

Ore Genesis of the Kalatongke Cu–Ni Sulfide Deposits, Western China: Constraints from Volatile Chemical and Carbon Isotopic Compositions

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Abstract: The Kalatongke Cu–Ni sulfide deposits located in the East Junggar terrane, northern Xinjiang, western China are the largest magmatic sulfide deposits in the Central Asian Orogenic Belt (CAOB). The chemical and carbon isotopic compositions of the volatiles trapped in olivine, pyroxene and sulfide mineral separates were analyzed by vacuum stepwise-heating mass spectrometry. The results show that the released volatiles are concentrated at three temperature intervals of 200–400°C, 400–900°C and 900–1200°C. The released volatiles from silicate mineral separates at 400–900°C and 900–1200°C have similar chemical and carbon isotopic compositions, which are mainly composed of H₂O (av. ~92 mol%) with minor H₂, CO₂, H₂S and SO₂, and they are likely associated with the ore-forming magmatic volatiles. Light $\delta^{13}\text{C}_{\text{CO}_2}$ values (from –20.86‰ to –12.85‰) of pyroxene indicate crustal contamination occurred prior to or synchronous with pyroxene crystallization of mantle-derived ore-forming magma. The elevated contents of H₂ and H₂O in the olivine and pyroxene suggest a deep mantle-originated ore-forming volatile mixed with aqueous volatiles from recycled subducted slab. High contents of CO₂ in the ore-forming magma volatiles led to an increase in oxygen fugacity, and thereby reduced the solubility of sulfur in the magma, then triggered sulfur saturation followed by sulfide melt segregation; CO₂ contents correlated with Cu contents in the whole rocks suggest that a supercritical state of CO₂ in the ore-forming magma system under high temperature and pressure conditions might play a key role in the assemblage of huge Cu and Ni elements. The volatiles released from constituent minerals of intrusion 1[#] have more CO₂ and SO₂ oxidized gases, higher CO₂/CH₄ and SO₂/H₂S ratios and lighter $\delta^{13}\text{C}_{\text{CO}_2}$ than those of intrusions 2[#] and 3[#]. This combination suggests that the higher oxidation state of the volatiles in intrusion 1[#] than intrusions 2[#] and 3[#], which could be one of key ore-forming factors for large amounts of ores and high contents of Cu and Ni in intrusion 1[#]. The volatiles released at 200–400°C are dominated by H₂O with minor CO₂, N₂+CO and SO₂, with $\delta^{13}\text{C}_{\text{CO}_2}$ values (–25.66‰ to –22.98‰) within the crustal ranges, and are considered to be related to secondary tectonic–hydrothermal activities.

Key words: Ore genesis, chemical composition, carbon isotope, volatile, mafic–ultramafic intrusion, Kalatongke Cu–Ni sulfide deposits, Xinjiang

1 Introduction

The magmatic Cu–Ni sulfide deposit is an important metallogenic type of copper–nickel resources. The Kalatongke magmatic Cu–Ni sulfide deposits are the largest of several large-scale magmatic sulfide deposits

occurring in the Central Asian Orogenic Belt (CAOB) (Song and Li, 2009; Zhang et al., 2009; Zhang et al., 2011; Li et al., 2012). The orogenic belt tectonic setting of this deposit formation is different from the cratons of other world-class magmatic deposits, such as the Noril'sk–Thalnak, Bushveld and Duluth Cu–Ni sulfide deposits (Arndt et al., 1998; Lightfoot and Keays, 2005; Ripley and

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Li, 2007). The zircon SHRIMP U-Pb dating from the norite in intrusion 1[#] has yielded the intrusive age of 287 ± 5 Ma (Han et al., 2004), Re-Os geochronological dates of the Cu-Ni sulfide ores are 282.5–290.2 Ma (Zhang et al., 2005), which is close to the time of mantle plume activity in Tarim to the south. The ore genesis and tectonic setting of the Kalatongke magmatic Cu-Ni sulfide deposits remain controversial.

Petrogeochemical data of the Kalatongke deposits have shown that sulfur saturation and sulfide segregation might be caused by olivine and pyroxene fractional crystallization or sulfur addition (Qian et al., 2009; Li et al., 2012), or crustal silicate addition (Zhang et al., 2003; Zhang et al., 2009). The mafic-ultramafic intrusions may be considered as either the roots or ascent and supply conduits of a large scale of magma related to a mantle plume (Mao et al., 2006, 2008; Pirajno et al., 2008; Qin et al., 2011), or as products of the subducted Junggar ocean plate (Han et al., 2007), the magmatic sulfide mineralization in a post-collisional environment (Zhang et al., 2008; Song and Li, 2009), or formed by decompression melting in the upwelling oceanic asthenosphere in a slab break-off window setting (Li et al., 2012).

The formation of a large scale Cu-Ni sulfide deposit needs the assemblage of huge amounts of Cu and Ni. The CO₂ in a supercritical state of mantle-derived magma system can leach and carry large amounts of Cu and Ni metal elements (Lowenstern et al., 1991; Fleet and Wu,

1995), which may be the critical factor for accumulation of huge ore-forming metal elements. On the other hand, CO₂ addition into an ore-forming magma will elevate the oxidation state of a magma system, and hence reduce the sulfur solubility of a silicate magma, which results in sulfide saturation and segregation (Lehmann et al., 2007).

The volatiles in the minerals of a magmatic Cu-Ni sulfide deposit contain the volatiles equilibrated with high temperature magma and volatiles related to secondary tectonic-hydrothermal activities. The chemical and carbon isotopic compositions of the volatiles may reveal the magma source and evolution (Trull et al., 1993; Demeny et al., 2004; Shaw et al., 2004) and provide clues to ore genesis and tectonic settings (Ripley et al., 2005; Zhang et al., 2010; Feng et al., 2011). In the present study, the chemical compositions and carbon isotopic compositions of volatiles in the olivine, pyroxene and sulfide in the Kalatongke deposits have been determined by vacuum stepwise-heating mass spectrometry combined with major and trace elements and Sr-Nd isotopic data (Jia et al., 2009; Li et al., 2012) to reveal the ore-forming magmatic source, evolution and tectonic setting.

2 Geological Background

The Kalatongke Cu-Ni sulfide deposits are located in the East Junggar terrane, the southern part of the CAOB, which is situated between the Siberian and North China-

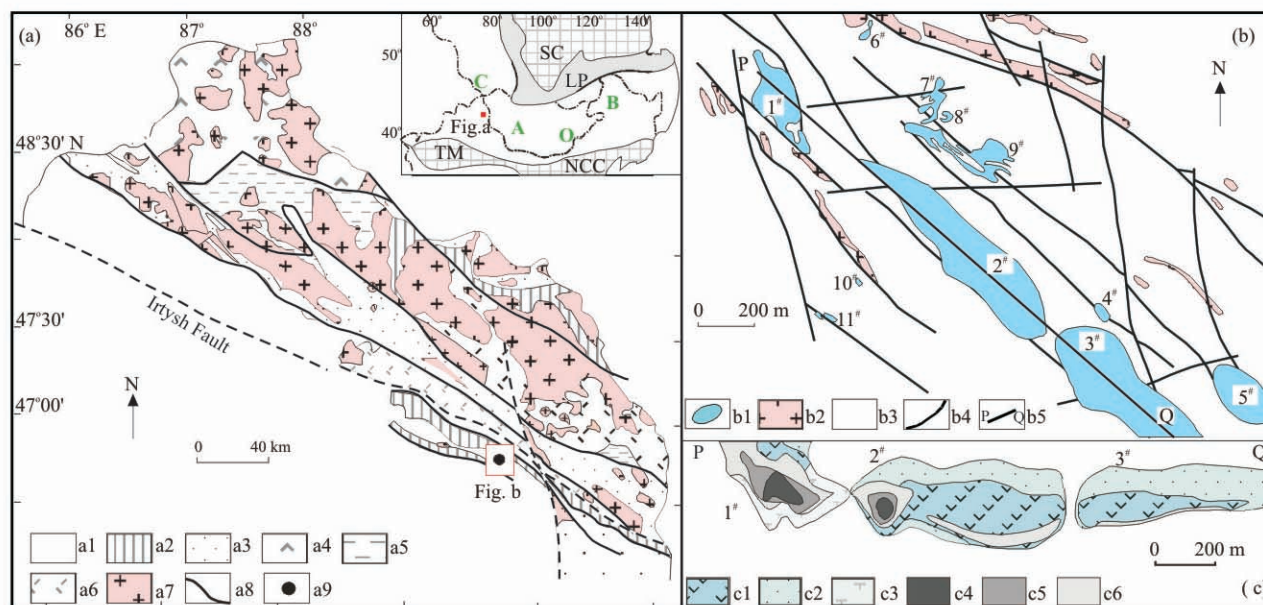


Fig. 1. (a) Location of the study area in the Central Asian Orogenic Belt and geological map of the southern Altay Range (modified from Jahn et al., 2000; Windley et al., 2002); (b) Simplified geological map (modified from Song and Li, 2009); and (c) Sections of the Kalatongke deposits (modified from Wang and Zhao, 1991).

a1, post-Triassic sedimentary rocks; a2, Carboniferous; a3, Devonian; a4, Cambrian and Ordovician; a5, Silurian; a6, schist and gneiss; a7, gabbro; a8, fault; a9, copper-nickel deposit; b1, ultramafic-mafic intrusion; b2, granophyre; b3, Carboniferous slate and tuff; b4, fault; b5, section; c1, norite; c2, diorite; c3, diabase; c4, massive sulfide; c5, net-textured sulfide; c6, disseminated sulfide. CAOB, Central Asian Orogenic Belt; NCC, North China Craton; SC, Siberian Craton; TM, Tarim; LP, Late Proterozoic.

Tarim cratons and extends more than 5000 km from west to east (Sengör et al., 1993; Fig. 1a). The details of regional geology were described by Li et al. (2012). Eleven ore-bearing or ore-barren mafic–ultramafic intrusions in the Kalatongke deposits are well differentiated, compositionally zoned, composed of hornblende-dolerite, olivine-norite, norite and diorite, and emplaced into Lower Carboniferous tuffs and shales at around 330 Ma (Mao et al., 2008; Fig. 1b). The sulfide ore bodies are hosted in intrusions 1[#], 2[#], 3[#] and 9[#], where they occur at the bases.

Intrusion 1[#] is characterized by the smallest volume with the largest Cu–Ni sulfide mineralization among four ore-bearing intrusions, and its set of lithologies consist of olivine-norite, norite and diorite from base to top (Fig. 1c). The zoned sulfide ore bodies are overlain by olivine-norite. Some disseminated sulfides occur farther up in the olivine-norite unit and in the lower part of the norite unit. Both unexposed intrusions 2[#] and 3[#] show tabular shapes, and are characterized by horizontal lithological zoning consisting of norite and diorite from base to top. The disseminated sulfide ores occur in the lower part of the norite unit (Song and Li, 2009). The primary base metal sulfides include pyrrhotite, chalcopyrite and pentlandite, with lesser pyrite, violarite, magnetite, Ag–Au minerals and telluride.

3 Sample Preparation and Analysis Process

3.1 Samples

The 48 samples used in this study were collected systematically from two recently drilled bore cores in intrusion 3[#] and underground adits in intrusions 1[#] and 2[#]. Based on petrological observations and the data of major elements, trace elements and platinum group elements (PGE) and the Sr–Nd isotopes of whole-rock and mineral separate samples (Jia et al., 2009; Li et al., 2012), we chose fresh olivine-norite, norite and sulfide ore for the selection of olivine, pyroxene and sulfide mineral separates. The petrological characteristics and microphotos of rock samples studied have been described by Li et al. (2012).

3.2 Sample pretreatments

Pieces of weathered surfaces and altered parts of samples were removed and washed carefully in distilled water. The fresh interiors of samples were crushed to a size range of 0.4–0.25 mm, then olivine, pyroxene and sulfide mineral separates were hand-picked under a binocular to ensure that they were free of petrological alteration.

All the samples were ultrasonically cleaned with

0.3mol/L HCl, rinsed with distilled water until the pH value reached around 7, and then ultrasonically cleaned again with analytical grade dichloromethane (CH₂Cl₂) before being dried at 110°C.

3.3 Experimental procedure

The volatiles in the silicate and sulfide mineral separates have been determined by vacuum stepwise-heating mass spectrometry. This technique can isolate volatiles stored in different residence sites by setting the heating temperatures of each step, and ensure excellent analytical reproducibility of mass spectrometry without detectable products of high temperature reaction among the released volatiles (Ivankin et al., 1988; Javoy and Pineau, 1991; Zhang et al., 2007). Each sample was loaded into a quartz tube and preheated at 110°C, and out-gassed for 2 hours under an high vacuum (1×10^{-8} Pa) to ensure no detectable volatiles possibly absorbed on sample released prior to analysis. Volatiles in mineral separates were extracted by stepwise heating from 200°C to 1200°C with 100°C increments for 50 minutes duration at top temperature of each step. The chemical composition of volatiles was analyzed by a MAT-271 mass spectrometer. For more on the volatile extraction line and experimental procedures see Zhang et al. (2007).

Carbon isotopes of CO₂ and CH₄, which can be expressed as $\delta^{13}\text{C} (\text{‰}) = ([^{13}\text{C}/^{12}\text{C}]_{\text{sample}}/[^{13}\text{C}/^{12}\text{C}]_{\text{PDB}} - 1) \times 10^3$ (where PDB, Pee Dee Belemnite, is the reference standard), were analyzed by a Gas Chromatography-Combustion-Mass spectrometer (GC-C-MS) system. Each sample was loaded into the quartz tube, and degassed at 110°C for 120 minutes by helium carrier gas to remove potentially surface-adsorbed gases and air in the sample and extraction line. CO₂ and CH₄ in sample were extracted at 200–400°C, 400–900°C and 900–1200°C by stepwise-heating extraction procedure based on the release character of the volatiles (see section “4 Result” for details), and heated for 50 minutes at top temperature of each step. The volatiles released at each step were introduced into the GC-C-MS system by helium carrier gas. The CO₂ and CH₄ were separated by an Agilent 6890N gas chromatographic analyzer, and then oxidized into CO₂, and finally sent into a Delta plus XP mass spectrometer for carbon isotopic analysis. The reported $\delta^{13}\text{C}$ (relative to PDB) values have a relative error less than 1.6‰. For details of the extraction methods of CO₂ and CH₄ see Zhang et al. (2007).

4 Results

Volatiles from constituent mineral separates of the Kalatongke magmatic Cu–Ni sulfide deposits were

released in pulses with elevated temperatures, and concentrated at three temperature intervals of 200–400°C, 400–900°C and 900–1200°C, which could be derived from different residence sites in minerals with distinct sources (Zhang et al., 2007, 2009; Tang et al., 2012). The contents of each volatile speciation at the three temperature intervals are summed respectively; the chemical and carbon isotopic compositions of volatiles released are listed in Table 1.

The characteristics of volatile contents in the Kalatongke Cu–Ni sulfide deposits are shown in Fig. 2. The contents of volatiles from olivine and pyroxene separates are 3109 and 18847 mm³.STP/g on average (STP=standard temperature and pressure), respectively, which are much higher than those in the Siberian large igneous province (Tang et al., 2011). The volatiles at 400–900°C interval are the highest in the contents (Fig. 2a), and are dominated by H₂O (Fig. 2b). H₂O, H₂ and H₂S are mainly released at 400–900°C (Fig. 2b, 2c and 2e), whereas CO₂ and SO₂ are released in the temperature

interval of 600–1200°C (Fig. 2d and 2f); CH₄ is mainly released at 400–700°C and 900–1200°C (Table 1).

The volatiles from sulfide separates in the Kalatongke deposits are av. 3700 mm³/g in content, and released mainly at 400–1000°C except for SO₂. They are dominated by SO₂ with minor H₂S, CO₂ and CH₄. SO₂ and CH₄ are released at 400–1200°C (Fig. 3a). CO₂ is mainly released at the 400–1000°C interval with higher amounts at 500°C (Fig. 3b). H₂S, similar to CO₂, is released at 400–1000°C (see Table 1).

4.1 Chemical compositions of volatiles

The volatiles released from the Kalatongke deposits vary in chemical composition with mineral species. The volatiles in the silicate minerals are dominated by H₂O (av. ~91 mol%), with minor H₂ and CO₂ (2.6 and 2.4) and trace H₂S and SO₂, whereas volatiles in the sulfide are dominated by SO₂ (av. ~83 mol%) with minor H₂S (9), CO₂ (3), CH₄ (2) and H₂O (2).

The volatiles released in the three temperature intervals

Table 1 Volatile concentrations (mm³.STP/g) and carbon isotopes (vs. PDB‰) of CO₂ and CH₄ of olivine (Olv), pyroxene (Pyx), and sulfide (Sfd) separates in mafic–ultramafic intrusions of the Kalatongke deposits, Xinjiang, west China

No.	Rock type	Mineral	Temperature (°C)	Contents (mm ³ .STP/g)								$\delta^{13}\text{C}$ (‰)		
				H ₂	CH ₄	N ₂ +CO	C ₂ H ₆	CO ₂	H ₂ S	SO ₂	H ₂ O	CO ₂	CH ₄	
Intrusion 1 [#]														
KTL1-9	Olivine Norite	Olv	200–400	2.13	0.25	4.07	0.12	6.56	2.75	1.77	37.59			
			400–900	30.37	4.92	16.47	7.96	149.4	406.8	13.83	2223			
			900–1200	0.26	1.61	8.28	0.86	42.06	18.55	67.75	59.66			
KTL1-11	Norite	Pyx	200–400	1.61	0.34	5.02	0.47	22.47	0.21	19.70	59.31	–25.66	–41.96	
			400–900	430.7	1.87	65.30	5.86	1061	193.9	12.81	17128	–15.69	–31.49	
			900–1200	0.44	6.95	12.14	0.86	64.09	17.94	334.7	252.1	–12.85	–34.72	
KTL1-13	Norite	Pyx	200–400	1.94	0.06	6.57	0.01	10.96	0.00	0	63.65	–23.75	–45.32	
			400–900	303.50	2.55	25.82	0.94	64.92	163.0	0	7492	–18.54	–34.71	
			900–1200	0.11	2.38	15.25	0.74	77.11	23.41	109.9	767.4	–20.09	–38.63	
KTL1-14	Norite	Pyx	200–400									–24.28	–46.75	
			400–900										–20.86	–37.27
			900–1200										–20.52	–38.24
KTL1-7	Net-textured ore	Sfd	200–400	0.29	0.71	2.81	0.74	19.15	0.37	28.30	3.23			
			400–900	0.21	30.66	3.19	3.01	81.52	6.49	1725	18.32			
			900–1200	0.10	10.29	3.70	0.81	5.41	2.19	493.8	5.38			
KTL1-10	Massive ore	Sfd	200–400	0.04	0.63	5.40	1.05	21.78	0.37	21.16	5.56			
			400–900	0.29	68.71	7.52	3.23	48.75	72.26	3239	175.4			
			900–1200	0.20	110.6	7.65	0.37	8.62	15.97	5126	21.11			
Intrusion 2 [#]														
KTL2-3	Norite	Pyx	200–400	3.65	0.15	49.14	16.79	6.72	0.02	0.86	73.1			
			400–900	629.3	3.59	35.51	4.58	115.50	191.00	10.11	18608			
			900–1200	0.64	6.22	18.34	0.70	173.29	22.33	260.66	401.56			
KTL2-5	Norite	Pyx	200–400	2.77	0.16	81.94	6.84	6.80	0.01	0.41	68.47	–22.98	–42.97	
			400–900	348.5	5.94	36.29	13.17	235.50	473.1	8.92	16200	–19.02	-	
			900–1200	0.67	2.73	23.67	0.90	131.50	14.09	117.84	215.54	–13.19	-	
KTL2-7	Norite	Pyx	200–400	3.06	0.29	0.76	4.40	4.34	0	0.80	96.09	–24.07	–46.43	
			400–900	653.5	3.33	26.35	6.14	127.50	210.7	2.81	19075	–16.20	–39.64	
			900–1200	0.45	7.14	12.75	0.46	99.35	4.72	315	79.63	–14.88	-	
KTL2-8	Norite	Pyx	200–400	10.36	0.09	65.40	0.15	5.74	0.32	1.34	47.03			
			400–900	631.5	4.09	31.27	3.23	221.3	152.3	6.92	22577			
			900–1200	4.83	8.77	12.51	0.94	147.6	19.62	360.1	548.6			
Intrusion 3 [#]														
KTL3-14	Disseminated ore	Sfd	200–400	0.12	1.44	6.67	0.70	21.31	0.52	64.42	8.89			
			400–900	3.56	15.33	1.19	24.09	110.0	771.9	401.1	25.46			
			900–1200	0.09	5.65	7.65	1.71	13.36	52.17	236.6	11.14			
KTL3-16	Disseminated ore	Sfd	200–400	0.03	1.00	4.09	0.62	49.30	0.33	46.09	3.90			
			400–900	2.18	13.15	0.96	10.84	88.05	374.9	557.8	20.89			
			900–1200	0.13	7.57	3.89	1.52	9.88	36.11	341.4	7.98			

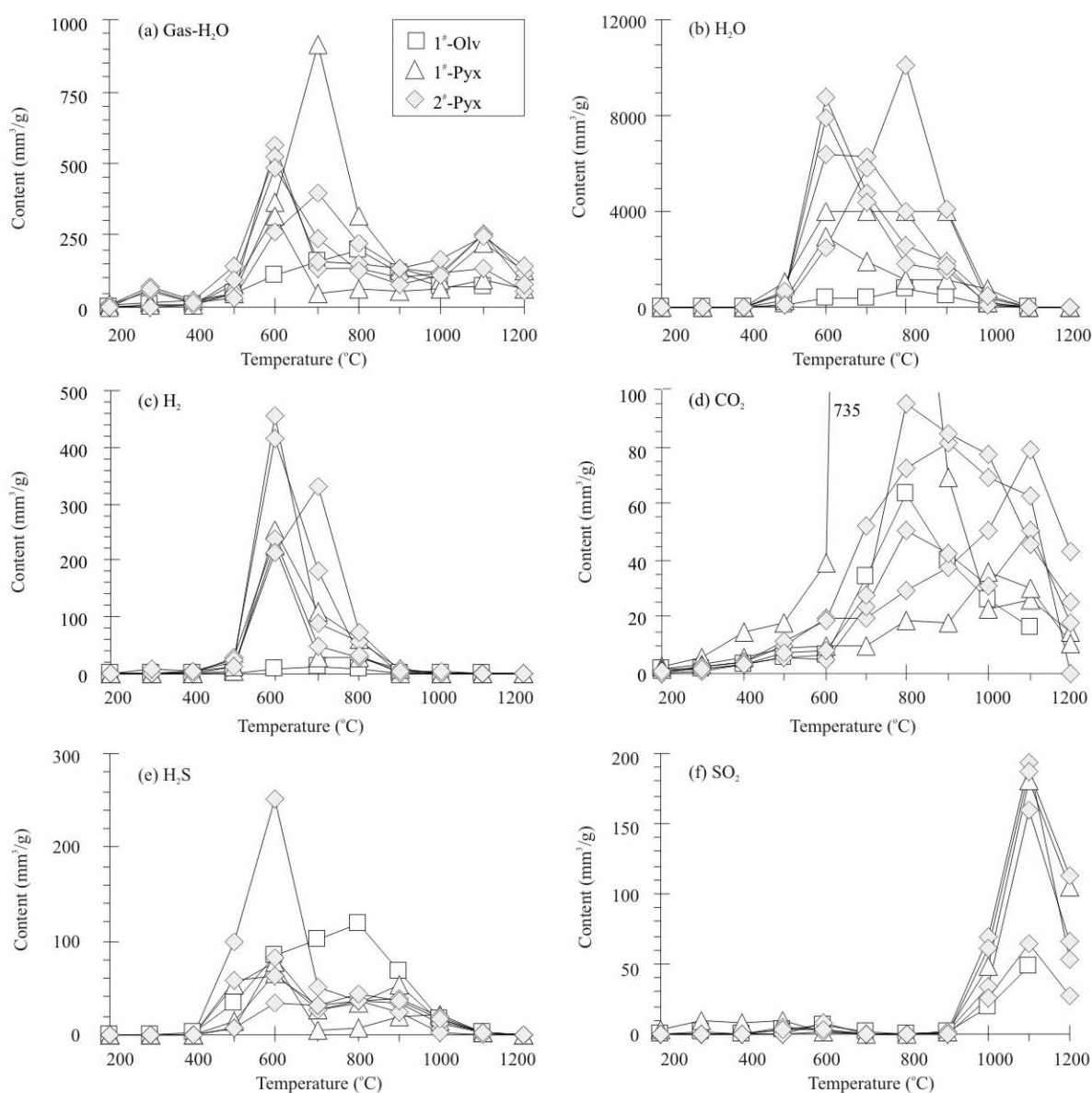


Fig. 2. Plots of volatile contents in the olivine (Olv) and pyroxene (Pyx) of the Kalatongke deposits vs. the temperature.

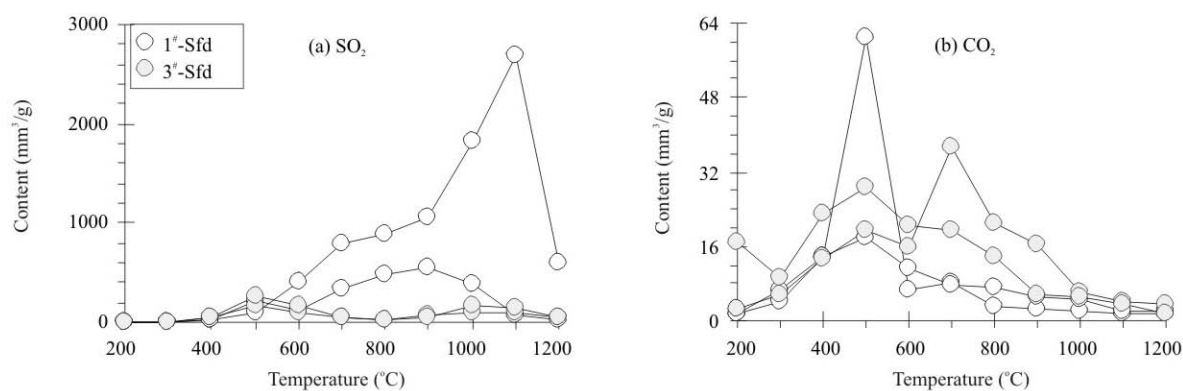


Fig. 3. Plots of volatile contents in the sulfide (Sfd) of the Kalatongke deposits vs. the temperature.

(200–400°C, 400–900°C and 900–1200°C) in the Kalatongke deposits show distinct chemical compositions. The volatiles at 400–900°C show the highest content and are

dominated by H₂O in olivine and pyroxene (av. 78 and 94 mol%, respectively) and by SO₂ in sulfides (av. ~75 mol%). When H₂O is excluded, the remaining components

of the volatiles in olivine are av. 630 mm³/g in content, and are dominated by H₂S (av. ~65 mol%) and CO₂ (24), with minor H₂ (5), N₂+CO (3) and SO₂ (2) and trace CH₄ and C₂H₆, whereas those in pyroxene are av. 1088 mm³/g in content, and are dominated by H₂ (av. ~46 mol%), CO₂ (28) and H₂S (21), with minor N₂+CO (3). The volatiles in the sulfides except for SO₂ are dominated by H₂S (av. ~61 mol %), with minor CO₂ (16) and H₂O (12), and trace CH₄ (6) and C₂H₆ (2).

The volatiles at 900–1200°C in the olivine and pyroxene separates in the Kalatongke deposits have higher content, and are dominated by SO₂ (av. ~34 and 32 mol%, respectively), H₂O (30 and 48) and CO₂ (21 and 15), with minor H₂S (9 and 2) and N₂+CO (4 and 2). Whereas the volatiles in the sulfide separates are dominated by SO₂ (av. ~95 mol %), the amount of the rest of the components are 89 mm³/g, which are dominated by CH₄ (av. ~38 mol%) and H₂S (30), with minor H₂O (13) and CO₂ (11).

The volatiles released at 200–400°C display the lowest content, and are dominated by H₂O in the olivine and pyroxene separates (av. 67 and 54 mol%, respectively) and by SO₂ in the sulfide separates (49 mol%). The amounts of volatiles excluding H₂O in the olivine and pyroxene separates are 19 mm³/g and 58 mm³/g, respectively, and are dominated by CO₂ (av. ~35 and 16 mol%) and N₂+CO (22 and 60), with minor H₂S, H₂, SO₂, CH₄ and C₂H₆. The volatiles excluding SO₂ in the sulfides are av. 41 mm³/g in content, and are dominated by CO₂ (av. ~68 mol%), with minor H₂O (13) and N₂+CO (12).

The chemical compositions of volatiles at 400–900°C interval vary distinctly among intrusions 1[#], 2[#] and 3[#]. The contents of CO₂ and SO₂, etc. oxidized gases of intrusion 1[#] are higher than those in intrusions 2[#] and 3[#]. The volatiles excluding H₂O in the pyroxene separates are dominated by CO₂ (av. ~48 mol%) and H₂ (31) in intrusion 1[#], and by H₂ (54) and H₂S (24) in intrusion 2[#]. The ratio of CO₂/CH₄ in intrusion 1[#] (av. 254) is much higher than that in intrusion 2[#] (41). The volatiles released from the sulfide separates in intrusion 1[#] have higher content of volatiles (av. 2744 mm³/g) and SO₂/H₂S ratios (av. 63) than intrusion 3[#] (1216 mm³/g and 1). The volatiles in intrusion 1[#] are dominated by SO₂ (av. ~91 mol %), whereas H₂S (av. ~47 mol%) and SO₂ (av. ~39 mol%) are dominated in intrusion 3[#].

4.2 Carbon isotopic compositions

The carbon isotopic values of CO₂ and CH₄ released from the constituent minerals of the Kalatongke deposits show differences at different temperature intervals (see Table 1 and Fig. 4a, b). The $\delta^{13}\text{C}$ values of CO₂ and CH₄ released at 400–900°C and 900–1200°C have similar ranges from –20.86‰ to –12.85‰ with a mean of

–17.18‰ for CO₂ and from –39.64‰ to –31.49‰ (av. –36.39‰) for CH₄. The $\delta^{13}\text{C}$ values of CO₂ and CH₄ at 200–400°C interval are noticeably lighter than those at 400–900°C and 900–1200°C intervals (Fig. 4), and $\delta^{13}\text{C}_{\text{CO}_2}$ values at 200–400°C vary from –25.66‰ to –22.98‰ (av. –24.15‰), within the range of the crustal endmember, and $\delta^{13}\text{C}_{\text{CH}_4}$ from –46.75‰ to –41.96‰ (av. –44.69‰). In addition, a positive correlation exists between $\delta^{13}\text{C}$ values of CO₂ and $\delta^{13}\text{C}$ values of CH₄ (Fig. 4b).

The carbon isotopic compositions of CO₂ and CH₄ at different temperature intervals vary distinctly between intrusions 1[#] and 2[#]. The $\delta^{13}\text{C}_{\text{CO}_2}$ values in intrusion 1[#] are lighter than those in intrusion 2[#]. The average values of $\delta^{13}\text{C}_{\text{CO}_2}$ for intrusions 1[#] are –17.82‰ at 900–1200°C, –18.36‰ at 400–900°C, and –24.56‰ at 200–400°C, while those for intrusions 2[#] are –14.03‰, –17.61‰ and –23.53‰ at the three corresponding temperature intervals, respectively.

5 Discussion

5.1 Sources of volatiles released

The chemical and carbon isotopic compositions of volatiles released from the mineral separates of the Kalatongke deposits exhibit differences at 200–400°C, 400–900°C and 900–1200°C intervals, implying that the volatiles are derived from different occurrence modes in minerals with distinct sources. The analogous release patterns of H₂O, H₂ and H₂S indicate their same residence modes in minerals. Two release peaks of CH₄ imply two kinds of occurrence modes or sources.

Volatiles released from mantle-derived magmatic minerals during stepwise heating could come from the following five possible sources: (1) air-derived volatiles absorbed on sample surfaces; (2) the decomposition of carbonates and sulfides at high temperatures; (3) new volatiles generated by high-temperature reactions among volatiles released; (4) volatiles trapped during mineral crystallization; and (5) volatiles trapped in any subsequent secondary processes, such as radiogenic ingrowths, metasomatism, metamorphism and alteration (Zhang et al., 2009).

Volatiles absorbed in sample surfaces, filling in cracks and broken fluid inclusions, and decompressed from carbonate minerals can be readily eliminated by our rigorous sample pretreatments and high-vacuum degassing prior to experimentation (Jackson et al., 1988). Because sulfides decompose at relatively high temperatures (610°C) under anhydrous conditions (Dunn, 1997), the large amounts of SO₂ (av. ~95 mol %) released at 900–1200°C from sulfide minerals might be related to the decomposition of sulfides. The new volatiles formed by

reactions among volatiles released during sample heating can be effectively prevented by adopting a cold trap with liquid nitrogen (Zhang et al., 2007, 2009; Tang et al., 2012) whereas those formed by volatile reactions in the sample interiors are regarded as a reversion of the volatiles equilibrated at the magma conditions of high temperature and pressure (Zhang et al., 2000, 2009; Tang et al., 2012). Therefore, volatiles released from the constituent minerals during stepwise heating are mainly derived from the volatiles trapped during mineral crystallization or in secondary processes.

Volatiles trapped from mantle-derived magma during mineral crystallization should be main volatiles preserved in minerals; they are stored in primary fluid inclusions and crystal lattices, structural defects or vacancies of minerals (Zhang et al., 2009, 2010; Tang et al., 2012). The volatiles released at 400–900 °C from silicate mineral separates in the Kalatongke deposits show the highest content and similar chemical and carbon isotopic compositions to those at 900–1200 °C; both are considered to be derived from primary fluid inclusions, crystal lattices, structural defects or vacancies of minerals, and represent the ore-forming magmatic volatiles. The volatiles released at 400–900 °C from sulfide separates are dominated by SO₂ (av. ~75 mol %), with minor H₂S (16) and trace CO₂ (4) and H₂O (3), and might represent the volatiles in sulfide solidification except for the abundant SO₂ which could be derived from the decomposition of the sulfide.

The volatiles at 200–400 °C from the silicate mineral separates are dominated by H₂O (av. ~55 mol%) and N₂+CO (26), with minor CO₂ (8), C₂H₆ (4) and SO₂ (3), and the average values of $\delta^{13}\text{C}_{\text{CO}_2}$ (–24.15 ‰) is in the range of the crust (Fig. 4); the volatiles released from sulfide separates are dominated by SO₂ (av. ~49 mol%) and CO₂ (34), with minor H₂O (7) and N₂+CO (6). The volatiles at 200–400 °C might be linked to secondary tectonic–hydrothermal activities; $\delta^{13}\text{C}$ values vary within the range of the crust, suggesting secondary metasomatism or alteration volatiles derived from the crust.

$\delta^{13}\text{C}_{\text{CO}_2}$ values are positively correlated with the $\delta^{13}\text{C}_{\text{CH}_4}$ values in intrusions 1[#] and 2[#] of the Kalatongke deposits, suggesting a possible equilibration of carbon isotope between CO₂ and CH₄. On the other hand, the $\delta^{13}\text{C}_{\text{CO}_2}$ values vs. the contents of CO₂ released at different temperatures does not correspond with the Rayleigh model of carbon isotope fractionation (Fig. 5). Therefore, the carbon isotopes of CO₂ and CH₄ could reveal the source and nature of volatiles in magma evolution.

5.2 The volatile system and sulfide saturation factors in ore-forming magma

Ore-forming magmatic volatiles released at 400–900 °C

and 900–1200 °C in the Kalatongke deposits are characterized by dominant H₂O (av. ~92 mol%) and minor H₂, CO₂, H₂S and SO₂, with trace CH₄ and C₂H₆. Sulfide solidifying volatiles are characterized by CO₂ and H₂O. Cu contents in the whole rocks display a clear correlation with the CO₂ contents released at 400–900 °C, and a rough correlation with the CO₂ contents at 900–1200 °C, but are irrelevant to CO₂ contents of secondary volatiles (200–400 °C) (Fig. 6). CO₂ is in a supercritical state in a mantle-derived magma system, and can leach and enrich great quantity of the ore-forming metal elements from penetrated mantle rocks (Ballhaus et al., 1994; Anthony et al., 1995). The relationships between the Cu and CO₂ contents at the different temperature intervals suggest that the supercritical state of CO₂ in the ore-forming magma system might be involved in the enrichment of large amounts of Cu and Ni elements under high temperature and pressure (Fleet and Wu, 1995).

The $\delta^{13}\text{C}_{\text{CO}_2}$ values (from –20.86 ‰ to –12.85 ‰) of the ore-forming magmatic volatiles range between mantle and crust values (Fig. 4), indicating a crustal contaminant (about 3–10%) has been added into the mantle-derived ore-forming magma (Fig. 7). Positive $\varepsilon_{\text{Nd}}(t)$ values (from +4 to +10, Zhang et al., 2006; Li et al., 2012) and (⁸⁷Sr/⁸⁶Sr)_i (0.7034–0.7057, Song and Li, 2009; Zhang et al., 2009; Li et al., 2012) in the Kalatongke deposits vary among the depleted mantle, the upper continental crust and coeval A-type granites in this region (Fig. 7); combined with Re–Os and O isotopic data (av. $\gamma_{\text{Os}}=183$, $\delta^{18}\text{O}=5.4\text{‰}–10.2\text{‰}$, Han et al., 2006; Zhang et al., 2006, 2009), suggesting that a crustal contamination was involved in the ore-forming magma of the Kalatongke deposits that originated from depleted asthenospheric mantle (Han et al., 2006; Zhang et al., 2006, 2009; Li et al., 2012).

The Kalatongke Cu–Ni sulfide deposits were formed by fractional evolution processes of the ore-forming parent magma of high MgO content (Li et al., 2012). The sulfur saturation was caused by the addition of sulfur, crustal contamination or olivine and pyroxene fractional crystallization, and thereby accelerated sulfide melt segregation (Zhang et al., 2003; Qian et al., 2009; Zhang et al., 2009; Jia et al., 2009; Zou et al., 2011; Li et al., 2012). The lighter $\delta^{13}\text{C}_{\text{CO}_2}$ values of the pyroxene at 400–900 °C and 900–1200 °C indicate crustal contamination occurred prior to or coincided with pyroxene crystallization in the ore-forming magma (Zhang et al., 2009). Because sulfide solubility is correlated inversely with oxygen fugacity (Haughton et al., 1974; Buchanan and Nolan, 1979), the elevated contents of CO₂ in the magma can increase oxygen fugacity, and thereby reduce the solubility of sulfur in the magma, and then trigger

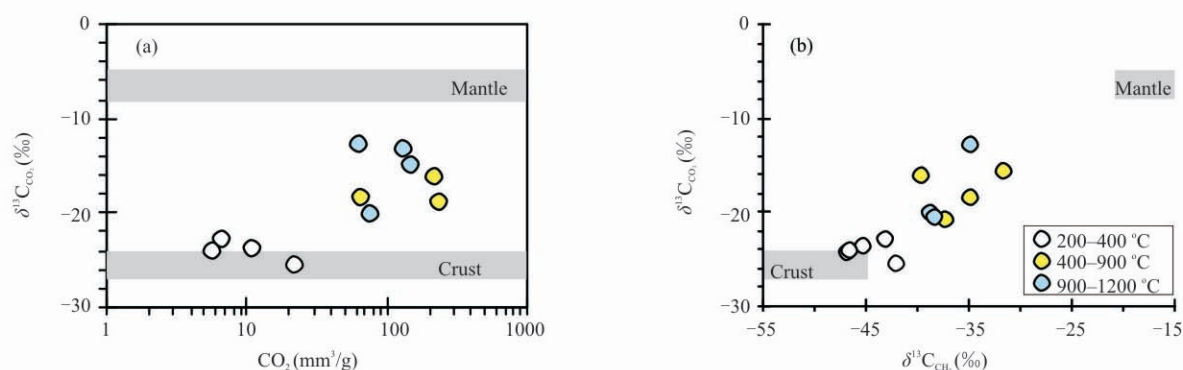


Fig. 4. Plots showing variations of $\delta^{13}\text{C}$ (‰ vs. PDB) of CO_2 and CH_4 in the Kalatongke deposits.

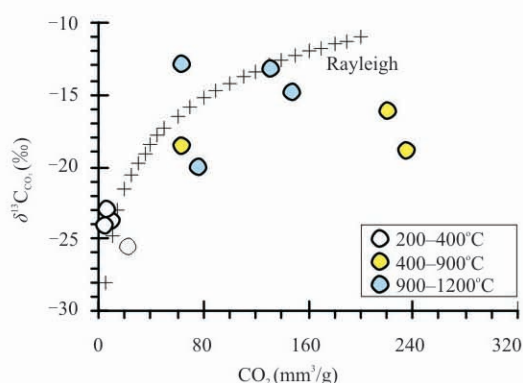


Fig. 5. Plot of $\delta^{13}\text{C}$ (‰ vs. PDB) with the contents of CO_2 in the Kalatongke deposits. Rayleigh-Rayleigh degassing model of carbon isotope (initial value of $\delta^{13}\text{C}$ is -11‰ , $1000\ln\alpha = -4.6\text{‰}$).

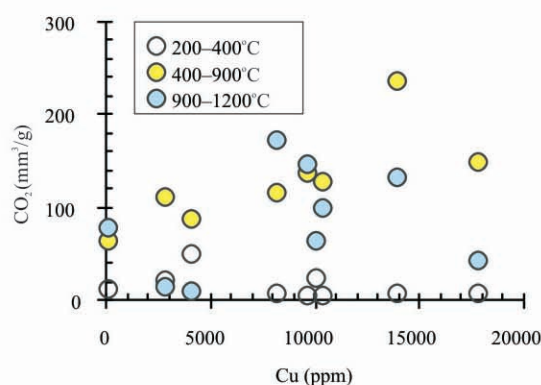


Fig. 6. Relationship between the contents of CO_2 released at different temperatures in the Kalatongke deposits and the contents of Cu in the whole rock (Cu data from Jia et al., 2009).

sulfide melt segregation (Lehmann et al., 2007).

Intrusion 1[#] has lighter $\delta^{13}\text{C}_{\text{CO}_2}$, higher CO_2 contents and CO_2/CH_4 ratios than intrusion 2[#], and higher SO_2 content (av. 2482 mm^3/g), and $\text{SO}_2/\text{H}_2\text{S}$ ratios (63) than intrusion 3[#] (av. 479 mm^3/g , 1 for $\text{SO}_2/\text{H}_2\text{S}$). These suggest that the intrusion 1[#] contain more oxidized components in ore-forming magma volatiles than the intrusions 2[#] and 3[#], more oxidized volatiles may cause sulfur saturation and sulfide segregation more easily and finally lead to the greater sulfide ores hosted in intrusion 1[#].

5.3 Tectonic implications

The elevated H_2 contents in the olivine and pyroxene (av. $\sim 4.2\text{ mol}\%$ and $\sim 37.4\text{ mol}\%$, respectively) in the Kalatongke deposits suggest that the ore-forming magmatic volatiles might be derived from the deep mantle (Navon et al., 1988; Javoy and Pineau, 1991; Liu et al., 1996; Zhang et al., 2005). The Nd, Sr, Pb and C isotopes of the Kalatongke deposits show the features of MORB with the presence of asthenosphere matters (Jiang et al., 2009; Song and Li, 2009), and followed by the addition of

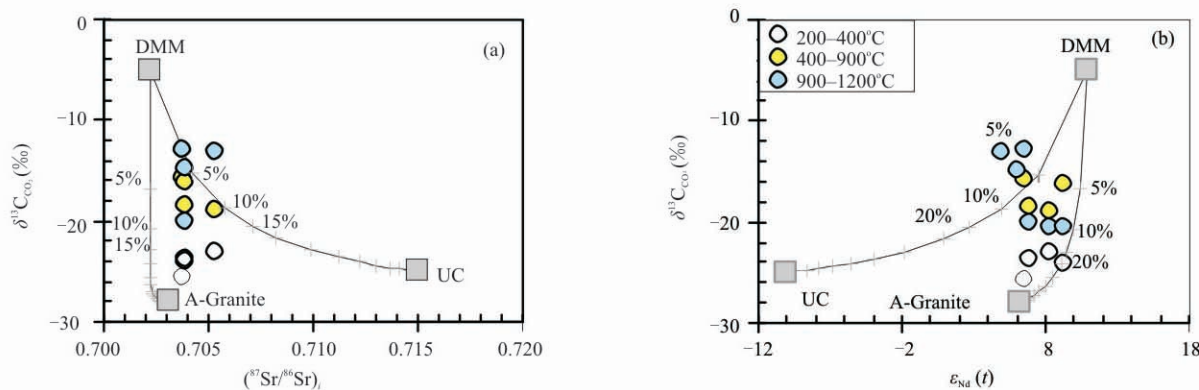


Fig. 7. Plots showing variations of $\delta^{13}\text{C}$ (‰ vs. PDB) of CO_2 with $(^{87}\text{Sr}/^{86}\text{Sr})_i$ and $\epsilon_{\text{Nd}}(t)$ (Sr and Nd isotopic data from Li et al., 2012).

crustal matters during the ascent of the mantle-derived ore-forming magma.

The abundant volatiles (av. 16599 mm³/g) and H₂O (av. 15000 mm³/g) exist in the olivine and pyroxene separates in the Kalatongke deposits, especially the H₂O content is much higher than that in the Siberian large igneous provinces (av. 4500 mm³/g, Tang et al., 2011). The hydrous minerals (e.g. amphibole and biotite) in the norite suggest that the ore-forming magma had the substantial H₂O and probably formed in a subduction-related source. The strong enrichment of LILEs and LREE, depleted in HFSE, and the significantly negative anomalies of Nb–Ta–Ti also support the derivation of water-rich volatiles from a subducted plate. Consequently, the primary magma of the Kalatongke Cu–Ni sulfide deposits was derived from metasomatized asthenosphere modified by aqueous volatiles from a recycled subducted slab during a previous subduction (Zhang et al., 2009), which was most likely formed related to the slab window (Li et al., 2012).

6 Conclusions

(1) The ore-forming magma volatiles in the Kalatongke Cu–Ni sulfide deposits are characterized by dominant H₂O (av. ~92 mol %), and relatively high contents of H₂, CO₂, H₂S and SO₂. The elevated contents of H₂ and H₂O suggest a deep mantle origin for the ore-forming volatiles with aqueous volatiles from recycled subducted slab. CO₂ contents correlated with whole rock Cu contents suggests that a supercritical state of CO₂ in the ore-forming magma might play a key role in the assemblage of huge Cu and Ni elements under high temperature and pressure.

(2) The lighter $\delta^{13}\text{C}_{\text{CO}_2}$ (from –20.86‰ to –12.85‰) and $\delta^{13}\text{C}_{\text{CH}_4}$ values (from –39.64‰ to –31.49‰) of the pyroxene samples indicate crustal contamination occurred prior to or synchronous with pyroxene fractional crystallization in mantle-derived ore-forming magma, and sulfur saturation caused by crustal contamination or olivine and pyroxene fractional crystallization. Meanwhile high CO₂ content in the ore-forming magma volatiles could be a factor of triggering sulfur saturation and sulfide melt segregation.

(3) Intrusion 1[#] shows more oxidized volatiles than intrusions 2[#] and 3[#], which may cause sulfur saturation and sulfide segregation more easily and finally lead to the greater sulfide ores hosted in intrusion 1[#].

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