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A 20 m.y. long-lived successive mineralization in the giant Dahutang W–Cu–Mo deposit, South China

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ABSTRACT

The connection between prolonged granitic magmatism and the formation of giant tungsten (W) polymetallic deposits has long been disputed. In this study, we present 6 mica Ar–Ar plateau ages and 22 molybdenite Re–Os model ages data on the newly discovered giant Dahutang W–Cu–Mo deposit in South China, which is one of the largest W deposits in the world. New and published zircon U–Pb, mica Ar–Ar, and molybdenite Re–Os age data reveal that the Mesozoic Dahutang magmatism and mineralization occurred in two major periods: (1) the Late Jurassic (ca. 153–147 Ma), forming the hydrothermal breccia, large wolframite-bearing quartz vein, and scheelite-dominated disseminated/veinlet type orebodies, which is mainly associated with the emplacement of porphyritic biotite granite; (2) the Late Jurassic to Early Cretaceous (ca. 146–130 Ma), forming the Cu–Mo–W ± Sn mineralization overprinting the Late Jurassic W–Mo ± Cu orebodies, which is mainly related to the successively emplacement of the Early Cretaceous granites. We suggest that continuous accumulation of mineralization for a long period of time (151–130 Ma) have contributed to the formation of the giant Dahutang deposit.

1. Introduction

The possible correlation between the duration of magmatism and the size of hydrothermal deposits has been a long and controversial topic, especially when super-large/giant-scale mineralization is concerned (Cathles et al., 1997; Mercer et al., 2015; Buret et al., 2016; Rezeau et al., 2016). What is often neglected in these discussions is the accurate occurrence of multi-stage magmatism and mineralization, which can substantially enlarge and enrich the ore deposits.

The recently discovered Dahutang W–Cu–Mo deposit in South China is a giant deposit, hosting an estimated resource of up to 1.1 million tonnes (Mt) of WO_3, 0.65 Mt Cu and 0.08 Mt Mo (Mao et al., 2013; Zhang et al., 2014; Jiang et al., 2015). Dating of the ore-forming granites yielded a very wide age range from ca. 151.7 Ma–130.3 Ma (Huang and Jiang, 2012, 2013, 2014; Xiang et al., 2012; Mao et al., 2015; Ye et al., 2016; Zhang et al., 2016), while the mineralization ages reported mainly cluster around 150 Ma (Xiang et al., 2013; Zhang et al., 2016), 143 Ma (Feng et al., 2012; Jiang et al., 2015) and 139 Ma (Mao et al., 2013; Zhang et al., 2017). How the wide magmatic age range and the seemingly discrete mineralization episodes are related is poorly understood, which leads to different interpretations of the ore-forming processes. In this contribution, based on detailed documentation of the mineral assemblages and alteration/mineralization paragenesis, we have identified four distinct magmatic-metallogenic phases at Dahutang. Timing of these four magmatic-metallogenic phases is newly constrained by our new molybdenite Re–Os and mica Ar–Ar dating, augmented by the published radiometric ages. We propose that the prolonged and multi-stage magmatism/mineralization has contributed to the giant-scale mineralization at Dahutang.

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2. Regional and deposit geology

South China has long been recognized as one of the world's major W mineral provinces (Hsu, 1943), boasting a resource of over 5.6 Mt of WO_3 (Shen et al., 2015; USGS, 2017). The newly discovered Dahutang W–Cu–Mo deposit comprises the North Dahutang, Central Dahutang and South Dahutang ore blocks (Supplementary Fig. A.1). At Dahutang, the low-grade metamorphic rocks of the Neoproterozoic Shuangqiaoshan Group were intruded by the Neoproterozoic Jiuling biotite granodiorite (zircon U–Pb age: 819 ± 9 Ma, Li et al., 2003). Multiple phases of Late Mesozoic granitic intrusions, closely mineralization-related, were emplaced as stocks and dikes into the Neoproterozoic granodiorite and Neoproterozoic Shuangqiaoshan Group metamorphic rocks. Field geological and petrographic observations suggest that the Late Mesozoic granites consist of porphyritic biotite granite (G1), fine-grained biotite granite (G2), porphyritic muscovite granite (G3), granite porphyry (G4), fine-grained muscovite granite (G5), granite porphyry (G6), medium- to fine-grained muscovite granite (G7), porphyritic two-mica granite (G8), and fine-grained two-mica granite (G9).

The W–Cu–Mo mineralization at Dahutang mainly occurs around the intrusive contacts of the Late Mesozoic granite stocks. Major ore types include disseminated/veinlets, large wolframite-bearing-quartz veins, and hydrothermal breccias. Previous field observations and molybdenite Re–Os dating suggest no discernible spatial or temporal relationships among the three types of mineralization. Ore minerals at Dahutang include mainly scheelite, wolframite, chalcopyrite, and molybdenite, with minor cassiterite, sphalerite, bornite, arsenopyrite, pyrite, and stannite. Gangue minerals at Dahutang include predominately quartz, with minor mica, feldspar, fluorapatite, fluorite, chlorite, and calcite.

3. Analytical methods

3.1. Mica $^{40}Ar/^{39}Ar$ dating

Five muscovite and one biotite samples for $^{40}Ar/^{39}Ar$ dating were collected from drill cores and underground tunnels of the Central and South Dahutang ore blocks. Locations and descriptions of these samples are given in Supplementary Table B1. One biotite (DH-31) and two muscovite samples (DH-42, DH-126) were conducted at the Key Laboratory of Orogenic Belts and Crustal Evolution, Peking University, the other three muscovite samples (DH-02, DH-120 and DH-94) were performed at the Western Australian Argon Isotope Facility of Curtin University. The $^{40}Ar/^{39}Ar$ analytical methods as follows:

$^{40}Ar/^{39}Ar$ analytical method at Curtin University: The samples were crushed to 0.10–0.20 mm size fraction. The muscovite was carefully handpicked under a binocular microscope, and then washed in an ultrasonic bath using ultrapure water. Purity of the mineral separates exceeds 99%. The selected muscovite grains were loaded into a small aluminum disc (1.9 cm in diameter, 0.3 cm deep), which was Cd-shielded to minimize nuclear interference reactions. The samples were irradiated for 40 h in the US Geological Survey nuclear reactor (Denver, USA) in central position over two irradiations. The $^{40}Ar/^{39}Ar$ analysis was performed at the Western Australian Argon Isotope Facility of Curtin University. The samples were step-heated using the 110 W Spectron Laser Systems, with a continuous Nd-YAG (IR; 1064 nm) laser rastered over the sample during 1 min to ensure a homogenously distributed temperature. The gas was purified in a stainless steel extraction line using three SAES AP10 getters and a liquid nitrogen condensation trap. Argon isotopes were measured in the static mode using a MAP 215-50 mass spectrometer (resolution: ~500; sensitivity: 4×10^{-14}). The age was calculated using the recommended Fish Canyon sanidine (FCs) age (28.294 ± 0.037 Ma; Jourdan and Renne, 2007; Merle et al., 2009). Mean J-value computed from the monitor mineral is 0.01055080 ± 0.00000422. The correction factors for interfering isotopes were $(^{39}Ar/^{37}Ar)_{ca} = 7.6 \times 10^{-4}$ (± 12%), $(^{36}Ar/^{37}Ar)_{ca} = 2.3 \times 10^{-4}$ (± 1%), $(^{40}Ar/^{39}Ar)_{k} = 7.3 \times 10^{-4}$ (± 13%). Our criteria for the plateau determination are as follows: The plateaus must include at least 70% of ^{39}Ar. The plateau should also be distributed over a minimum of three consecutive steps agreeing at 95% confidence level, and satisfying a probability of fit of at least 0.05. Plateau ages are given at the 2σ level and are calculated using the mean of all the plateau steps, each weighted by the inverse variance of their individual analytical error.

$^{40}Ar/^{39}Ar$ analytical method at Peking University: The samples were crushed to 0.18–0.28 mm size fraction. About 20–60 mg muscovite or biotite was packed in pure aluminous foil for each sample. The foils were placed in evacuated quartz tubes and irradiated in position B4 of 49-2 Nuclear Reactor at the Institute of Chinese Atomic Energy for 24 h, with an irradiation flux of 2.65×10^{13} fast neutrons/cm^2. Neutron flux variation (J) was measured using the standard mineral Zhoukoudian biotite (ZBH-25; 132.7 Ma). After irradiation, the samples were heated to total fusion in a tantalum furnace, during 10–16 stepwise-heating stages. Extracted gas was analyzed using a RGA10 mass spectrometer at the Key Laboratory of Orogenic Belts and Crustal Evolution, Peking University. Argon isotope data were corrected for system blanks, mass discrimination, and interfering neutron reactions with Ca and K. Correction factors are: $(^{36}Ar/^{37}Ar)_{Ca} = 0.000271$, $(^{39}Ar/^{37}Ar)_{Ca} = 0.000652$, and $(^{40}Ar/^{39}Ar)_{K} = 0.00703$. The analyzed argon isotope data were corrected using the $^{40}Ar/^{39}Ar$ Dating 1.2 software developed by the Key Laboratory of Orogenic Belts and Crustal Evolution, Peking University. Plateau and isochron ages were calculated using the Isoplot 3.0 software of Ludwig (2003).

3.2. Molybdenite Re–Os dating

Twenty-two molybdenite samples for the Re–Os dating have been systematically collected from drill cores and underground tunnels of the North, Central and South Dahutang ore blocks. Sampling locations/depths and mineralogical features of the samples are summarized in Supplementary Table B.2.

Molybdenite grains were magnetically separated and then hand-picked under a binocular microscope to obtain a > 99% purity. Re–Os isotope analyses were conducted at the Key Laboratory of Mineralogy and Metallogeny, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences (GIGCAS). The Carius tube method was used for the dissolution of molybdenite and equilibration of samples, and tracer ^{185}Re and ^{190}Os at 225–230 °C for 24 h (Sun et al., 2001, 2010; Li et al., 2011; Leng et al., 2012). From the opened tube, an approximate amount of the supernatant was transferred to a 30 ml quartz beaker, heated to dryness at 150 °C, and then 0.5 ml HNO_3 was added and dried-down. This step was repeated twice to ensure the removal of Os as OsO_4, and finally diluted to 10 ml (using 2% HNO_3) for the Re isotope analysis by ICP–MS. The supernatant was poured into a 50 ml distillation flask redesigned after Sun et al. (2001), and distilled at 110 °C for 20 min for Os. 5 ml of H_2O was used to extract the Os from a water–ice bath, and the solution was used for ICP–MS measurement of the Os isotopes.

4. Results

A total of six mica Ar–Ar ages and twenty-two molybdenite Re–Os model ages were obtained in this study. Summary of the Ar–Ar data of the biotite and muscovite from Dahutang are given in Supplementary Tables B.3-1–3-2. Plots of the $^{40}Ar/^{39}Ar$ age spectra for the Dahutang deposit are illustrated in Fig. A.2. Published and new granite ages (zircon and monazite U–Pb, mica $^{40}Ar/^{39}Ar$ dating) and molybdenite Re–Os model ages are also compiled and summarized in Supplementary Tables B.4–B.5 and illustrated in Fig. 1.