boundary of the ELIP is the Ailao Shan-Red River Fault zone (ASSR), a major crustal structure separating the Yangtze Craton from the Gandise and Yunnan fold belts.

The ELIP consists of a succession of predominantly tholeiites, with minor picritic and rhyolitic lava flows. In addition to lava flows, mafic-ultramafic layered complexes, dikes and sills, syenite, and other alkaline intrusions, are part of the ELIP (Xu et al. 2001; Boven et al. 2002; Zhang et al. 2004b; Xiao et al. 2004). The maximum thickness of the lava flows is estimated at about 5400 m in the Binchuan region in the Pan-Xi rift, decreasing to less than 500 m eastward from the rift (Xiao et al. 2003, 2004; He et al. 2003; Xu et al. 2004).

The ELIP magmatism appears to be associated with Late Permian rifting of the Qiangtang terrane from the Yangtze block. The flood basalts were subsequently uplifted, deformed, and seg-

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mented by block faulting during the closure of the Tethys Ocean in the Jurassic (Chung et al. 1998). The region was reworked further during the collision between the Indian and Eurasian continents (Leloup et al. 2001).

The total eruptive volume is estimated to be between 0.3 and 0.5×10^6 km^3 (Yin et al. 1992; Jin and Shang 2000); however, given the large amount of erosion, and by comparison with other continental flood basalt privinces, this volume may represent less than 50% of the total amount of magma emplaced. The lava successions rest unconformably on the Early Permian Maokou Formation, composed chiefly of marine limestones, and are overlain directly by Late Permian and locally Early Triassic marine strata. Lo et al. (2002) concluded from ^{40}Ar-^{39}Ar dating that the main phase of flood basalt magmatism occurred at 251–253 Ma, more or less coeval with the Siberian Traps, which also contain picrites (Sharma 1997).

Picrites are present in the southern (Shiman) and northern (Daju) portions of Lijiang county, Yunnan province. These two occurrences are about 26 km apart. The geological characteristics of the two picrite occurrences are similar, with the picritic lavas being located in the lower portions of the flood basalt pile. In the Shiman section, three picritic flows are intercalated with pyroxene-phyric basaltic lavas, whereas four picritic flows occur in the lower part of the Daju section. It is possible, perhaps likely, that the Shiman section corresponds to the middle part of the much thicker Daju section. Massive aphyric basalts and amygdaloidal basalts are dominant in the middle to upper part of the Daju sequence. In the Shiman section, the lowermost and the uppermost picritic flows are 3 to 5 m thick, and the middle picritic layer is 15 to 20 m thick. In the Daju section, the lowermost picritic flow varies from 20 to 50 m in thickness, whereas the thickness of each of the other three flows is similar to those in the Shiman section. Unlike the central and eastern parts of the ELIP, where plagioclase-phyric basalts are widely exposed, in the Lijiang region, plagioclase-phyric basalts are absent.

The native gold-bearing picrite (DJ03-35) is located in the second picritic layer of the Daju section (Fig. 2). The picrite is highly porphyritic, and contains abundant forsteritic olivine phenocrysts with minor clinopyroxene and Cr-spinel. Olivine phenocrysts are generally euhedral to subhedral (Fig. 3a), ranging from 0.2 to 4 mm in size; Strained, kink-banded crystals are absent, but fractures usually occur in the olivine crystals. The olivine phenocrysts contain scattered inclusions of glass, and some enclose small, equant, euhedral to rounded, dark-brown Cr-spinel crystals, usually tens of micrometers across. Some Cr-spinel crystals are also present in the groundmass, which consists of fine-grained unaltered olivine, diopside, and plagioclase. Quenched olivine groundmass crystals are generally fresh or unaltered, and may be small (<0.1 mm), and near-equant "hop-

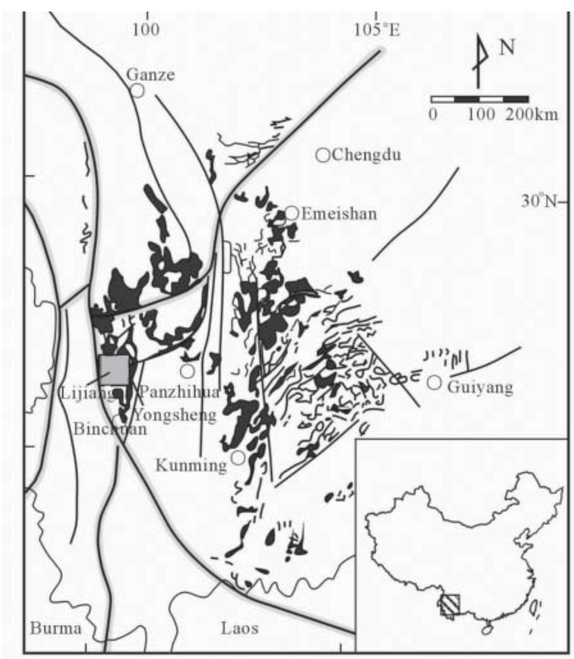


FIGURE 1. Simplified map showing outcrops of Emeishan flood basalts (black areas; simplified from Chung and Jahn 1995). The shaded area marks the location of the Figure 2.

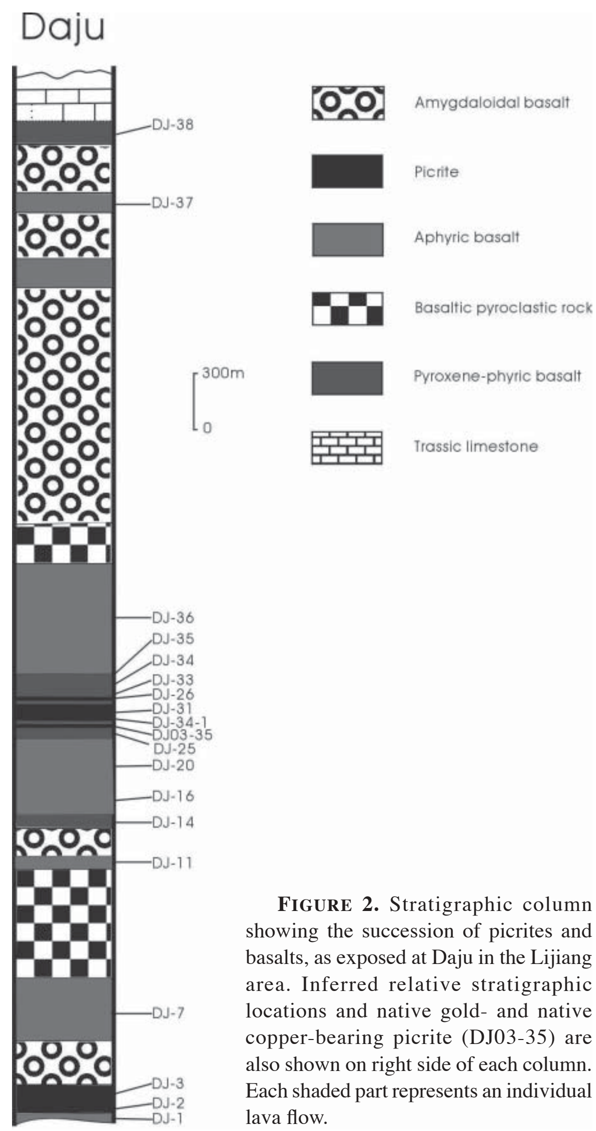
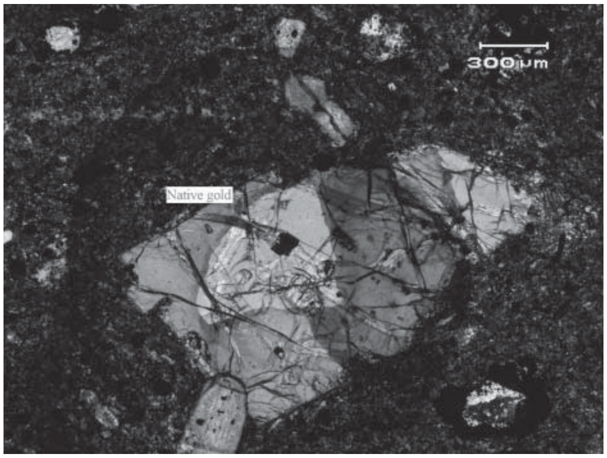


FIGURE 2. Stratigraphic column showing the succession of picrites and basalts, as exposed at Daju in the Lijiang area. Inferred relative stratigraphic locations and native gold- and native copper-bearing picrite (DJ03-35) are also shown on right side of each column. Each shaded part represents an individual lava flow.



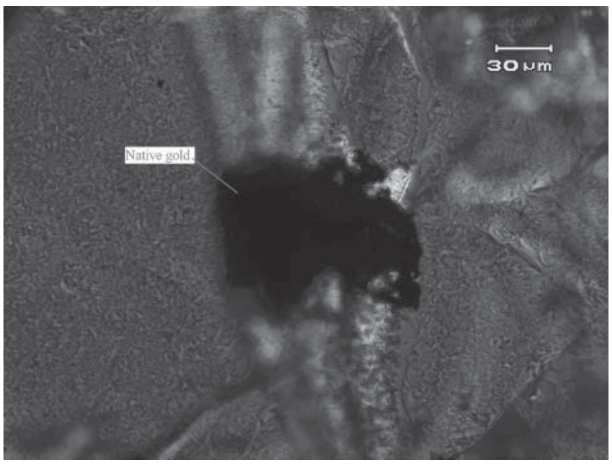


FIGURE 3. Photomicrographs of the native gold bleb enclosed by the olivine phenocryst in the picrite. (**a**) Olivine phenocrysts and the groundmass of the picrite. The black grain enclosed by olivine phenocsyst is native gold. The blue circle is an ink mark. Cross-polarized light; (**b**) the entire outline of native gold bleb can be recognized under cross-polarized light; (**c**) only part of native gold bleb is exposed in the polished thin section, whereas the unexposed appears as a blurred image under reflected light.

pers." Commonly, however, they are elongated, up to 0.5 mm long and 0.1 mm wide, skeletal, and consist of parallel-set rods and blebs in optical continuity. Groundmass plagioclase occurs as microcrystals rather than the long platy shapes seen in the basalts, and clinopyroxene is anhedrdal.

Electron microprobe analyses show that the olivine phenocrysts are magnesian ($Fo_{85.2}$–$Fo_{91.4}$) with 0.22–0.45 wt% CaO and 0.03–0.17 wt% Cr_2O_3 (Zhang et al. 2005a). Olivine in mantle peridotites is characterized by much lower Ca and Cr contents (e.g., Gurenko et al. 1996; Thompson and Gibson 2000). Along with the presence of glass inclusions and lack of strain texture, these features indicate that the olivines in the ELIP picritic lavas investigated in this work crystallized from a melt and are not accidental xenocrysts of mantle olivine.

The MgO content of this picrite sample is 19.42 wt%, and it is characterized by enrichment of light rare earth elements and large ion lithophile elements, and absence of negative Nb and Ta anomalies. The ($^{87}Sr/^{86}Sr$)t and (ε_{Nd})t values are 0.70425 and +2.7, respectively (Zhang et al. 2004b).

DISCOVERY AND OCCURRENCE OF GOLD AND COPPER

We collected a 20 kg sample of picrite (DJ03-35) in the Lijiang (Yunnan Province), and obtained mineral separates including native gold, native copper, native zinc, and moissanite (SiC). To understand the paragenesis of these phases, we made 35 polished thin sections, and discovered a bleb of native gold and two grains of native copper in two of these sections, respectively. The gold bleb is spheroidal, ~30 μm in diameter, and enclosed by a fresh olivine phenocryst in the picrite (Fig. 3). The copper grains are anhedral to subhedral, and about 20 μm in diameter (Fig. 4). However, they are enclosed in serpentinized olivine phenocrysts. Unfortunately, we were not able to detect the other separated minerals (e.g., native zinc and moissanite) in the polished thin sections.

The polished thin section containing the gold bleb was never coated with gold. The carbon coat was later removed from the gold-bearing grain by scrubbing with a cotton swab and solvent to verify that the bleb is not a surface contamination. Although the gold bleb is not exposed completely, it can be confirmed that it is not an anthropogenic contaminant introduced during sample preparation.

COMPOSITIONS OF NATIVE GOLD AND NATIVE COPPER

The grains were analyzed using a JXA8800R microprobe and a S-3500N Scanning Electron Microscope equipped with an energy dispersive spectrometer at the Institute of Mineral Resources of Chinese Academy of Geological Sciences. The operating conditions were 20 kV accelerating voltage and 15 nA beam current. SW9100 NIST multiple element standards were used for calibration. Detection levels for Se, Te, Bi, Hg, As, and PGE were 0.03 wt%. Results are presented in Table 1.

Four microprobe analyses of the native gold bleb reveal a very homogeneous composition, characterized by only gold. Other elements such as Se, Te, Ag, Cu, Hg, Bi, Os, Ir, Pd, and Pt are below detection levels from center to rim of the grain (Table 1). In contrast, the native gold grains in rocks affected by hydrothermal processes always contain varying amounts of silver (Chisholm

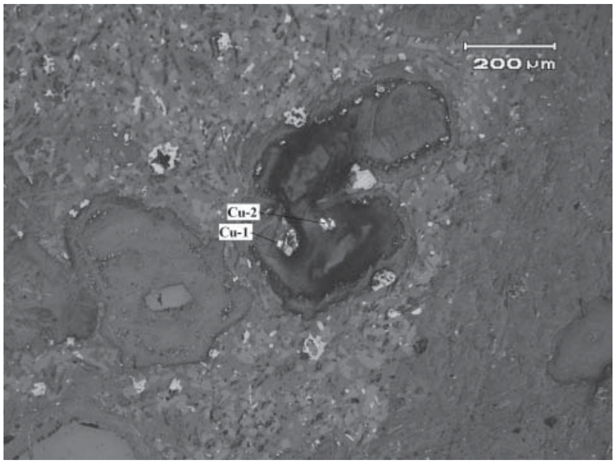


FIGURE 4. Photomicrograph of native copper grains enclosed by the serpentinized olivine phenocryst in the picrite (reflected light).

TABLE 1. Electron microprobe analyses of native gold (Au) and native copper (Cu) grains

wt%	Au-1	Au-2	Au-3	Au-4	Cu-1	Cu-2
Au	100.08	100.50	100.68	100.04	0.00	0.00
Ag	0.00	0.00	0.00	0.00	0.00	0.00
Pt	0.00	0.00	0.00	0.00	0.00	0.00
Pd	0.00	0.00	0.00	0.00	0.00	0.00
Cu	0.00	0.00	0.00	0.00	98.45	98.49
Si	0.00	0.00	0.00	0.00	0.68	0.60
Al	0.00	0.00	0.00	0.00	0.95	0.92
Total	100.08	100.50	100.68	100.04	100.08	100.01

1979; Piestrzyński and Sawlowicz 1999), whereas those associated with mafic-ultramafic intrusions usually contain minor platinum-group elements (PGE) and silver or copper (Boyle 1987; Robinson et al. 2004). The native copper grains contain minor Si and Al, both of which are less than 1 wt% (Table 1). These compositions exclude the possibility that the native copper is the result of anthropogenic contamination introduced during sample preparation because the commercially refined copper or copper alloys do not contain either Si or Al.

DISCUSSION

Natural metallic alloys have been reported from many localities and have been identified both in thin sections of ultramafic rocks (e.g., Stockman and Hlava 1984; Legendre and Augé 1986; Melcher et al. 1997, Malich 1999; Bai et al. 2000; Robinson et al. 2004) and in placers or soils associated spatially with ultramafic rocks (Cabri and Harris 1975; Bird and Bassett 1980; Nakagawa and Franco 1997). Metallic alloys also are associated commonly with Cr-spinel in layered basic intrusions (Naldrett et al. 1987; Kinloch and Peyerl 1990). To our knowledge, only in the Kilauea volcano district, Hawaii, has native gold been discovered in a Fe-sulfide grain in volcanic rocks (Sisson 2003). The native gold hosted in an olivine phenocryst in the ELIP picritic rocks is the first documented case of its kind.

Native gold is generally considered the product of hydrothermal or magmatic processes. However, the gold bleb reported in this paper is enclosed in the fresh olivine phenocryst and does not occur as a fracture filling. Moreover, native gold formed by hydrothermal processes always contains minor Ag and other elements such as Te, Bi, Cu, Sb, etc. (Chisholm 1979; Fulignati and Sbrana 1998). Thus, the possibility that the gold identified here is a product of alteration of the sample or hydrothermal processes can be excluded. Sisson (2003) proposed that the gold bleb in the Hawaii basanite was formed by precipitation from an immiscible gold liquid resulting from resorption of magmatic sulfide during crystallization-differentiation. The determination of the gold content of the picrite in this study shows that the whole rock contains only 3.8 ppb Au (our unpublished data), consistent with the gold concentrations of normal picrites. As gold has very low solubility in silicate melts (Borisov and Palme 1996), it is unlikely that the gold bleb was formed by the precipitation of an immiscible gold liquid at the magmatic stage. In addition, the presence of native copper, native zinc, and SiC indicates an origin under reducing conditions. In general, tholeiites (including picrites) form under lower oxygen fugacity compared to calc-alkaline basalts (Helz 1973; Macdonald et al. 2000). From this point of view, the original magma should have been sufficiently enriched in elements such as Cu, Zn, and Au to form primary minerals. On the other hand, because both copper and zinc are chalcophile elements and therefore highly compatible with S, the presence of native copper and native zinc and absence of sulfide in the picrites indicates that the primary magma was strongly S-undersaturated. Based on the PGE concentrations of the picrites, Zhang et al. (2005) also concluded that the picritic magmas were S-undersaturated. Thus, it is unlikely that the gold bleb was released from sulfides during the progressive resorption of sulfides.

Native copper grains also were discovered in some sedimentary rocks, and they were interpreted to result from diagenetic or low-temperature hydrothermal processes (Subías et al. 2003; Distler et al. 2004). Although native copper is hosted in the serpentinized olivine phenocrysts, the mineral is unlikely to have resulted from serpentinization or diagenesis because it contains minor Si and Al, whereas the native copper formed by alteration generally does not include Si or Al (Bai et al. 2000; Buseck 1968). Bai et al. (2000) proposed that Si-bearing native elements were derived from deep-mantle sources. Therefore, we infer that these native copper blebs also may be derived from a deep-mantle source and are not the result of alteration processes.

Natural SiC occurs as hexagonal and trigonal polymorphs (α-SiC) known as moissanite (Leung 1990) and as a cubic polymorph (β-SiC). Moissanite (SiC) is a naturally occurring mineral closely associated with diamonds. It occurs both as inclusions within diamonds (Moore and Gurney 1989; Otter and Gurney 1989; Leung 1990; Leung et al. 1990, 1996) and as an accessory mineral in diamond-bearing rocks. Therefore, it is generally considered an ultrahigh-pressure mineral. Although we have not established the paragenetic relationship of native gold, native copper, and native zinc with moissanite in the thin sections, it is likely that these minerals have an ultrahigh-pressure origin.

Recently, Zhang et al. (2005a) proposed that the generation of ELIP could be attributed to melting of a mantle plume head because of the high mantle potential temperature (~1600 °C), which was estimated from high-Mg olivine phenocrysts. In addition, the regional lithospheric uplift immediately preceding flood volcanism also supports a plume-head origin (Xu et al. 2004). As discussed above, native gold, native copper, native zinc, and moissanite were not formed by either hydrothermal processes or by precipitation from a melt. Therefore, we argue that these native elements and the moissanite are xenocrysts derived from a deep-mantle source and, given the geological context of the ELIP, most likely a mantle plume. In addition, previous studies have shown that the Au contents of native gold increase with increasing temperature and pressure (Chisholm 1979 and references therein). Thus, the high purity of the native gold bleb in the picrite sample suggests that this gold formed under extremely high temperature. Consequently, the native gold, native copper, native zinc, and moissanite could represent phases left in the mantle during early stages of differentiation of Earth. They would have been transported to relatively shallow depths by a rising mantle plume, and then incorporated into the melts from which the picrites formed.

Concluding remarks and geological significances

The presence of some native elements and moissanite suggests that they were derived from a deep-mantle source under reducing conditions. The mantle plume, from which the ELIP was formed, is a good candidate because the plume is very hot, and the native gold bleb does not contain any other elements. Either these reduced phases formed earlier than the olivine phenocrysts or some special conditions allowed them to be preserved.

Although the Emeishan flood basalts themselves do not hold any economic reserves of gold, there are many gold deposits that formed during the Mesozoic time in the ELIP (e.g., Hu et al. 2004). On the basis of S and Pb isotopic data, some researchers proposed that the gold in these deposits may be derived from the mantle (Hu et al. 2005; Zhou 1996). As the S and Pb isotopic compositions of sulfides only indicate the sources of S and Pb, not Au, the source region of Au is still debated (Feng and Zhang 2004). The discovery of a native gold bleb enclosed in an olivine phenocryst provides direct evidence of a mantle source, or mantle plume sources for these gold deposits.

Ali et al. (2004) used Ar-Ar dating to define chronologically the post-Permian tectonothermal events in the Indo-China-western Yangtz region, affected by the Emeishan flood volcanism. These events cluster at ca. 175, 142, 98, and 42 Ma, which Ali et al. (2004) ascribed to strike-slip and collision tectonic movements along the complex major sutures that separate the Indian block in the west from Indo-China and the South China blocks to the east and Songpan-Garze basin to the northwest. In addition, the ASRR and adjacent structures were reactivated during the 55–50 Ma collision of the Indian plate with Eurasia. This reactivation of the ASSR is estimated at about 300 km of left-lateral strike-slip movement (Tapponier et al. 1990). We argue that the gold was incorporated into hydrothermal systems by convection of ore-fluids during these later thermal events.

Recently, several native copper deposits in the sedimentary interbeds of the Emeishan flood basalts and in amygdaloidal basaltic lavas (Keeweenawan type; Brown 1979) were discovered in the Ludian area, eastern Yunnan province, eastern part of the ELIP (Zhu et al. 2002), and in the border area between Yunnan and Guizhou Provinces (Zhu et al. 2003). The origin of such copper deposits (Zhu et al. 2002, 2003; Zhang et al. 2004a; Li et al. 2004) is hotly debated. The discovery of native copper grains in serpentinized olivine phenocrysts show that the mantle plume associated with ELIP may be a potential source of copper for these native copper deposits.

As stated above, moissanite (SiC) is a naturally occurring mineral closely associated with diamonds. Therefore, the discovery of diamonds in the Emeishan picrites is of great economic importance because it opens a new area for diamond exploration as exemplified by discoveries of diamonds in picritic rocks in the Kusya River Basin, Urals (Russia) (Lukyanova et al. 1980).

Acknowledgments

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