Geochronology, fluid inclusions and isotopic characteristics of the Chaganbulagen Pb–Zn–Ag deposit, Inner Mongolia, China

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A B S T R A C T

The large Chaganbulagen Pb–Zn–Ag deposit is located in the Derbugan metallogenic belt of the northern Great Xing’an Range. The vein-style orebodies of the deposit occur in the NWW-trending fault zones. The ore-forming process at the deposit can be divided into three stages: an early quartz–pyrite–arsenopyrite–pyrrhotite–sphalerite–galena–chalcopyrite stage, a middle quartz–carbonate–pyrite–sphalerite–galena–silver-bearing minerals stage, and a late quartz–carbonate–pyrite stage. The sericite sample yielded a 40Ar–39Ar plateau age of 138 ± 1 Ma and an isochron age of 137 ± 3 Ma, indicating that the ages of mineralization and monzogranite porphyry in the Chaganbulagen deposit should be the Early Cretaceous, and that the mineralization should be slightly later than the intrusion of monzogranite porphyry. There are only liquid inclusions in quartz veins of the Chaganbulagen deposit. Homogenization temperatures, densities, and salinities of the fluid inclusions from the early stage are 261–340 °C, 0.65–0.81 g/cm³, and 0.7–6.3 wt.% NaCl eqv., respectively. Fluid inclusions of the middle stage have homogenization temperatures, densities, and salinities of 200–265 °C, 0.75–0.86 g/cm³, and 0.5–5.7 wt.% NaCl eqv., respectively. For fluid inclusions of the late stage, their homogenization temperatures, densities, and salinities are 173–219 °C, 0.85–0.91 g/cm³, and 0.4–2.7 wt.% NaCl eqv., respectively. The ore-forming fluids of the deposit are generally characterized by moderate temperature and low salinity and density, and belong to an H2O–NaCl ± CO2 ± CH4 system. The δ18Owater values calculated for ore-bearing quartz vary from −17.9‰ to −10.8‰, and the δDwater values from bulk extraction of fluid inclusion waters vary from −166‰ to −127‰, suggesting that the ore-forming fluids consist dominantly of meteoric water. The δ34Swater values range from 1.4‰ to 4.1‰. The 206Pb/204Pb, 207Pb/204Pb, and 208Pb/204Pb values of the ore minerals are in the ranges of 18.302–19.037, 15.473–15.593, and 38.110–38.945, respectively. The data for the S and Pb isotopic systems indicate that the ore-forming metals and sulfur came from Mesozoic magma. The Chaganbulagen deposit is a low-sulfidation epithermal Pb–Zn–Ag deposit, and the temperature decrease is the dominant mechanism for the deposition of ore-forming materials.

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1. Introduction

The Manzhouli area, located in the northeastern part of Inner Mongolia, bordering Mongolia to the west and Russia to the north (Fig. 1a), is one of the most important Pb–Zn–Ag–Cu–Mo metallogenic regions in China. To date, a great number of economically important porphyry Cu–Mo deposits (e.g., Wunugetushan and Badaguan), medium–low temperature hydrothermal vein-type Pb–Zn–Ag deposits (e.g., Jiawula and Chaganbulagen), and low-sulfidation epithermal Ag deposits (e.g., Erentaolegai) have been discovered (Liu et al., 2014; Wu et al., 2010). The Chaganbulagen and Jiawula deposits are only 5 km apart and are connected by the NWW-trending Jiawula–Chaganbulagen (Jia–Cha for short) fault, with the former developed in the southeast segment and the later in the northwest segment of the Jia–Cha fault. In addition, a few Cu–Zn–Sn skarns (e.g., Longling) and numerous high-sulfidation epithermal Cu–Au mineral occurrences (e.g., Bayanhaolei and Daba) have also been discovered in this area (Fig. 1b).

Previous studies in this area have mainly described geological characteristics of typical deposits (Qi et al., 2005; Zeng, 2010), host rock features and mineral paragenesis (Chen et al., 2008; Qin et al., 1999; Sheng and Fu, 1999), genetic classification (Nie et al., 2011; Yang et al., 2009), and fluid inclusions properties (Shuang, 2012; Wu et al., 2007, 2010; Zhai et al., 2010). But reliable ore-forming ages for the Chaganbulagen
Pb–Zn–Ag deposit have not been reported. Furthermore, sources for the ore-forming fluids and genetic classification of the Chaganbulagen Pb–Zn–Ag deposit are also controversial. Zhao and Zhang (1997) and Sheng and Fu (1999) reported a K–Ar age of 133–110 Ma and a single grain zircon U–Pb age of 139.2 Ma, respectively, for monzogranite porphyry that is spatially related to the Chaganbulagen deposit. However, these ages have a wide range and cannot directly represent the age of mineralization. As for sources of the ore-forming fluids, Pan et al. (1990) pointed out that the ore-forming fluids were mainly from the magmatic water, but Xie and Liu (2001) considered that they were mainly from the meteoric water. The Pb–Zn–Ag deposits in the Manzhouli area have mainly been classified as one of the two genetic types of deposits: medium–low temperature hydrothermal vein-type deposits (Wu et al., 2010; Zhao and Zhang, 1997) or low-sulfidation epithermal deposits (Li et al., 2015; Xie and Liu, 2001; Zhao and Wu, 2002). In this paper, we present new sericite 40Ar − 39Ar age, zircon LA-ICP-MS (laser ablation multi-collector inductively coupled plasma mass spectrometry) U–Pb age, analyses of fluid inclusions, and H–O–S–Pb isotopes. The purpose of this study is to classify the deposit type, determine the mineralization age, reveal the characteristics of the ore-forming fluids and their evolution, and discuss the sources of ore-forming fluids and metals of the Chaganbulagen deposit.

2. Regional geology

The Manzhouli region is located in the southeastern margin of the Argun orogenic belt, also referred to as the Argun massif, which belongs to the eastern segment of the Sayan–Central Mongolia–Argun orogenic belt. The NE-trending Argun orogenic belt is one of the key tectonic units in northeastern China and is bounded by the Mesozoic Mongolia–Okhotsk suture to the northwest and the Derbugan fault to the southeast (Fig. 1a). This orogenic belt experienced a complicated
tectonic evolution involving at least six major geological events since the Neoproterozoic: (1) Middle–Late Neoproterozoic to Early Cambrian, development of a geosyncline and deposition of marine basalt, andesite, dacite, mudstone, siltstone, and carbonate of the Xinghuadukou Group, marine sandstone, siltstone, carbonate, dacite, and rhyolite of the Jiageda Formation, and marine carbonate, sandstone, siltstone, and mudstone of the Argunhe Formation (Wu et al., 2012); (2) Middle Cambrian–Early Ordovician Khanka orogeny resulted in amalgamation of the Argun block and the Vitim–Stanovoy block, which is located at the southeastern edge of the Siberian Craton, forming the Argun orogenic belt (Ge et al., 2007; Wu et al., 2005, 2012); (3) Early Paleozoic amalgamation of the above units with the Central Mongolia block to the subduction of the Paleo-Asian Oceanic plate beneath the Siberia Craton and the formation of the Okhotsk–Beringian back-arc basin. At this time, the Argun block separated from the Siberian Craton again and formed the basement of island-arc terranes between the Paleo-Asian and the Mongol–Okhotsk oceans; (4) Early Mesozoic subduction of the Mongol–Okhotsk Ocean basin toward both the north and the south resulted in the formation of calc–alkaline granitoids and related porphyry Cu–Mo deposits in the Argun orogenic belt (e.g., Badaguan, Babayi, and Wunugetushan); and (5) collision of the Mongol–North China continent with the Siberian Craton, resulting in the closure of the Mongol–Okhotsk ocean basin in the late Middle Jurassic (Sorokin et al., 2004). Subsequently, the Argun area evolved into either a post-collisional/post-orogenic setting resulting from collapse of the Mongol–Okhotsk orogenic belt (Fan et al., 2003) or a back-arc extensional environment caused by sequential slab break off, retreat, and roll back of the Paleopacific (Izanagi) plate (Pirajno and Zhou, 2015; Wang et al., 2006) during the Late Jurassic–Early Cretaceous. This induced large-scale magmatic activity and hydrothermal metallogeny, forming the Great Xing’an Range giant igneous province and the Argun Pb–Zn–Ag metallogenic belt (Pirajno and Zhou, 2015; Wu et al., 2007).

Pre-Mesozoic strata, consisting of rocks of the Neoproterozoic Jiageda and Sinian–Lower Cambrian Argunhe Formations, are sporadically distributed in the Manzhouli area. The Jiageda Formation consists of sericite–quartz schist, quartzite, sandstone, slate, rhyolitic tuff, and marble, whereas the Argunhe Formation mainly comprises marble, with a small amount of sericite–quartz schist and metasandstone. The Mesozoic strata are widely distributed and subdivided, from bottom to top, into the Middle Jurassic Wanbao and Tamulangou formations, Upper Jurassic Manketouebo and Manitu formations, and Lower Cretaceous Baiyingaolao, Meiletu, and Damoguaihe formations. The Wanbao Formation is composed of terrestrial and sandy conglomerates, sandstone, and thin pelitic siltstone with coal seams; the Tamulangou Formation consists of andesite and basaltic andesite; the Manketouebo Formation is mainly made of rhyolite; the Manitu Formation is composed of andesite and dacite; the Baiyingaolao Formation consists of dacite, rhyolite, and tuff; the Meiletu Formation is composed of basalt and andesite; and the Damoguaihe Formation comprises conglomerate, sandstone, and shale with coal seams (Meng et al., 2011).

Faults and folds are well developed in the Manzhouli area. The faults are mainly NW- and NE-trending. The NE-trending faults are represented by the Derbugan fault and controlled the distribution of a NE-trending uplift and depression. Many porphyry Cu–Mo and hydrothermal vein-type Pb–Zn–Ag deposits occurred in the uplift, along the northwestern side of the Derbugan fault, forming the NE-striking Derbugan metallogenic belt. The NW-trending faults mainly include the Badaguan, Hanigou, and Muhar faults, and they control the distribution of NW-trending smaller belts of mineralization. The pre-Mesozoic faults are mainly NE-striking and are characterized by tight and even reversed form, whereas the Mesozoic faults commonly show open shapes with short axes, forming dome and basin patterns. Widespread volcanic structures are superimposed on the older regional structures.

Regional magmatism can be divided into the Late Hercynian, Indosinian, Early Yanshanian, and Late Yanshanian periods (Wu et al., 2010). The Hercynian granitoids, consisting of dominantly granite, granodiorite, and monzogranite, occur as batholiths or stocks, with K–Ar isotope ages of 271–262 Ma (Zhao and Zhang, 1997). The Indosinian granitoids, consisting of dominantly monzogranite and syenogranite, with a small amount of granodiorite, occur as batholiths, with Rb–Sr isochron ages varying from 225 to 211 Ma (Qin et al., 1998) and zircon U–Pb age of 231 Ma (Kang et al., 2014), and are related to Indosinian porphyry Cu–Mo mineralization. The Early Yanshanian granitoids, consisting of biotite granite, granodiorite, and monzogranite, occur as batholiths or stocks, with a single grain zircon U–Pb age of 188.3 Ma (Qin et al., 1999) and whole rock Rb–Sr isochron age of 184–178 Ma (Chen et al., 2008; Qin et al., 1999), and are related to early Yanshanian porphyry Cu–Mo mineralization (Wu et al., 2010). The Late Yanshanian granitoids, mainly comprising granite porphyry, monzogranite porphyry, and syenogranite porphyry, occur as apophyses or stocks, with K–Ar isotope ages ranging from 138 to 93 Ma (Qin et al., 1995) and a single grain zircon U–Pb age of 139.2 Ma (Sheng and Fu, 1999), and are related to the Pb–Zn–Ag mineralization (Wu et al., 2010).

3. Ore deposit geology

The Chaganbulagen deposit is located 150 km southwest of Manzhouli City, Inner Mongolia. The deposit, discovered in 1985, contains proven reserves of 191,966 t Pb, 216,713 t Zn, 8478 t Cu, and 959 t Ag, with average grades of 2.62% Pb, 2.95% Zn, 0.15% Cu, and 154 g/t Ag. The Chaganbulagen deposit has been mined since 2002.

The exposed strata in the Chaganbulagen deposit are Middle Jurassic terrestrial conglomerate and sandstone of the Wanbao Formation, Middle Jurassic andesite and basaltic andesite of the Tamulangou Formation, Late Jurassic rhyolite of the Manketouebo Formation, Late Jurassic andesite and dacite of the Manitu Formation, and Early Cretaceous dacite, rhyolite and tuff of the Baiyingaolao Formation (Meng and Qin, 1997; Wu et al., 2010). The NWW-striking Jia–Cha fault is more than 3000-m-long, 200-m-wide and 500-m-deep, strikes 280°–290°, dips SW and controls the distribution of orebodies at the Chaganbulagen deposit. There are also the NW- and NE-trending faults. The intrusive rocks in the Chaganbulagen deposit are mainly composed of the Late Hercynian granite and the Yanshanian granitic porphyry (Wu et al., 2010).

The main orebodies of the Chaganbulagen deposit are the nos. I, II, V, IX, and X orebodies (Fig. 2). All of the orebodies occur in the NWW-trending fault zones; of these orebodies, the no. IX orebody is the largest, being more than 1400-m-long and 0.3–22.9-m-thick with an average thickness of 1.9 m. The average grades of the Chaganbulagen deposit are 2.86% Pb, 3.63% Zn, 146 g/t Ag, and 0.17% Cu, respectively. The ore minerals are mainly galena, sphalerite, pyrite, chalcopyrite, pyrrhotite, and arsenopyrite (Fig. 3). The main silver-bearing minerals are andorite, freibergite, pyrrargyrite, canfieldite, and argentite. The gangue minerals are mainly quartz, calcite, illite, hydromuscovite, sericite, kaolinite, and fluorite. The ores mainly comprise xenomorphic granular, exsolution, intergrowth, poikilitic, euhedral–subhedral granular, and intertextural textures (Fig. 3a–c). The sphalerite and galena are intergrowth (Fig. 3a). The xenomorphic granular pyrite (0.3–0.6 mm) is distributed in crystals of galena, developing a poikilitic texture (Fig. 3b). Exsolution blebs of chalcopyrite (5–25 μm) occur in crystals of sphalerite (Fig. 3c). The structures of the ores are massive, disseminated, brecciated, vein, or veinlet (Fig. 3d–f). The alteration assemblage includes silification, carbonatization, illitization, hydromuscovitization, chloritization, kaolinization, sericitization, and fluoritization. The dominant alteration related to the Pb–Zn–Ag mineralization is a silification, sericitization, carbonatization, and illitization assemblage, occurring in the NWW ore-bearing fault. The crosscutting relationship in the veins (Fig. 3g–i) indicates that the ore-forming process of the Chaganbulagen deposit can be divided into three stages: (1) an early quartz–pyrite–arsenopyrite–pyrrhotite–sphalerite–galena–chalcopyrite stage; (2) a middle quartz–carbonate–pyrite–sphalerite–galena–silver-bearing minerals stage; and
Institute of Atomic Energy. H8 channels were used and the neutron irradiation, which was conducted in the pool heap of the China Academy of Geological Sciences (CAGS). The sericite to be analyzed was sealed in a quartz bottle and placed in a nuclear reactor for neutron irradiation (Fig. 4b). Because sericite has a small particle size, it is difficult to obtain in its pure form. Therefore, we utilized the inverse-selection method by crushing the sample and maximally removing metallic sulfides and quartz, then using the remaining pure sericite for the \( ^{40}\text{Ar} - ^{39}\text{Ar} \) dating. \( ^{40}\text{Ar} - ^{39}\text{Ar} \) isotope dating was undertaken at the Ar–Ar Isotope Geology Laboratory of the Institute of Geology, Chinese Academy of Geological Sciences (CAGS). The sericite to be analyzed (purity >99%) was selected and ultrasonically cleaned. The cleaned sample was sealed in a quartz bottle and placed in a nuclear reactor for neutron irradiation, which was conducted in the pool heap of the China Institute of Atomic Energy. H8 channels were used and the neutron flux density was approximately \( 6.0 \times 10^{12} \text{neutrons/cm}^2 \text{s} \). The total irradiation time was 3064 min, and the integrated neutron flux was \( 1.10 \times 10^{18} \text{neutrons/cm}^2 \). Simultaneously, neutron irradiation was applied to the national reference ZBH-25 biotite of China as the monitoring standard. The systematic blank levels for \( ^{40}\text{Ar}, ^{39}\text{Ar}, ^{37}\text{Ar}, \) and \( ^{36}\text{Ar} \) were smaller than \( 3 \times 10^{-15}, 4 \times 10^{-16}, 8 \times 10^{-17}, \) and \( 2 \times 10^{-17} \) mol, respectively. Correction coefficients for the interference isotopes resulting from the neutron irradiation process were obtained by analyzing irradiated \( \text{K}_2\text{SO}_4 \) and \( \text{CaF}_2 \), with values of \( (^{37}\text{Ar}/^{39}\text{Ar})_\text{Ca} = 0.0002389, (^{40}\text{Ar}/^{39}\text{Ar})_\text{Ca} = 0.004782, \) and \( (^{39}\text{Ar}/^{37}\text{Ar})_\text{Ca} = 0.000806 \) being obtained. \( ^{37}\text{Ar} \) was corrected for radioactive decay with a decay constant \( (^{40}\text{K}) \) of \( 5.543 \times 10^{-10} \text{year}^{-1} \) (Steiger and Jager, 1977), and the uncertainty of the plateau age is given at the 2σ level. Isoplot was used to compute the positive and inverse isochrones (Ludwig, 2003). Details of the experimental procedures have been described by Mao et al. (2006) and Wu et al. (2014).

### 4.2. Zircon LA-ICP-MS U–Pb dating

The sample NJ-12 was collected from monzogranite porphyry in the Chaganbulagen deposit. The sample is light gray in color, has a porphyritic texture and massive structure, and reveals pyritization (Fig. 5a). The phenocrysts are mainly plagioclase (~20% of the rock mass), K-feldspar (~15%), quartz (~5%), and biotite (~3%). The groundmass is dominated by microlites of plagioclase (~15%), K-feldspar (~20%), quartz (~20%), and biotite (~2%) (Fig. 5b–d). Plagioclase phenocrysts are anhedral or subhedral tabular in shape with lengths of 0.6–4.5 mm; of these phenocrysts show weak sericitization and carbonation (Fig. 5b, c). K-feldspar phenocrysts are subhedral tabular or granular in shape with lengths of 0.5–2.5 mm; of these phenocrysts show weak argillization. Quartz phenocrysts are granular in shape with lengths of 0.4–1.5 mm (Fig. 5d). Biotite phenocrysts are flake in shape with lengths of 0.4–1.3 mm (Fig. 5b).

Zircons were separated from sample NJ-12 using the conventional heavy liquid and magnetic techniques and purified by handpicking under a binocular microscope at the Langfang Regional Geological Survey, Hebei Province, China. The handpicked zircons were mounted in epoxy and polished to expose the cores of the grains for subsequent cathodoluminescence (CL) imaging and zircon U–Pb analyses. The CL images were obtained at the Institute of Geology, CAGS. The samples were analyzed at the Ministry of Land and Resources (MLR) Key Laboratory of Metallogeny and Mineral Assesment, Institute of Mineral Resources, CAGS using a Finnigan Neptune MC-ICP-MS attached to a New Wave UP 213 laser ablation system with an in-house sample...
cell. The detailed analytical procedures are similar to those described by Yang et al. (2014) and Wu et al. (2015). U–Pb fractionation was corrected using zircon standard GEMOC GJ-1 (207Pb/206Pb age of 608.5 ± 1.5 Ma, Jackson et al., 2004), and accuracy was controlled using zircon standard Mud Tank (intercept age of 732.0 ± 5.0 Ma, Black and Gulson, 1978). U–Pb ages were calculated from raw signal data using Isoplot 3.0 (Ludwig, 2003). Because 204Pb could not be measured due to a low signal and interference from 204Hg in the gas supply, common lead correction was carried out using the EXCEL program ComPbCorr#3 15G (Andersen, 2002). Errors on individual analyses by LA-ICP-MS are quoted at the 1σ level, while errors on pooled ages are quoted at the 95% (2σ) confidence level.

4.3. Fluid inclusion microthermometry and laser Raman spectroscopy

Ten samples were selected for fluid inclusion (FI) analyses, including eight samples of the early stage, one sample of the middle stage, and one sample of the late stage. The eight samples of the early stage, collected from orebody no. II at the 510-m and 450-m levels, orebody no. X at the 510-m, 450-m, and 390-m levels, and orebody no. VI at the 450-m level, are quartz–pyrite–arsenopyrite–pyrrhotite–sphalerite–galena–chalcopyrite veins. The one sample from the middle stage is collected from orebody no. II at the 580-m level and is the quartz–carbonate–pyrite–sphalerite–galena–silver-bearing mineral vein. The one sample of the late stage, collected from orebody no. X at the 510-m level, is quartz–carbonate–pyrite vein.

Microthermometric measurements were conducted using a Linkam THMSG 600 heating–freezing stage mounted on an Olympus BX-50 microscope at the China University of Geosciences, Beijing. The estimated accuracies of the freezing and heating measurements were ±0.1 °C from −100 °C to 25 °C, ±1 °C from 25 °C to 400 °C, and ±2 °C from above 400 °C, respectively. Fluid salinities were calculated from ice-melting temperatures using the equation of Bodnar (1993). Fluid densities were calculated with the equation of Liu and Duan (1987).

Volatile compositions of single FIs were identified by using a Renishaw inVia laser Raman probe at the MLR Key Laboratory of Metallogeny and Mineral Assessment, Institute of Mineral Resources, CAGS. The wavelength of the Ar+ laser was 514.5 nm and the beam size is 1 μm. The spectrum was measured from 100 to 4500 cm⁻¹ and the acquisition time was 20 s. The spectrum resolution was ±2 cm⁻¹. Instrumental setting was kept constant during all analyses.

4.4. H–O–S–Pb isotopes

Eleven quartz samples from the early, middle, and late stages were analyzed for O (quartz) and H (fluid inclusions) isotopes. The samples include: (1) NC-1, NC-5, NC-10, NC-12, NC-13, NC-14, and NC-17,
collected from orebody no. X at the 510-m, 450-m, and 390-m levels, respectively, are the quartz–pyrite–arsenopyrite–pyrrhotite–sphalerite–galena–chalcopyrite veins of the early stage; (2) NC-21 and NC-48, collected from orebody no. II at the 510-m level and orebody no. VI at the 450-m level, respectively, are also the quartz–pyrite–arsenopyrite–pyrrhotite–sphalerite–galena–chalcopyrite veins of the early stage; (3) C2-5801 was collected from orebody no. II at the 580-m level and is the quartz–carbonate–pyrite–sphalerite–galena–silver-bearing mineral vein of the middle stage; and (4) NC-8 was collected from orebody no. X at the 510-m level and is the quartz–carbonate–pyrite vein of the late stage. The H–O isotope analyses were accomplished at the MLR Key Laboratory of Metallogeny and Mineral Assessment, Institute of Mineral Resources, CAGS, using a Finnigan MAT253 mass spectrometer. The accuracy of the O isotope analysis is better than ±0.2‰, and that of the H isotope analysis is better than ±2‰ (Mao et al., 2008). The δ18O of water was calculated from the O isotopes of quartz by using the fractionation equation 10001nwater = (3.38 × 106)

\[ T - 2 - 3.40, \]

where \( T \) is the temperature in degreeKelvin (Clayton et al., 1972), and the average fluid inclusion temperature of each stage was used to calculate the δ18Owater Value.

The sulfur and lead isotope compositions of the four pyrite, three galena, and three sphalerite samples, separated from the seven ore samples from the middle stage of the Chaganbulagen deposit, were measured at the Analytical Laboratory of the Beijing Research Institute of Uranium Geology. Of these seven samples, (1) NC-4, NC-10, and NC-81 were collected from orebody no. X at the 510-m, 450-m, and 390-m levels and are massive Pb–Zn ores with pyrite; (2) NC-44, NC-46, and NC-47, collected from orebody no. VI at the 450-m level, are massive Pb–Zn ores with pyrite; and (3) NC-20 was collected from orebody no. II at the 510-m level and is a massive Pb–Zn ore.

For the sulfur isotope analysis, each sample was weighed to 15 mg, mixed with CuO powder, placed in a vacuum quartz tube, and allowed to react for 15 min at a temperature of 1100 °C (Robinson and Kusakabe, 1975). After purification, SO2 was transferred to the sample tube and the S-isotopic ratio 34S/32S was measured on the mass spectrometer of a Finnigan MAT 251 instrument. The lead isotopic composition was measured using a GV IsoProbe-T multi-collector thermal ionization mass spectrometer. The 208Pb/206Pb and 207Pb/206Pb ratios had a precision better than 0.005% (2σ). The Pb-isotopic analyses are reported with respect to Pb standard reference NBS-981 values (Todt et al., 1993): 206Pb/204Pb = 16.934 ± 0.007, 207Pb/204Pb = 15.486 ± 0.012, and 208Pb/204Pb = 36.673 ± 0.033, respectively.

5. Analytical results

5.1. Sericite 40Ar–39Ar age

The 40Ar–39Ar analytical results for a sericite sample are summarized in Table 1 and illustrated in Fig. 6. In this study, an age plateau is defined as a sequence of five or more consecutive steps corresponding to at least 60% of the total 39Ar released that yield apparent ages reproducible at the 95% confidence level (2σ). The sample yielded a well-defined plateau age of 138 ± 1 Ma and an isochron age of 137 ± 3 Ma (Fig. 6a, b). The plateaus comprise ten continuous steps accounting for 89% of the total 39Ar released. The overwhelming majority of the 39Ar released all satisfy the plateau-forming conditions, and their correlation coefficients are very high, indicating that the sericite experienced no obvious disturbance after mineral precipitation. Therefore, both the plateau and the isochron ages obtained in this study can represent the true ore-forming time of the Chaganbulagen Pb–Zn–Ag deposit.
5.2. Zircon U–Pb age

Twelve zircons from sample NJ-12 of the monzogranite porphyry were selected for LA-ICP-MS U–Pb dating. The zircon U–Pb compositions were analyzed with reference to their CL images (Fig. 7). Sample NJ-12 contains a population of fine-grained, colorless to light yellow zircon crystals with adamantine luster. The zircons are pyramidal or bipyramidal in shape with length/width ratios of 1–3. A total of 15 spots were analyzed from the 12 grains; the results are presented in Table 2 and Fig. 7. Except for three analyses (spots NJ12-1.1, NJ12-8.1, and NJ12-9.1) presenting relatively new or old 206Pb/238U ages (136Ma, 148Ma, and 150Ma), the remaining 12 analytical spots show a tight, concordant grouping defining a weighted mean 206Pb/238U age of 143 ± 2 Ma with an MSWD value of 0.23 (Fig. 8a, b). All of the zircons in the sample have high Th/U ratios (1.49–4.21), indicative of a magmatic origin (Belousova et al., 2002).

5.3. Fluid inclusion study

5.3.1. Petrography

The fluid inclusions in the Chaganbulagen deposit are only liquid inclusions, including primary and secondary inclusions. These primary inclusions are oval, elongated, and irregular, with diameters of 4–32 μm, although mainly 7–18 μm, and contain bubbles typically accounting for 5–55% of the total volume. They homogenize to liquid when heated. Inclusions of this type account for approximately 65% of the total

<table>
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<th>Step</th>
<th>T(°C)</th>
<th>40Ar/39Ar</th>
<th>36Ar/39Ar</th>
<th>37Ar/39Ar</th>
<th>38Ar/39Ar</th>
<th>40Ar*/39Ar</th>
<th>39Ar (×10−14 mol)</th>
<th>39Ar (cum.) (%)</th>
<th>Age(Ma) (±1σ)</th>
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<td>0.0000</td>
<td>0.0209</td>
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<td>0.12</td>
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<td>127.5 ± 2.0</td>
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<td>4</td>
<td>1060</td>
<td>63.8319</td>
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<td>0.0141</td>
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<td>36.95</td>
<td>139.1 ± 1.3</td>
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<td>0.0000</td>
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<td>0.0113</td>
<td>0.0143</td>
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<td>2.01</td>
<td>63.71</td>
<td>137.6 ± 1.3</td>
</tr>
<tr>
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<td>1160</td>
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<td>0.0013</td>
<td>0.0113</td>
<td>0.0143</td>
<td>62.7305</td>
<td>0.85</td>
<td>72.31</td>
<td>137.7 ± 1.3</td>
</tr>
<tr>
<td>8</td>
<td>1200</td>
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<td>0.0000</td>
<td>0.0148</td>
<td>62.4895</td>
<td>0.91</td>
<td>81.56</td>
<td>137.1 ± 1.3</td>
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<tr>
<td>9</td>
<td>1250</td>
<td>64.5999</td>
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<td>0.0000</td>
<td>0.0145</td>
<td>63.5637</td>
<td>0.86</td>
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<td>139.4 ± 1.4</td>
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<td>10</td>
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<td>0.0000</td>
<td>0.0150</td>
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<td>0.65</td>
<td>98.86</td>
<td>145.3 ± 1.4</td>
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<tr>
<td>11</td>
<td>1400</td>
<td>76.6443</td>
<td>0.0190</td>
<td>0.0000</td>
<td>0.0196</td>
<td>71.0143</td>
<td>0.31</td>
<td>100.00</td>
<td>155.1 ± 1.7</td>
</tr>
</tbody>
</table>
number of inclusions and are distributed in isolation in quartz of every stage (Fig. 9). The secondary inclusions are generally smaller in size than the primary inclusions, and occur along transcristalline microfractures in quartz crystals of the early and middle stages, accounting for 35% of the total number of inclusions.

5.3.2. Microthermometry

The microthermometric results and calculated salinities and densities are shown in Table 3 and Fig. 10. Below, we summarize the results from each hydrothermal stage.

Fluid inclusions in quartz of the early stage: The final ice melting temperatures of the FIs range from $-3.9$ °C to $-0.4$ °C, corresponding to salinities of $0.7$–6.3 wt.% NaCl eqv. The homogenization temperatures are between 261 °C and 340 °C, and the fluid densities vary from 0.65 to 0.81 g/cm$^3$ (Table 3; Fig. 10a, b). The eutectic temperatures range from $-21.6$ °C to $-19.7$ °C with an average of $-20.6$ °C, which is basically the same as the eutectic temperature of an H$_2$O–NaCl system ($-20.8$ °C), indicating that the fluids belonged to an H$_2$O–NaCl system.

Fluid inclusions in quartz of the middle stage: The final ice melting temperatures of the FIs range from $-3.5$ °C to $-0.3$ °C, with salinities of 0.5–5.7 wt.% NaCl eqv. The homogenization temperatures are 209–265 °C, and the fluid densities range from 0.75 to 0.86 g/cm$^3$ (Table 3; Fig. 10c, d). The eutectic temperature from one inclusion is $-19.9$ °C, which is slightly higher than that of an H$_2$O–NaCl system, indicating that the fluids roughly belonged to an H$_2$O–NaCl system.

Fluid inclusions in quartz of the late stage: The final ice melting temperatures of the FIs range from $-1.4$ °C to $-0.2$ °C, with salinities of 0.4–2.7 wt.% NaCl eqv. The homogenization temperatures are 173–219 °C, and the fluid densities are 0.85–0.91 g/cm$^3$ (Table 3; Fig. 10e, f). The two eutectic temperatures are $-20.9$ °C and $-19.2$ °C with an average of $-20.1$ °C, which is slightly higher than that of an H$_2$O–NaCl system, indicating that the fluids roughly belonged to an H$_2$O–NaCl system.

5.3.3. Laser Raman spectra

The results of representative laser Raman spectroscopic analyses are given in Fig. 11. The gas phase component for FIs in quartz of the early stage is mainly H$_2$O and CO$_2$ with a small amount of CH$_4$ (Fig. 11a, b). The gas phase component for FIs in quartz of the middle stage is mainly H$_2$O with a small amount of CO$_2$ (Fig. 11c). The gas phase component for FIs in quartz of the late stage is only H$_2$O (Fig. 11d). In general, from early to late, the CO$_2$ and CH$_4$ contents of the ore-forming fluids gradually decrease, evolving from an early H$_2$O–NaCl–CO$_2$ ± CH$_4$ system through a middle H$_2$O–NaCl ± CO$_2$ system into a late H$_2$O–NaCl system, with ore-forming fluids generally having affinities to an H$_2$O–NaCl ± CO$_2$ ± CH$_4$ system.

5.3.4. Trapping pressure of fluid inclusions and ore-forming depth

Fluid inclusions may provide data for the trapping pressure and resultant depth estimates (Brown and Hagemann, 1995; Roedder and...
Bodnar, 1980; Shepherd et al., 1985). Estimates of the trapping pressure can be obtained only if an independent estimate of trapping temperature is known or if the inclusions were trapped under immiscible conditions (Brown and Hagemann, 1995; Liu et al., 2014; Roedder and Bodnar, 1980; Shepherd et al., 1985). However, when fluid immiscibility or boiling is not documented, the trapping pressures only represent the minimum of fluid pressure (Roedder and Bodnar, 1980).

The trapping pressures were estimated based on the fluid inclusion data of the early and middle stages by using the Flincor software of Bodnar, 1980; Shepherd et al., 1985). However, when fluid immiscibility or boiling is not documented, the trapping pressures only represent the minimum of fluid pressure (Roedder and Bodnar, 1980).

5.4. Isotope data

5.4.1. Hydrogen–oxygen isotopes

The analytical results for oxygen isotopes of 11 quartz samples and hydrogen isotopes of fluid inclusions of 11 quartz samples from the Chaganbulagen deposit are shown in Table 4. Of these, nine samples from the early stage have δD values of −166‰ to −127‰, δ18Oquartz values of −6.8‰ to −4.0‰, and δ18Owater values of −13.6‰ to −10.8‰, respectively; the δD, δ18Oquartz, and δ18Owater values of one sample from the middle stage are −134‰, −1.5‰, and −11.0‰, respectively; and the δD, δ18Oquartz, and δ18Owater values of one sample from the late stage are −142‰, −6.3‰, and −17.9‰, respectively. All of these samples plot between the magmatic water box and the meteoric water line, with most data near the meteoric water line in the δ18Owater–δD diagram of Sheppard (1977) (Fig. 12).

5.4.2. Sulfur isotopes

Sulfur isotopic compositions for the four pyrite samples, three sphalerite samples, and three galena samples from the main ore-forming stage of the Chaganbulagen deposit are presented in Table 5 and Fig. 13. The δ34S(‰) values range from 1.4 to 2.9 for pyrite, 3.3 to 4.1 for sphalerite, and 1.7 to 2.5 for galena. These are in disagreement with the equilibrium sequence, which is δ34S(py) > δ34S(sp) > δ34S(gn), suggesting that the hydrothermal system was non-equilibrium condition during the formation of sulfides (Ohmoto, 1986). The δ34S values of sulfide ore minerals deposited in a non-equilibrium system may have been obtained by multi-stage activity of both structural features (e.g., fractures/faults) and magmatic events, which led to complicated changes in the physicochemical conditions of migrating fluids before
mineralization, suggesting that this ore depositional system evolved under relatively open conditions (Zhai et al., 2013).

5.4.3. Lead isotopes

The lead isotopic compositions for the four pyrite samples, three sphalerite samples, and three galena samples from the main ore-forming stage of the Chaganbulagen deposit are displayed in Table 5 and Fig. 14. The $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios for four pyrite samples vary from 18.316 to 19.037, 15.473 to 15.593, and 38.132 to 38.945, respectively. The $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios for three sphalerite samples range from 18.315 to 18.345, 15.530 to 15.570, and 38.160 to 38.244, respectively. The ratios of $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, and $^{208}\text{Pb}/^{204}\text{Pb}$ for three galena samples vary from 18.302 to 18.329, 15.509 to 15.555, and 38.110 to 38.218, respectively. In the $^{206}\text{Pb}/^{204}\text{Pb}$ versus $^{207}\text{Pb}/^{204}\text{Pb}$ diagrams of Zartman and Doe (1981) (Fig. 14a), the Pb isotopic compositions of the Chaganbulagen deposit are projected in the area between the orogenic belt and the mantle evolution lines; in the $^{206}\text{Pb}/^{204}\text{Pb}$ versus $^{208}\text{Pb}/^{204}\text{Pb}$ diagrams (Fig. 14b), all of the projected data points are located on the orogenic belt line or its vicinity.

6. Discussion

6.1. Mineralization age

In this study, the sericite from the Chaganbulagen deposit yielded a well-defined $^{40}\text{Ar}–^{39}\text{Ar}$ plateau age of 138 ± 1 Ma and an Ar–Ar isochron age of 137 ± 3 Ma. We also obtained a zircon LA-ICP-MS U–Pb age of 143 ± 2 Ma for monzogranite porphyry. The sericite Ar–Ar plateau and isochron ages represent an alteration event that is associated with the Pb–Zn–Ag mineralization, and therefore the ages are interpreted here as the mineralization age of the Chaganbulagen Pb–Zn–Ag deposit. The zircon LA-ICP-MS U–Pb age records the timing of magma crystallization. For the Jia–Cha ore field, Sheng and Fu (1999) reported a single grain zircon U–Pb age of 139.2 Ma for the monzogranite porphyry. Li et al. (2015) reported sulfide Rb–Sr isochron ages of 143–142 Ma for seven sphalerite and six pyrite samples from the Jiawula deposit. The dating results indicate that the deposits in the Jia–Cha ore field were formed approximately 143–137 Ma ago and that the monzogranite porphyry emplaced at approximately 143–139 Ma. Meng and Qin (1997), Wu et al. (2010), and Zhai et al. (2013) thought that the intrusion activity of the monzogranite porphyry had closely space–time

Table 3

<table>
<thead>
<tr>
<th>Stage</th>
<th>Type</th>
<th>Size (μm)</th>
<th>V (vol.%)</th>
<th>Number</th>
<th>$T_e$ (°C)</th>
<th>$T_m,\text{ice}$ (°C)</th>
<th>$T_h$ (°C)</th>
<th>Salinity (% NaCl equiv.)</th>
<th>Density (g/cm$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Early stage</td>
<td>WL</td>
<td>4–32</td>
<td>10–55</td>
<td>34</td>
<td>−21.6 to −19.7</td>
<td>−3.9 to −0.4</td>
<td>261–340</td>
<td>0.7–6.3</td>
<td>0.65–0.81</td>
</tr>
<tr>
<td>Middle stage</td>
<td>WL</td>
<td>5–22</td>
<td>5–30</td>
<td>28</td>
<td>−19.9</td>
<td>−3.5 to −0.3</td>
<td>205–265</td>
<td>0.5–5.7</td>
<td>0.75–0.86</td>
</tr>
<tr>
<td>Late stage</td>
<td>WL</td>
<td>5–20</td>
<td>5–30</td>
<td>33</td>
<td>−20.9 to −19.2</td>
<td>−1.4 to −0.2</td>
<td>173–219</td>
<td>0.4–2.7</td>
<td>0.85–0.91</td>
</tr>
</tbody>
</table>

Notes: V, volume fraction of gas phase in the total volume of inclusion; $T_e$, eutectic temperature; $T_m,\text{ice}$, final ice-melting temperature; $T_h$, total homogenization temperature.
relationship with the mineralization. Thus the monzogranite porphyry should be the intrusion related to the mineralization. Generally, the mineralization age is about 4 Ma younger than the associated intrusion (Du et al., 2010). Furthermore, the closure temperature of zircon U–Pb age is higher than that of sericite Ar–Ar age (Harris, 1996), indicating the magma crystallization should be earlier than the alteration related to the mineralization. As discussed above, the ages of mineralization and monzogranite porphyry in the Chaganbulagen deposit should be the Early Cretaceous, and the mineralization occurred slightly later than emplacement of the monzogranite porphyry.

The Derbugan metallogenic belt is characterized by porphyry Cu–Mo and epithermal Pb–Zn–Ag, Ag, and Au–Cu deposits. For porphyry Cu–Mo deposits, Kang et al. (2014) obtained a molybdenite Re–Os age of 228.7 ± 3.1 Ma and granodiorite porphyry zircon U–Pb ages of 230.6 ± 2.8 Ma and 230.5 ± 4.4 Ma in the Badaguan porphyry deposit; Chen et al. (2010) reported a zircon LA-ICP-MS U–Pb age of 202 ± 5.7 Ma for the granodiorite porphyry and a Re–Os isochron age of 203.6 ± 4.6 Ma for molybdenite from the Taipingchuan porphyry deposit; Chen et al. (2011) defined a molybdenite Re–Os age of 177.6 ± 4.5 Ma, and Li et al. (2012) reported a molybdenite Re–Os isochron age of 178.0 ± 10 Ma in the Wunugetushan porphyry Cu–Mo deposit. For the epithermal Pb–Zn–Ag, Ag, and Au–Cu deposits, Xu et al. (2014) reported a zircon SHRIMP U–Pb age of 138.6 ± 2.3 Ma for a quartz porphyry intrusion, which was considered as being closely related to the mineralization of the Erentaolegai Ag deposit. Considering the ages obtained by us in this study and other geochronological data obtained by previous workers, we considered that important mineralization in the

![Fig. 10. Histograms of homogenization temperatures of fluid inclusions in the Chaganbulagen Pb–Zn–Ag deposit, showing average temperatures of various mineralization stages.](image)

![Fig. 11. Laser Raman spectra of fluid inclusions of the Chaganbulagen Pb–Zn–Ag deposit: (a) FIs of the early stage; (b) FIs of the early stage; (c) FIs of the middle stage; and (d) FIs of the late stage.](image)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Stage</th>
<th>Mineral</th>
<th>T_h(°C)</th>
<th>δD</th>
<th>δ18O_quartz</th>
<th>δ18O_water</th>
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<tr>
<td>NC-1</td>
<td>Early stage</td>
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<td>−153</td>
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<td>302</td>
<td>−162</td>
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<td>−13.4</td>
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<td>302</td>
<td>−145</td>
<td>−6.2</td>
<td>−13.0</td>
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<td>NC-12</td>
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<td>302</td>
<td>−138</td>
<td>−5.5</td>
<td>−12.3</td>
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<tr>
<td>NC-13</td>
<td>Early stage</td>
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<td>302</td>
<td>−153</td>
<td>−4.0</td>
<td>−10.8</td>
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<td>NC-14</td>
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<td>302</td>
<td>−154</td>
<td>−6.4</td>
<td>−13.4</td>
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<td>NC-21</td>
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<td>302</td>
<td>−127</td>
<td>−5.7</td>
<td>−12.5</td>
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<td>302</td>
<td>−131</td>
<td>−4.9</td>
<td>−11.7</td>
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<tr>
<td>C2-5801</td>
<td>Middle stage</td>
<td>Quartz</td>
<td>237</td>
<td>−134</td>
<td>−1.5</td>
<td>−11.0</td>
</tr>
<tr>
<td>NC-8</td>
<td>Late stage</td>
<td>Quartz</td>
<td>199</td>
<td>−142</td>
<td>−6.3</td>
<td>−17.9</td>
</tr>
</tbody>
</table>

Table 4: H and O isotopic compositions of the Chaganbulagen Pb–Zn–Ag deposit.
Derbugan metallogenic belt mainly occurred in three periods: (1) Late Triassic porphyry Cu–Mo mineralization; (2) Early Jurassic porphyry Cu–Mo mineralization; and (3) Early Cretaceous epithermal Pb–Zn–Ag, Ag, and Au–Cu mineralization. Of these three mineralization periods, the first two periods generated only porphyry Cu–Mo deposits, which occurred in a continental margin arc environment associated with the southward subduction of the Mongolia–Okhotsk oceanic slab beneath the Argun massif during the Late Triassic–Early Jurassic (Chen et al., 2010, 2011); the third mineralization period formed mainly epithermal Au–Ag–polymetallic deposits, which were related to volcanic–subvolcanic hydrothermal activity of the early Cretaceous (Li et al., 2015; Shuang, 2012; Wu et al., 2010).


The ore-forming fluids of the early stage in the Chaganbulagen deposit are characterized by medium–high homogenization temperatures (261–340 °C), low salinity (0.7–6.3 wt.% NaCl equiv.) and low density (0.65–0.81 g/cm³) (Fig. 15). Although petrographic studies did not indicate the presence of CH₄- and CO₂-bearing inclusions, the laser Raman spectroscopy revealed significant CO₂ and CH₄ peaks, and thus the early stage ore-forming fluid belongs to an H₂O–NaCl–CO₂ ± CH₄ system.

The fluids of the middle stage are characterized by relatively medium homogenization temperatures (200–265 °C), low salinity (0.5–5.7 wt.% NaCl equiv.), and low density (0.75–0.86 g/cm³) (Fig. 15). The laser Raman spectroscopy identified H₂O and CO₂ confirming that the middle stage fluid comprises an H₂O–NaCl ± CO₂ system.

The hydrothermal fluid of the late stage is of low temperature, very low salinity, and medium–low density, varying from 173 °C to 219 °C, 0.4 wt.% to 2.7 wt.% NaCl equiv., and 0.85 to 0.91 g/cm³, respectively (Fig. 15). The laser Raman spectroscopy identified only H₂O, confirming that the hydrothermal fluid of the late stage is composed of an H₂O–NaCl system.

Wu et al. (2010) reported microthermometric data of the Chaganbulagen deposit. They obtained homogenization temperatures of 264–329 °C, salinities of 2.7–6.9 wt.% NaCl equiv., and densities of 0.77–0.83 g/cm³ for 13 inclusions from the early stage; the homogenization temperatures, salinities, and densities from 12 inclusions of the middle stage are 231–262 °C, 2.9–6.6 wt.% NaCl equiv., and 0.84–0.86 g/cm³, respectively; and the homogenization temperatures, salinities, and densities from five inclusions of the late stage vary from 176 °C to 208 °C, 2.2 to 4.0 wt.% NaCl equiv., and 0.89 to 0.91 g/cm³, respectively.

The above microthermometric results obtained by us as well as previous researchers indicate that the fluids of the early and middle stages in the Chaganbulagen deposit were medium–high temperature and low salinity and density, and generally belonged to an H₂O–NaCl–CO₂ ± CH₄ in composition; and that the fluids of the late stage are characterized by low temperature, very low salinity, medium–low density, and lack of CO₂ and CH₄. From the early, through the middle, and to the late stage, the temperature and salinity of the hydrothermal fluids gradually decreased but fluid density gradually increased (Fig. 15).

6.3. Sources of ore-forming fluid and materials

6.3.1. Source of ore-forming fluid

The δD values range from −166‰ to −127‰, and the δ¹⁸Owater values vary from −17.9‰ to −10.8‰. The H–O isotope data of the early, middle, and late stage all are plotted near the meteoric water line, but far away from the magmatic water field in the δ¹⁸Owater–δD diagram (Fig. 12), implying that the hydrothermal fluids consist dominantly of meteoric water. As abovementioned, the ore-forming fluids of the Chaganbulagen deposit are characterized by low salinity; from the early stage to the late stage, the temperature and salinity of the

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**Table 5**

Sulfur and lead isotope compositions of sulfides from the Chaganbulagen deposit.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Stage</th>
<th>Mineral</th>
<th>δ²⁸Sᵥ₋ᵥᵥ (%)</th>
<th>δ¹⁸O_H₂O (%)</th>
<th>²⁰⁸Pb/²⁰⁴Pb (2σ)</th>
<th>²⁰⁶Pb/²⁰⁴Pb (2σ)</th>
<th>²⁰⁸Pb/²⁰⁴Pb (2σ)</th>
<th>μ</th>
<th>ω</th>
</tr>
</thead>
<tbody>
<tr>
<td>NC-10</td>
<td>Middle</td>
<td>Pyrite</td>
<td>2.9</td>
<td>1.4</td>
<td>18.507 ± 0.004</td>
<td>15.536 ± 0.003</td>
<td>38.269 ± 0.008</td>
<td>9.3</td>
<td>34.8</td>
</tr>
<tr>
<td>NC-44</td>
<td>Middle</td>
<td>Pyrite</td>
<td>1.9</td>
<td>0.4</td>
<td>19.037 ± 0.002</td>
<td>15.593 ± 0.002</td>
<td>38.945 ± 0.004</td>
<td>9.4</td>
<td>35.2</td>
</tr>
<tr>
<td>NC-46</td>
<td>Middle</td>
<td>Pyrite</td>
<td>2.3</td>
<td>0.8</td>
<td>18.983 ± 0.002</td>
<td>15.473 ± 0.004</td>
<td>38.954 ± 0.010</td>
<td>9.2</td>
<td>34.3</td>
</tr>
<tr>
<td>NC-47</td>
<td>Middle</td>
<td>Pyrite</td>
<td>1.4</td>
<td>−0.1</td>
<td>18.316 ± 0.003</td>
<td>15.537 ± 0.002</td>
<td>38.132 ± 0.005</td>
<td>9.4</td>
<td>35.2</td>
</tr>
<tr>
<td>NC-4</td>
<td>Middle</td>
<td>Sphalerite</td>
<td>4.1</td>
<td>3.7</td>
<td>18.339 ± 0.002</td>
<td>15.570 ± 0.002</td>
<td>38.218 ± 0.004</td>
<td>9.4</td>
<td>35.7</td>
</tr>
<tr>
<td>NC-20</td>
<td>Middle</td>
<td>Sphalerite</td>
<td>3.3</td>
<td>2.9</td>
<td>18.315 ± 0.002</td>
<td>15.530 ± 0.001</td>
<td>38.160 ± 0.003</td>
<td>9.3</td>
<td>35.3</td>
</tr>
<tr>
<td>NC-47</td>
<td>Middle</td>
<td>Sphalerite</td>
<td>3.7</td>
<td>3.3</td>
<td>18.345 ± 0.002</td>
<td>15.569 ± 0.002</td>
<td>38.244 ± 0.004</td>
<td>9.4</td>
<td>35.8</td>
</tr>
<tr>
<td>NC-4</td>
<td>Middle</td>
<td>Galena</td>
<td>1.7</td>
<td>4.1</td>
<td>18.302 ± 0.002</td>
<td>15.509 ± 0.002</td>
<td>38.218 ± 0.007</td>
<td>9.3</td>
<td>35.4</td>
</tr>
<tr>
<td>NC-46</td>
<td>Middle</td>
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<td>2.0</td>
<td>4.4</td>
<td>18.317 ± 0.002</td>
<td>15.535 ± 0.002</td>
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</tr>
<tr>
<td>NC-81</td>
<td>Middle</td>
<td>Galena</td>
<td>2.5</td>
<td>4.9</td>
<td>18.329 ± 0.002</td>
<td>15.555 ± 0.001</td>
<td>38.181 ± 0.003</td>
<td>9.4</td>
<td>35.5</td>
</tr>
</tbody>
</table>

Notes: μ = ²⁰⁶Pb/²⁰⁴Pb, ω = ²³⁵Pb/²³⁴Pb, the age for calculating the values of μ and ω is 138 Ma.
hydrothermal fluids gradually decreased but fluid density gradually increased. These phenomena are usually linked with water–rock interaction during meteoric water convection. The fluids of the early and middle stages with relatively high temperature developed fluid–rock reaction with volcanic rocks in circulation process, elevating not only the salinity but also the $\delta^{18}O$ value of the ore-forming fluids. The late stage fluids are difficult to react with country rocks because of the relatively low fluid temperature, more retaining signatures of the original meteoric water such as very low salinity and lower $\delta^{18}O$ value. We, therefore, conclude that the ore-forming fluids of the deposit were primarily sourced from circulating meteoric water although some researchers believed that mixing between magmatic and meteoric water may be a plausible explanation for the low $\delta$D and $\delta^{18}O$ values of the ore-forming fluids (Li et al., 2014; Ouyang et al., 2014; Rye, 1993; Taylor, 1974; Wu et al., 2010; Zhai et al., 2013).

6.3.2. Sources of sulfur and ore-forming metals

The isotope composition of sulfides from hydrothermal deposits is a function of total S isotopic compositions ($\delta^{34}S$), oxygen fugacity ($f_O$), pH, ionic strength, and temperature. Sulfur isotopic composition of hydrothermal sulfides depends not only on the $\delta^{34}S$ value of source materials, but also on the physicochemical condition of the ore-forming fluid (Ohmoto, 1972). Therefore, the S sources must be determined based on the total S isotopic signature of the hydrothermal fluids when the sulfides were deposited. However, when the hydrothermal fluids are dominated by $H_2S$ with low oxygen fugacity and low pH values, the relationship $\delta^{34}S_{H_2S} \approx \delta^{34}S_{min} \approx \delta^{34}S_{pyrite}$ holds in the equilibrium state (Wu et al., 2014). The sulfides of the Chaganbulagen deposit during the mineralization are dominated by pyrite, sphalerite, galena, and chalcopyrite, and no sulfate minerals have been detected. Therefore, $H_2S$ is dominated in the hydrothermal system during the mineralization of the Chaganbulagen deposit, and pyrite, sphalerite, galena, and chalcopyrite formed in the environment with low $f_O$ and low pH values. According to Table 5, the average $\delta^{34}S$ values of pyrite, sphalerite, and galena from the Chaganbulagen deposit are 2.1‰, 4.1‰, and 2.1‰, respectively. The basic sequence ($\delta^{34}S_{py} < \delta^{34}S_{sp}$ and $\delta^{34}S_{sp} > \delta^{34}S_{gn}$) is inconsistent with the $\delta^{34}S$ enrichment condition of $\delta^{34}S_{py} > \delta^{34}S_{sp} > \delta^{34}S_{gn}$ at isotopic equilibrium, indicating that the S isotopes of pyrite and sphalerite and galena had not reached equilibrium but those of sphalerite and galena had reached equilibrium (Ohmoto, 1986). Under the circumstance, $\delta^{34}S_{H_2S}$ values of sphalerite and galena can represent the total S isotopic compositions of hydrothermal system (Xu et al., 1993). The $\delta^{34}S_{H_2S}$ values were calculated with the equation $\delta^{34}S_{H_2S} = \delta^{34}S_{i} - A_{i} (10^{6} \times T^{-3})$, where $i$ stands for different sulfides; $A_{i}$ value is 0.4 for pyrite, 0.1 for sphalerite, and −0.63 for galena, respectively; $T$ is the temperature in Kelvin (Xu et al., 1993); and the average of fluid inclusion temperatures from the middle stage was used to calculate the $\delta^{34}S_{H_2S}$ value. The $\delta^{34}S_{H_2S}$ values of the Chaganbulagen deposit vary from 2.9‰ to 3.7‰ for sphalerite and 4.1‰ to 4.9‰ for galena (Table 5). The $\delta^{34}S$ value is slightly higher than that of the meteorite sulfur but consistent with that of intermediate–felsic magmatic rock (Ohmoto and Rye, 1979), indicating that the sulfur of the Chaganbulagen deposit was mainly derived from intermediate–felsic magmatic source.

The Pb isotope data are useful geochemical tracers and can indicate crustal evolution, a source of lead, and thus deposit genesis (Lu et al., 2000). In the $^{206}Pb/^{204}Pb$ versus $^{207}Pb/^{204}Pb$ diagram (Fig. 14a), the Pb isotope data obtained by us in this study are projected primarily toward the area between the orogenic belt and the mantle evolution lines; in the $^{208}Pb/^{204}Pb$ versus $^{206}Pb/^{204}Pb$ diagram (Fig. 14b), almost all of the Pb isotope data are plotted in the field of the orogenic belt evolution line. The $\mu$ values of metal sulfides from the Chaganbulagen deposit are between 9.2 and 9.4, which are obviously higher than the range of 8–9 in the mantle and lower than 9.38 in the upper crust (Doe and...
6.4. Mechanism of mineral deposition and genetic type

6.4.1. Mechanism of mineral deposition

The following mechanisms may cause precipitation of metal chloride complexes from a solution (Barnes, 1979; Chi and Xue, 2011; Ouyang et al., 2014; Ramboz et al., 1982; Robb, 2004): (1) increased concentration of sulfur in solution, (2) decrease in chloride concentration, (3) temperature decrease, (4) pressure decrease, (5) phase separation, (6) fluid mixing/dilution, and (7) pH and Eh shifts caused by fluid/rock interaction.

There are only low-salinity liquid inclusions in the Chaganbulagen deposit, excluding fluid immiscibility, boiling, or phase separation caused by pressure decrease. The low H–O isotope values, which are close to the meteoric water line but far away from the magmatic water field, as well as low salinity and simple inclusion type, indicate that the ore-forming fluid was dominantly derived from meteoric water and that fluid mixing between meteoric water and magmatic water can be ignored. The ore-forming fluids of the early and middle stages have higher salinity and δ18Owater value than the hydrothermal fluids of the late stage, implying that water–rock interaction occurred during meteoric water convection, increasing the concentration of ore-forming elements such as S, Pb, Zn, and Ag. Therefore, we propose that temperature decrease of the ore-forming fluids is the dominant mechanism for the deposition of ore-forming materials in the Chaganbulagen deposit. The temperature decrease can not only reduce the solubility of the ore-forming substance but also destroy the stability of the complexes (Brimhall and Crear, 1987; Crear and Barnes, 1976), giving rise to the precipitation of metals from the hydrothermal system.

6.4.2. Genetic type

The Chaganbulagen deposit was generally defined as medium–low temperature hydrothermal vein-type deposit based on mineralization temperatures of 176–367 °C obtained by fluid inclusion microthermometry (Wu et al., 2010; Zhao and Zhang, 1997) or low-sulfidation epithermal deposit according to mineral assemblage, wall-rock alteration type, and mineralization environment (Qi et al., 2005; Shuang, 2012; Zhao and Wu, 2002). For the Pb–Zn–Ag deposits in the Derbugan metallogenic belt, northern Great Xing’an Range, previous studies showed that the Pb–Zn–Ag deposits were located in the transition zone of the Mesozoic volcanic basin and pre-Mesozoic basement uplift (Wu et al., 2010); the Pb–Zn–Ag mineralization was related to volcanic–subvolcanic hydrothermal activity, and mineralization age should be Early Cretaceous (Li et al., 2014; Mao et al., 2013); and the deposits were formed either in a post-orogenic extensional regime, which resulted from the collapse of the Mongol–Okhotsk orogenetic belt after the closure of the Mongolia–Okhotsk Ocean (Fan et al., 2003; Ge et al., 2007; She et al., 2009, 2012; Wang et al., 2015), or in a back-arc extensional environment induced by sequential slab break off, retreat, and roll back of the Pacificocean (Izanagi) plate (Pirajno and Zhou, 2015; Wang et al., 2006). Our new data from the fluid inclusion study in this work indicate that the ore-forming fluids of the Chaganbulagen deposit are characterized by moderate temperature, low salinity and density, and meteoric water. The paleodepth of the Chaganbulagen deposit is 0.87–1.80 km. The ore minerals of the deposit are mainly galena, sphalerite, pyrite, chalcopyrite, silver-bearing minerals, pyrrhotite, and arsenopyrite; the main alteration types include silicification, sericitization, hydrodynamicization, carbonation, chloritization, and kaolinization. Both geological and geochemical characteristics above as well as adular discovered by Shuang (2012) are similar to those of many low-sulfidation epithermal deposits in the word proposed by Heald et al. (1987) and Hedenquist (1994). Thus, we propose that the Chaganbulagen deposit is a low-sulfidation epithermal deposit.

7. Conclusions

(1) The sericite sample yielded a 40Ar/39Ar plateau age of 138 ± 1 Ma and an isochron age of 137 ± 3 Ma, and the zircon LA-ICP-MS U–Pb age of the monzogranite porphyry was 143 ± 2 Ma.

(2) The ore-forming fluids of the Chaganbulagen deposit are characterized by moderate temperature, low salinity and density, and an H2O–NaCl ± CO2 ± CH4 composition.

(3) The ore-forming fluids of the Chaganbulagen deposit consist dominantly of meteoric water, and the ore-forming materials mainly came from the Mesozoic magma.

(4) The Chaganbulagen deposit is a low-sulfidation epithermal deposit, and the temperature decrease is the dominant mechanism for the deposition of ore-forming materials.

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References


