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In situ chemical and isotopic analyses and element mapping of multiple-generation pyrite: Evidence of episodic gold mobilization and deposition for the Qiucun epithermal gold deposit in Southeast China

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ABSTRACT

Gold deposits are often the result of complex mineralization and remobilization processes. Interpretation of bulk geochemical and sulfur isotope data of the gold deposits is frequently hampered by complex zoning in pyrite, which calls for in situ determination of geochemical and sulfur isotope composition of sulfide minerals. The Qiucun deposit is a good representative of epithermal gold deposits in the Mesozoic Coastal Volcanic Belt of southeastern China. It represents a complex mineralization history, comprising three hydrothermal stages: (I) early stage of pyrite-quartz-chalcedony; (II) main ore stage of quartz-polymetallic sulfide; and (III) post-ore stage of quartz-carbonate. Detailed backscattered electron imaging (BSE) and in situ trace element and sulfur isotope analyses using laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) and LA-multicollector (MC)-ICP-MS were applied to reveal the gold mineralization and remobilization history of this deposit. Four texturally distinct generations of pyrite were identified, all of which host invisible gold: Py_{1a} and Py_{1b} in Stage I and Py_{2a} and Py_{2b} in Stage II. A detailed study of the texture, chemistry, and sulfur isotopic composition as well as hydrothermal evolution of auriferous pyrite from the Qiucun deposit revealed the behavior of gold in the course of pyrite evolution. Pyrite of Stages I and II contains invisible gold, whereas later-stage visible native gold and re-enrichment in invisible gold is associated with alteration rims around the primary pyrite grains. Py_{1a} is rich in silicate inclusions, enriched in Co and Ni, and depleted in As and Au relative to later pyrite generations. This redistribution is attributed to the alteration of biotite in the sub-volcanic host rocks that effectively destabilized gold in the ore fluid during Py_{1a} deposition. Py_{1b} and Py_{2a} show oscillatory zoning with bright bands having elevated As and Au contents. The oscillatory zoning is interpreted to reflect pressure fluctuations and repeated local fluid boiling around the pyrite crystals. These three pyrite generations (Py_{1a}, Py_{1b}, Py_{2a}) record a narrow range of $\delta^{34}S_{V-CDT}$ values between –3.6 and 4.6‰, consistent with a magmatic sulfur source. Gold and some trace elements (As, Ag, Sb, Pb, Tl, and Cu) that were initially incorporated into Py_{2a} became partially exsolved and remobilized during the replacement of porous and invisible gold-rich Py_{2b}. This replacement was likely due to coupled dissolution and re-precipitation reactions triggered by oxidation of the mineralizing fluids. Fluid oxidation is further supported by a general decrease trend of $\delta^{34}S_{V-CDT}$ from Py_{2a} (–3.2 to 4.6‰) to Py_{2b} (–15.2 to –2.3‰). Last, previously formed auriferous pyrite underwent post-mineralization fracturing, causing local pulverization of pyrite. Thus, newly created porosity facilitated fluid circulation, hydrothermal alteration of the pyrite, and remobilization of invisible gold, which re-precipitated with pyrite in the form of electrum as small inclusions or as larger grains within fractures. Our study emphasizes that pressure-driven hydrothermal processes play a vital role in the initial enrichment and re-concentration of gold and some other trace metals during episodic deposition, replacement, and hydrothermal alteration of gold-bearing pyrite in epithermal gold deposits, ultimately forming visible gold and high-grade ore shoots as exemplified by the Qiucun deposit.

Keywords: Pyrite formation and replacement, sulfur isotopes, gold remobilization, epithermal gold mineralization