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Discovery of Cu-Zn, Cu-Sn intermetallic minerals and its significance for genesis of the Mianning-Dechang REE Metallogenic Belt, Sichuan Province, China

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Abstract Mianning-Dechang Himalayan REE Metallogenetic Belt in Sichuan Province lies along the western margin of the Yangtze Craton. We have conducted detailed mineralogical studies on ore minerals collected from Maoniuping and Dalucao, the two largest deposits in the belt. With optical microscope, SEM/EDS, and EPMA, three rare intermetallic minerals, *i.e.*, zinccopperite (Cu_2Zn), Sn-bearing native copper, and Cu-bearing jupiter were found to occur in the main ore along with barite, fluorite, apatite, sulfide and bastnaesite. Since the conditions under which zinccopperite and Sn-bearing native copper formed are quite unique, finding of these minerals, for the first time in domestic REE deposits, has significant implications for the genesis of the ore deposits in which they occur. In comparison with Cu-Zn intermetallic minerals in other occurrences, we propose that the formation of this REE metallogenetic belt is associated with fast upwelling of the Himalayan magma from deep source.

Keywords: Mianning-Dechang REE metallogenetic belt, Cu-bearing intermetallic minerals, EPMA, genesis significance, deep source magmatic activity.

The Mianning-Dechang REE metallogenetic belt is among the most important of its kind in China. The typical geologic features and multiple economically-valuable components in this belt have caught attention from more and more geologists. Wide investigations from petrology, petrochemistry to rare earth and trace element geochemistry, have been carried out recently, and a large quantity of geochemical data have already been obtained[1−11]. Maoniuping REE deposit is the largest of this belt, the third largest in the world behind Bayan Obo, China, and Mountain Pass, USA. Understandably, previous studies of ore geology, petrochemistry, isotope geochemistry, fluid inclusion, etc. have been heavily focused upon Maoniuping[1−2,7−15], which leads to lack of the thorough studies and poor understanding of other deposits in the same belt. Genesis and ore-forming mechanism of these deposits remain debatable. Mineral associations of Maoniuping and Dalucao REE deposits were carefully studied by Yuan Zhongxin *et al*. (1995)[2]; China University of

Geosciences (1998)[1)] and the 109th Geological Team of Sichuan Bureau of Mineral Exploration (1997)[2)], but none of them noticed the existence of some rare intermetallic minerals. In this study, we will report our discovery of three rare intermetallic minerals, namely zinccopperite, Sn-bearing copper and jupiter, made by using ore microscopy, SEM/EDS(Scanning Electron Microscope/Energy Dispersion) and EPMA (Electro Probe Micro-Analysis). The discovery is important to our understanding of the genesis and physico-chemical conditions of this type of REE deposits. Cu-bearing minerals themselves are also important ore-minerals.

1 Geological outline of Mianning-Dechang REE metallogenic belt

The Mianning-Dechang REE metallogenic belt (Panxi REE Metallogenic Belt), ~150 km long and ~10 km wide, striking NNE, in Sichuan extends from Mianning, through Xichang to Dechang. Five REE deposits (mineralization spots) were found in this belt. They are Maoniuping, Muluo, Maidi, Lizhuang, and Dalucao. Besides REE, Pb, Mo, Ag, Th and barite, celestite, fluorite are also mined.

Tectonically, the Mianning-Dechang REE metallogenic belt is located in the mid-north of the Panxi rift of the Kangdian Anteclise, the Yangtz Craton. Extrusive and intrusive rocks that are exposed along this belt are alkali-rich ultramafic, mafic, and intermediate-felsic. REE-mineralized intrusive rocks in the rift usually occur as supper-hypabyssal stock on juncture between pre-sinian diastrophic block and Mesozoic marine strata in the west of An'ninghe fault. From north to south, intrusive REE-rich bodies are the following: Maoniuping nordmarkite, Muluo alkali granite, Lizhuang nordmarkite, Xichang-Taihe alkali granite and nordmarkite, Cida alkali granite in Dechang, Dalucao nordmarkite[2]. Ore bodies in the Maoniuping deposit occur mainly in nordmarkite and the rest in rhyolite. Ore minerals of the Maoniuping REE deposit include bastnaesite, bunsite and cerianite; gangue minerals include barite, fluorite, quartz and calcite. Minor metallic sulfides and oxide minerals are also present. Varied ore types are distinguished as calcite type with mass-mottled structure REE ore; fluorite, barite porous REE ore, veinlet and mixed ore[2]. Ore bodies in Dalucao REE deposit occur as big lentiforms, veins and veinlets. No.1 ore body is a breccia type composed of fluorite, barite-celestite and REE minerals. Bastnaesite is the dominating REE mineral. Fluorite, celestite-barite, calcite and minor metallic sulfide (galena, pyrite, chalcopyrite) and metallic oxide (hematite, magnetite, ilmenite, unidentified Fe-Mn oxides) are the gangue minerals.

2 Occurrence, microscopic characteristics and chemical composition of Cu-bearing intermetallic minerals

In ore samples from Maoniuping and Dalucao, three intermetallic minerals, including zinccopperite, Sn-bearing copper and jupiter, were first identified with the optical microscope and then further characterized by SEM/EDS and EPMA. SEM/EDS was performed in the State Key Laboratory for Advanced Metals and Materials at the University of Science and Technology Beijing. The SEM instrument is a Cambridge S250-MK2 with resolution of 6 nm, accelerating voltage of 30 kV, most amplification of 3×10^5. The EDS instrument is a LINK860 with working voltage of 1–30 kV, least beam spot of 5 nm and analytical diameter of 1—10 μm. The detection limit is 0.1%—0.3% for elements after Na in the periodic table. The EPMA Lab of the Institute of Mineral Resource, Chinese Academy of Geological Sciences helped with electron microprobe analysis. The instrument is a JXA8800-R electron probe with electric current set at 2×10^{-8}A and accelerating voltage 20 kV. Intermetallic minerals occur in irregular shape in the cement of breccia ore accompanied by barite (celestite), fluorite, bastnasite, magnetite and sulfide; some by limonite in the zone of oxidation.

(i) Zinccopperite. Gold looking under reflecting microscope with golden yellow color, irregular shapes, 4—140 μm in diameter (Figs. 1(a), 2(a)). The reflectivity is higher than that of pyrite. Nick or groove oc-

1) China University of Geosciences (Wuhan), Ore-forming conditions of DL REE deposit in Dechang, Sichuan Province, 1998
2) The 109th Geological Team of Sichuan Bureau of Mineral Exploration, REE occurrence and production utilization of Dalucao REE deposit in Dechang, Sichuan Province, 1997

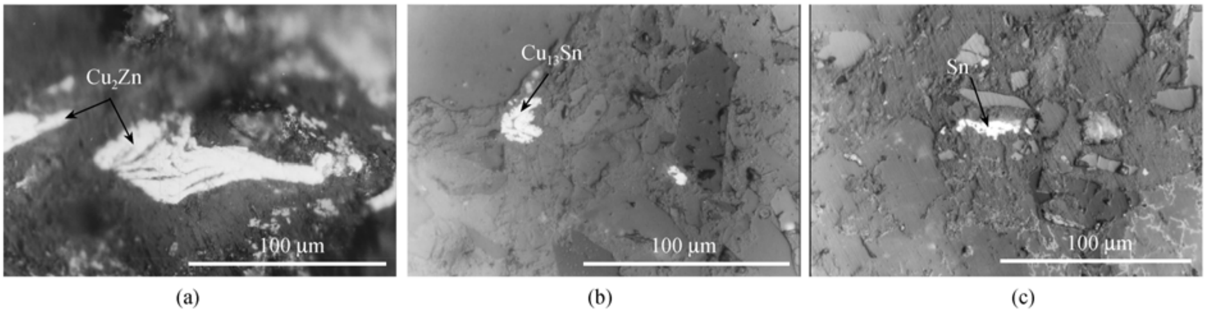

Fig. 1. Photos of zinccopperite, Sn-bearing copper and jupiter under microscope. (a) Zinccopperite (sample: mnp-37); (b) Sn-bearing copper (sample No. mnp-33); (c) jupiter (sample No. dl-04).

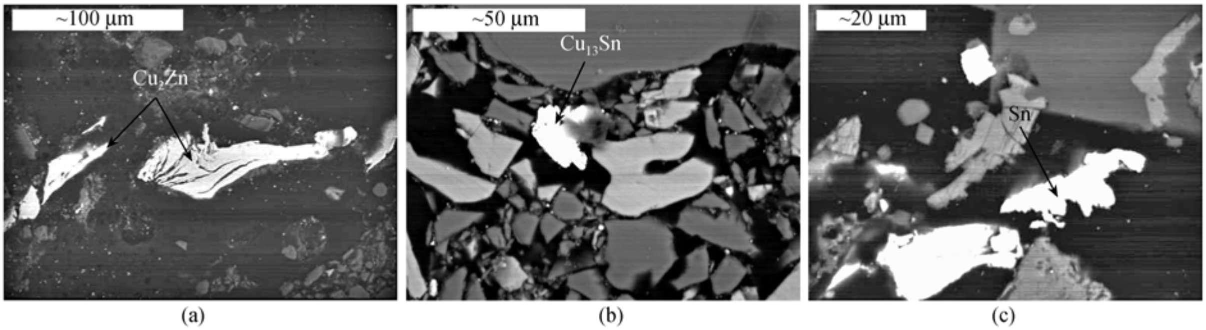

Fig. 2. Backscattered electron images of zinccopperite, Sn-bearing copper and Jupiter. (a) zinccopperite (sample: mnp-37); (b) Sn-bearing copper (sample:mnp-33); (c) jupiter (sample: dl-04).

Table 1 EPMA results of Cu-bearing intermetallic minerals (wt%)[a)]

Sample	Mg	Al	Si	P	K	Ca	Ti	Cr	Mn	Fe	Ni	Cu	Zn	Sn	Total	Fomula
mnp-33-2	0.12	0.00	0.20	0.00	0.00	0.01	0.06	0.01	0.04	0.32	0.05	83.80	2.67	11.57	98.85	$Cu_{13}Sn$
mnp-33-3	0.05	0.00	0.24	0.00	0.00	0.00	0.00	0.00	0.03	2.22	0.05	82.79	2.82	10.82	99.02	$Cu_{13}Sn$
mnp-37-1	0.15	0.00	0.15	0.00	0.00	0.04	0.00	0.00	0.00	0.68	0.08	67.33	31.03	0.12	99.58	Cu_2Zn
mnp-37-2	0.26	0.00	0.12	0.02	0.01	0.00	0.02	0.00	0.01	0.86	0.03	67.37	30.41	0.17	99.28	Cu_2Zn
dl-08-1	0.00	0.00	0.13	0.00	0.00	0.01	0.00	0.02	0.05	0.12	0.57	84.56	1.97	11.47	98.90	$Cu_{13}Sn$
dl-04-1	0.08	0.00	0.26	0.01	0.00	0.00	0.00	0.00	0.06	0.39	0.16	1.01	0.09	97.05	99.11	Sn
dl-01-1	0.18	0.00	0.08	0.00	0.00	0.00	0.00	0.00	0.03	0.91	0.10	67.06	31.11	0.09	99.56	Cu_2Zn

a) Performed by Zhou Jianxong and Chen Zhenyu in EPMA Lab of Institute of Mineral Resource, CAGS.

curs on the surface of a grain. Zinccopperite are found in the polished sections from both Maoniuping and Dalucao. In Maoniuping REE ore, Zinccopperite occurs in breccia barite vein developed in the fracture of euhedral quartz. Cement barite is porous and can be easily damaged during sample preparation. Bastnaesite, magnetite, fluorite and sulfide are present in barite vein as well. In Dalucao REE ore, Zinccopperite also occurs in cement of breccia ore. Energy spectrums show (Fig. 3(a)) Cu, Zn and Fe peaks. The quantitative analyses by EPMA yield the chemical composition as follows: Cu 67.06－67.37 wt%, Zn 30.41－31.11 wt%, and Fe 0.68－0.91 wt%. This leads to a simplified formula of Cu_2Zn，which falls in α portion in the Cu-Zn alloy phase diagram. Zinccopperite has been reported from other different occurrences, such as in silicification belt of Yulong and Malasongduo porphyry copper deposits, Tibet[16], in Ruo'ergai gold deposit, Sichuan Province[17], Shanchakou Cu-bearing intrusion of Xinjiang[18] and in Tuolugou SEDEX type cobalt deposit[19]. The similarity in its chemical composition of zinccopperite from a variety of places strongly suggests that this intermetallic mineral is quite stable in nature.

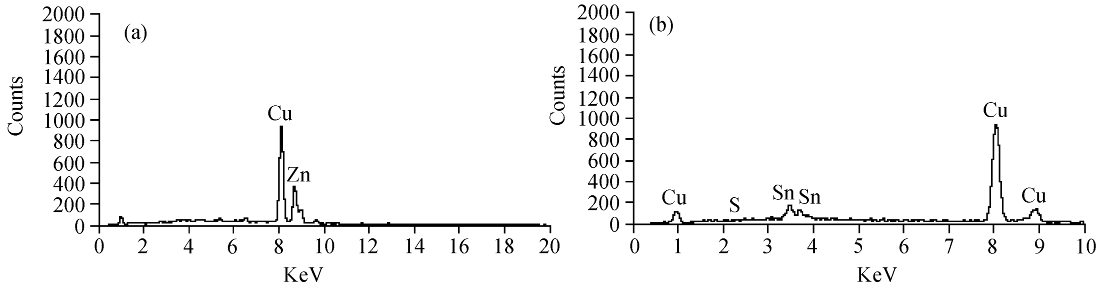

Fig. 3. Energy spectra of zinccopperite and Sn-bearing copper. (a) Zinccopperite (sample No. mnp-37); (b) Sn-bearing copper (sample No. mnp-33).

(ii) Sn-bearing copper. Similar to zinccopperite under a reflecting microscope with golden yellow color, albeit being slight darker than zinccopperite, irregular shapes, and sizes varied from <5 to 30 μm (Figs. 1(b), 2(b)). Like zinccopperite, the reflectivity is higher than that of pyrite. Minerals of Sn-bearing copper are found in the ore from both Maoniuping and Dalucao. Chemically, it consists of 82.79—83.80 wt% of Cu, 10.82—11.57 wt% of Sn and 1.97—2.82 wt% of Zn, determined by EPMA . The peaks in the energy spec- trum are shown in Fig. 3(b). This composition is rep- resented by formula $Cu_{13}Sn$. The occurrence of Sn- bearing copper in Maoniuping is identical to that of zinccopperite described above. Similar native Sn-bearing copper minerals were previously reported by Wang (1999)[20] and Zhang *et al*. (2002)[19].

(iii) Jupiter. Bright white color under a reflecting microscope, in irregular shapes, ~30 μm in size (Figs. 1(c), 2(c)). The reflectivity is also higher than that of pyrite. So far, they are only seen in samples from Dalucao. It occurs in cement of breccia ore companied by barite, celestite, magnetite and minor sulfides. Chemical composition determined by EPMA shows 97.05wt% of Sn and only 1.01wt% of Cu, and therefore the chemical formula is simply Sn. Juipiter is found in many occurrences, which differs from native copper minerals. Because allomerism between Cu and Sn is not so easy to happen, jupiter is usually characterized by lower content of Cu (<1%), although jupiter in Dalucao has relatively higher Cu content compared with others.

3 Summary of research on native Cu-bearing intermetallic minerals

Unlike copper which is commonly seen in many ore deposits, Cu-Zn intermetallic minerals have rare occurrences. Copper and zinc intermetallic minerals were first found in lunar rocks (Apollo-11)[21] and later found in several other different occurrences including in meteorolites. Zhanghengite (Cu_3Zn) was found in a meteorolite from Boxian, Anhui Province[22] and Danbaite ($CuZn_2$) in a Cu-Ni sulfide deposit within sheet-like ultrabasic rocks in Danba, Sichuan Province[23]. Cu-Zn intermetallic minerals are seen in many occurrences, including Xifanping porphyry copper deposit ($Cu_{6\text{-}7}Zn$), Sichuan Province[24], Ruo'ergai gold deposit (Cu_2Zn), Sichuan Province[17], Yulong and Malasongduo porphyry copper deposit (Cu_2Zn) , Tibet[16], Sanchakou copper deposit (Cu_2Zn), Xinjiang[18], Kalatongke Cu-Ni sulfide deposit (Cu_3Zn), Xinjiang[25], Tuolugou SEDEX type cobalt deposit (Cu_2Zn)[19]. Most of zinccopperite minerals fall in α portion in Cu-Zn alloy phase diagram, but Danbanite and Zhanghengnite are in γ and β portion respectively. Danbanite and Zhanghengnite are the formal names for these two minerals while others have been called differently in publications. Cu_2Zn has been called "copper-zinc"[17], "Zn-copper"[19] and zinccopperite[24,26,27], *etc*. Sun Yan *et al*. (2003)[27] called Cu_3Zn, $Cu_{6\text{-}7}Zn$ and Cu_2Zn zinccopperite which is what we adopt in the paper. Internationally, studies of Cu-Zn intermetallic minerals are scarce. Distlera *et al*. (2004)[28] reported a Cu-Zn intermetallic mineral in Sukhoi Log gold deposit occurring as quartz vein in black shale, but no detailed description was given. Cu-Zn intermetallic mineral in REE deposit has not been seen in any report worldwide. The similarity in its chemical composition of zinccopperite from a variety of places strongly suggests that this intermetallic mineral is quite stable in nature.

Copper and jupiter are common native metal minerals but Sn-bearing copper or Cu-bearing jupiter rarely occurs in nature. As stated earlier, Cu and Sn are not proximal elements, so, it is difficult for them to form an intermetallic mineral by allomerism. Thus far, reports on Sn-bearing copper are very limited. Besides Mianning-Dechang REE belt, similar minerals are only seen in volcanic tuff in Changkeng-Fuwan polymetal ore field, Guangdong Province[20] and in Tuolugou SEDEX cobalt deposit, East Kunlun[19]. The physico-chemical conditions of forming Sn-bearing copper remain unknown and need to be further studied.

4 Significance of the discovery of Cu-bearing intermetallic minerals

Except for lunar rocks and meteorolites, almost all occurrences of Cu-Zn alloy minerals are spatially related to the suture between two plates, which indicates that mineralization is associated with deep source magmatism derived by subduction and collision processes of plates. Yulong and Malasongduo porphyry copper deposits, for example, are located in India-Asia collision zone; other examples include Kalatongke Cu-Ni sulfide deposit in North Jungar folded zone between Siberia and Kazakstan-Jungar plates[29], Sanchakou copper deposit of Xinjiang in the joint between Tarim and Kazakstan-Jungar plates[30], Ruo'ergai Gold deposit of Sichuan in the northern margin of the Yangtze Craton[17], both Danba Pt-bearing Cu-Ni sulfide deposit[23] and Mianning-Dechang REE metallogenic belt in the western margin of Yangtze Craton. In summary, although Cu-Zn alloy minerals occur in a variety of rocks and ore types and spread out over geologic times, they all seem to be present only in very few but similar tectonic settings that are dictated by plate movements and associated basic to ultrabasic magmatic activities originated from deep sources.

Jupiter and copper are frequently seen in nature. Jupiter was first found in sand deposit, uranium deposit, cupreous sandstone, and volcanic pneumatolytic minerals. Recently, it became known that jupiter is also present in ophiolite mantle[31], kimberlite[32−34] basic-ultrabasic rocks. A large number of literatures on native metal or intermetallic minerals in ophiolite have been published lately. Native metal minerals including native iron, copper, chromium, jupiter, lead, nickel, zinc, silicon, bismuth and silver were reported to be found in ultrabasic rocks and chromite deposit in Far-east Koryak Plateau. Native gold, copper, chromium, silver, iron, palladium, osmium, iridium, *etc*. were found in chromium deposit in ophiolite mantle in Ural area. PGE intermetallic minerals and several native base metal minerals were also found in chromium deposit in Kompirsai ophiolite[31]. Gold-platinum intermetallic mineral was found in mantle xenolith[35].

The occurrences of native metal minerals in ophiolite mantle, mantle plume basalt (Omei basalt), and mantle xenolith indicate that mantle is rich in native metal and intermetallic minerals, especially rare native metal minerals. It is even suggested that the presence of abundant native metals is one of the exclusives of the mantle. Studies on native metal and intermetallic minerals in lunar rocks and meteorolites also indicate the similarities in composition and conditions with the deep earth. Native metal minerals that occur near the surface are almost certainly brought up by magma from deep source (mantle). Bai Wenji *et al*. (2004)[31] believe that native metal minerals form in a strongly reductive environment. The native metals in mantle are likely core materials that are left in mantle during core-forming process and later are brought up to the upper part of the earth by mantle plume. Since copper and zinc are adjacent elements in the same period in the periodic table, they have similar atomic radius, atomic weight and chemical features. It is, therefore, easy for them to form alloy. But, due to their sulphophile and lithophile nature, they tend to form either sulfide and oxide if sulfur or oxygen is available. This leads Luo *et al*. (1999)[17] to conclude that the occurrence of native and intermetallic mineral of Cu and Zn must indicate a geologic setting that is strongly reductive, S and O absent, and at a high temperature. The presence of abundant sulfate in Mianning-Dechang REE metallogenic belt, however, does not suggest the absence of sulfur. Our alternative explanation for intermetallic minerals in Mianning-Dechang is that forming of Cu-Zn native minerals is also dictated by the duration of the rise of magma. We believe that fast upwelling and cooling of magma makes combination reaction of S and Cu/Zn difficult as the duration in which the condition of the environment is suitable for the reaction is not long enough. As a result, native copper/zinc and intermetallic minerals will quickly form instead. Cn-Zn intermetallic minerals in Mianning-Dechang generally occur in the cement of breccia rocks with the cement mainly consisting of cellular and skeletal barite. This is obviously due to fast cooling and crystallization of a magma with high volatile components. Comparing it with similar occurrence of intermetallic minerals in vocalnic pneumatogenic phases, tuff, and SEDEX cobalt, we can conclude that the Mianning-Dechang REE metallogenic belt is associated with a quick magmatic process that started from a deep source (mantle). Pb, Sr, and Nd isotopic signatures have clearly shown that carbonatite is derived from the EMI mantle source[14]. Studies of fluid inclusions also support the point of view that the mantle served as the source of magma [6,11,15].

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