

Age and source constraints for the giant Muruntau gold deposit, Uzbekistan, from coupled Re-Os-He isotopes in arsenopyrite

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ABSTRACT

The Muruntau gold deposit, Uzbekistan, is one of the largest gold deposits known worldwide, but its origin remains controversial. We used Re-Os arsenopyrite geochronology to precisely determine the age of main-stage gold mineralization at Muruntau to be 287.5 ± 1.7 Ma, which overlaps the emplacement of proximal post-tectonic granitoid magmatism. Additionally, we suggest that arsenopyrite growth may have occurred over an interval of at least 2 m.y. Os initial ratios derived from arsenopyrite were coupled with He isotopic data from fluid inclusions within arsenopyrite to constrain the source of ore metals and fluids. Muruntau arsenopyrite yields relatively unradiogenic initial Os (0.37 ± 0.27) and elevated $^3\text{He}/^4\text{He}$ ratios ($0.23\text{--}0.33R_a$) relative to purely crustal Os-He reservoirs. These data suggest the presence of a mantle-derived component in the ore system that was probably introduced during the generation of the granitoid magmas. These new timing and source constraints provide important new insight into the generation of this giant gold deposit, and they necessitate reexamination of genetic models for Muruntau and potentially other giant “orogenic gold” deposits worldwide.

Keywords: rhenium, osmium, arsenopyrite, geochronology, helium, Muruntau.

INTRODUCTION

“Orogenic gold deposits” (Groves et al., 1998) are a class of deposit represented episodically in the geologic record since ca. 3.2 Ga that occur in broad coincidence with the production of juvenile continental crust (Goldfarb et al., 2001; Groves et al., 2005). The exact nature of this association is unknown, though mantle-to-crust heat transfer in crust-forming environments is considered necessary for providing energy to drive gold-bearing hydrothermal systems (Goldfarb et al., 2001). A major Phanerozoic crust-forming episode occurred during development of the Central Asian Altiid tectonic collage, which involved volcano-plutonic island-arc development, formation of multiple subduction-accretion complexes (Yakubchuk et al., 2002), and emplacement of voluminous post-tectonic granitoid intrusions of predominately juvenile character (Jahn et al., 2000). Collage development generated a vast metallogenic wealth, including one of the richest gold provinces on Earth, the late Paleozoic Tien Shan orogen, which is an extensive accretionary complex that formed after back-arc basin closure in the early mid-Carboniferous (Yakubchuk et al., 2002). Multiple world-class orogenic gold deposits occur within the southern portion of the orogen (southern Tien Shan orogen). Here, we evaluate a possible relationship between juvenile crust

production and orogenic gold mineralization at the Muruntau gold deposit, Uzbekistan, by precise determination of the timing of a major gold mineralizing event, age comparisons with regional magmatic events, and new isotopic constraints on the source of metal (Os) and fluid (He) components of the ore system.

GEOLOGY AND EXISTING GEOCHRONOLOGY OF THE MURUNTAU GOLD DEPOSIT

The giant Muruntau deposit has immense gold reserves (>5100 metric tons), and it exhibits characteristics typical of most orogenic gold deposits: it is hosted in a metamorphosed terrane and is structurally controlled (Drew et al., 1996) and derived from aqueous-carbonic, low-salinity fluids (Graupner et al., 2001). Gold mineralization and associated hydrothermal alteration are confined to deformed and metamorphosed Cambrian-Ordovician shales and carbonaceous siltstones of the Variegated Besapan unit (Drew et al., 1996), but they also exhibit a close spatial relationship to granitoid plutons (e.g., the Murun granite and Saradin granodiorite; Kostitsyn, 1996), quartz dioritic to syenitic dikes, and lamprophyre dikes (Drew et al., 1996; Graupner et al., 2006). The location of the Muruntau deposit in the thermal aureole of an unexposed pluton (Drew et al., 1996) and the presence of

hydrothermally altered dikes along fault zones within the ore field (Kostitsyn et al., 1996) imply a relationship between granitoid magmatism and the ore-forming process (e.g., Drew et al., 1996; Yakubchuk et al., 2002). Ages of 286–293 Ma likely represent the main stage of granitoid magmatism in the vicinity of Muruntau, as defined by Rb-Sr whole rock + mineral isochron (Sardarin and Murun plutons; Kostitsyn, 1996) and reversely discordant U-Pb sensitive high-resolution ion microprobe (SHRIMP) zircon analyses from the North Tamdinskii granite, located 31 km northwest of Muruntau (Kempe et al., 2004). Existing constraints for the timing of gold mineralization (Kostitsyn, 1994; Kempe et al., 2001; Wilde et al., 2001) indicate a hiatus of at least ten million years (m.y.) between cessation of the main period of granitoid magmatism and the onset of gold mineralization. These age data have, in part, led to diverse explanations for the origin of the Muruntau deposit (e.g., Wilde et al., 2001; Graupner et al., 2006).

Recently, precise dates for gold mineralization have been obtained by Re-Os geochronology of arsenopyrite (Arne et al., 2001; Morelli et al., 2005). To determine the timing of gold mineralization at Muruntau, we analyzed three coarsely crystalline arsenopyrite samples from the open pit. The samples include one (9824) from a subvertical stockwork quartz veinlet from the first ore body, and two (18 and 20A) from steeply dipping quartz-arsenopyrite veinlets from the Southern fault zone (Fig. 1). Arsenopyrite crystallization is, at least in part, genetically related to gold mineralization at Muruntau, as evidenced by the presence of gold inclusions within arsenopyrite in sample 9824. Rare Bi-Te phases constitute the only other inclusions detected by optical and electron microscopy.

Re-Os RESULTS

Arsenopyrite was mechanically separated and crushed using agate tools; analytical protocols for Re-Os analysis are described elsewhere (Morelli et al., 2005). All three arsenopyrite samples yielded well-fitted individual Re-Os isochrons and precise model 1 miner-

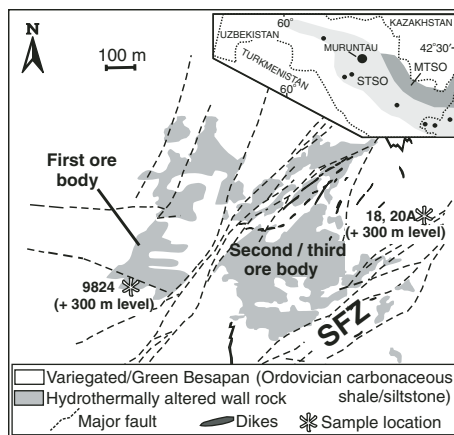


Figure 1. General geology of Muruntau gold deposit within open pit, showing location of arsenopyrite samples used in study (STSO—southern Tien Shan orogen, MTSO—Middle Tien Shan orogen, SFZ—Southern fault zone).

alization ages: 288.6 ± 5.6 Ma (9824; mean square of weighted deviates [MSWD] = 1.1, initial $^{187}\text{Os}/^{188}\text{Os} = -0.2 \pm 1.9$ [Os_i]), 290.4 ± 4.5 Ma (18; MSWD = 0.81, Os_i = 0.14 ± 0.41), and 296.4 ± 6.1 Ma (20A; MSWD = 1.5, Os_i = 0.07 ± 0.59 ; Fig. 2A). All ages overlap within 2σ uncertainties and clearly indicate that arsenopyrite formation and associated gold mineralization occurred at ca. 290 Ma. However, internal age structure may be revealed through collective regressions of multiple samples. Whereas collective regression of all analyses of samples 9824 and 18 yields a precise model 1 isochron age of 287.5 ± 1.7 Ma (MSWD = 0.98, Os_i = 0.37 ± 0.27 ; Fig. 2B), addition of sample 20A fails to result in a model 1 solution (Fig. 2C). This difference is evident when the data are plotted to avoid use of ^{188}Os , the precise quantification of which is challenging in these samples for which blank ^{188}Os contributes significantly to ^{188}Os (Stein et al., 2000; Morelli et al., 2005). Samples 9824 and 18 collectively yield a precise model 1 age of 287.7 ± 0.7 Ma (MSWD = 1.0) on a plot of radiogenic ^{187}Os ($^{187}\text{Os}^*$) versus ^{187}Re (e.g., Stein et al., 2000), whereas sample 20A yields a distinguishable model 1 age of 294.3 ± 3.9 Ma (MSWD = 0.82; Fig. 2D). This may suggest that arsenopyrite 20A crystallized at least two million years prior to samples 18 and 9824, but confirmation of this result will require additional high-precision Re-Os data.

TIMING OF GOLD MINERALIZATION AT MURUNTAU

A major problem in understanding the generation of orogenic gold deposits generally is the lack of reliable age determinations for mineralization (Groves et al., 2003). The new dates presented here, which are the completed data set of

a previously reported preliminary age (Morelli et al., 2004), clearly demonstrate the utility of the Re-Os method for directly dating sulfide minerals associated with gold mineralization, and also they reveal the shortcomings of other methods. Gold mineralization at Muruntau was previously determined to be ca. 275 Ma, based on Rb-Sr data from hydrothermal alteration minerals (Kostitsyn, 1994) and Sm-Nd scheelite analyses (Kempe et al., 2001), or as young as ca. 245 Ma from $^{40}\text{Ar}/^{39}\text{Ar}$ dates of vein selvage sericite (Wilde et al., 2001). The Re-Os arsenopyrite ages show that sulfide and gold mineralization occurred at ca. 290 Ma, 15–20 m.y. older than dates based on hydrothermal silicate/tungstate gangue minerals, and that it temporally overlapped regional late-tectonic magmatism (Fig. 3). The small but statistically significant Re-Os age difference between arsenopyrite samples may also indicate that hydrothermal activity occurred over at least 2 m.y., suggesting a protracted or multistage hydrothermal event for formation of this very large deposit.

IMPLICATIONS OF Os_i AND He ISOTOPES FOR METAL AND FLUID SOURCES

Information about metal sources in ore systems is carried by the Os_i ratios of sulfide minerals (e.g., Arne et al., 2001; Levresse et al., 2004; Morelli et al., 2005), and He isotopes in fluid inclusions hosted by ore minerals are a powerful tracing tool for detection of mantle-derived volatiles (Turner and Stuart, 1992; Stuart et al., 1995; Levresse et al., 2004). Gold in orogenic environments, including Phanerozoic deposits such as Muruntau (Kostitsyn, 1994; Wilde et al., 2001), is commonly thought to originate from crustal rocks via leaching and transport by circulating metamorphic fluids. The most precise determination of Os_i, identical (within uncertainty) to Os_f of each individual regression, results from the conventional regression of samples 18 and 9824 (Fig. 2B; Os_i = 0.37 ± 0.27) and allows contributions from potential Os sources to be assessed despite the large uncertainty that results from the very radiogenic Os isotopic compositions of arsenopyrite. Figure 4 compares Os_i values of Muruntau arsenopyrite to reservoirs that may have contributed Os to the hydrothermal system, including upper (Peucker-Ehrenbrink and Jahn, 2001; Sun et al., 2003; Hattori et al., 2003) and lower (Saal et al., 1998) continental crust, and primitive upper mantle (Meisel et al., 2001). These data indicate that regional metamorphism of crustal rocks is unlikely to have been the only source of Os in the Muruntau hydrothermal fluids, as Os_i values of upper continental crust are too radiogenic (Fig. 4). Limited overlap between Muruntau Os_i and lower continental crust suggests that a solely lower crustal origin for Os is also unlikely. A plausible

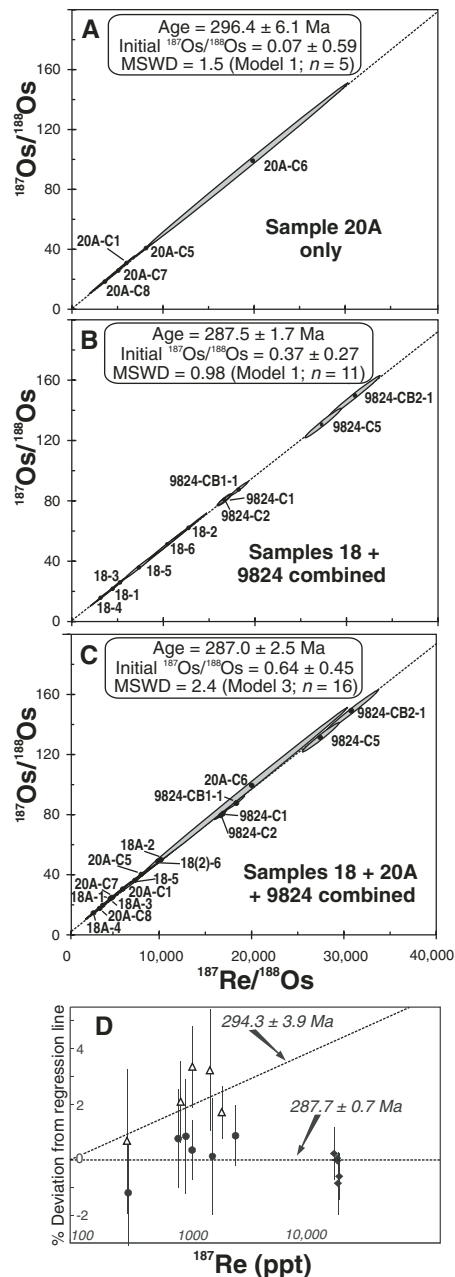


Figure 2. A: Re-Os model 1 isochron from regression of analyses of Muruntau arsenopyrite sample 20A. B: Re-Os model 1 isochron from regression of analyses of arsenopyrite samples 18 and 9824 from Muruntau. Age quoted is from $^{187}\text{Re}/^{188}\text{Os}$ versus $^{187}\text{Os}/^{188}\text{Os}$ regression. C: Re-Os model 3 “errorchron” from regression of analyses of all arsenopyrite samples from Muruntau (9824 + 18 + 20A). D: Re-Os ages derived from regression of analyses of independent arsenopyrite samples in ^{187}Re versus $^{187}\text{Os}^*$ (radiogenic Os) space. This plot reveals deviations outside 2σ uncertainties of analyses of sample 20A (open triangles), which individually yield a model 1 age of 294.3 ± 3.9 Ma, from regression line defined by samples 18 (filled circles) and 9824 (filled diamonds). Regression of all data in this study was performed using IsoPlot version 3 (Ludwig, 2003). MSWD—mean square of weighted deviates.

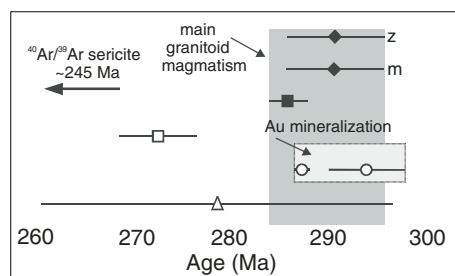


Figure 3. Comparison of Re-Os arsenopyrite isochron ages (with associated 2σ uncertainties; open circles) to existing ages for Muruntau gold mineralization. Open symbols represent inferred mineralization ages; filled symbols represent minimum crystallization ages for local felsic intrusives. Square—Rb-Sr whole rock (Kostitsyn, 1994); triangle—Sm-Nd scheelite (Kempe et al., 2001); diamonds—U-Pb zircon (z) and monazite (m) (Kempe et al., 2004).

explanation for the Muruntau arsenopyrite Os_i value of 0.37 ± 0.27 is one that requires contribution of Os from both older crust and contemporaneous mantle-derived magmas. Alternately, the Os_i could be explained solely from crust formed shortly before ca. 290 Ma—assuming typical crustal and mantle Re-Os values, only ~40 m.y. of prehistory is required to evolve from mantle Os values to the Muruntau arsenopyrite value at 0.37 (Fig. 4).

Assuming that He followed a similar pre-mineralization pathway to Os, the presence of an unradiogenic Os component is supported by He isotopes from fluid inclusions in arsenopyrite. The $^3He/^4He$ ratios of fluid inclusions determined here show a narrow range ($0.23\text{--}0.33R_a$; supplementary data table in the GSA Data Repository¹), consistent with other He isotope data from Muruntau arsenopyrite, quartz, and scheelite (Graupner et al., 2006). These values are an order of magnitude higher than continental crust-derived He ($^3He/^4He < 0.01R_a$; Stuart et al., 1995), and they require a contribution of mantle-derived (primordial) 3He . Assuming a depleted-mantle R_a value (~8 for modern day; Stuart et al., 1995), at least a few percent of He in Muruntau ore fluids must be from a mantle source. A similar conclusion was reached by Graupner et al. (2006).

The significance of the Os_i and He isotopic data in relation to the origin of gold at Muruntau remains uncertain, due to limited understanding of the exact source and transport relationships between Os, He, and Au in hydrothermal systems. The Os and He isotope data do

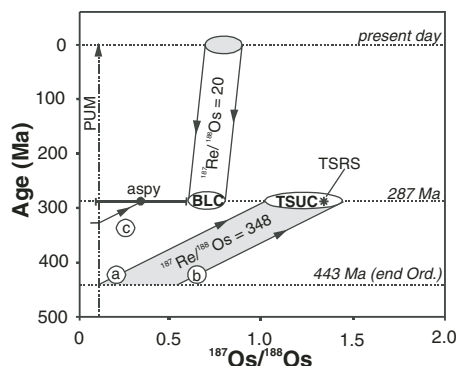


Figure 4. Comparison of Os_i from arsenopyrite (aspy) to terrestrial Os reservoirs that may have contributed Os to Muruntau hydrothermal system. PUM—primitive upper mantle (Meisel et al., 2001). TSUC—Tien Shan upper crust, which is a minimum estimate of $^{187}Os/^{188}Os$ for Cambrian-Ordovician (Ord.) host sediments at Muruntau, made by using mantle (A) and Ordovician seawater (Widom et al., 2004) (B) initial $^{187}Os/^{188}Os$ ratios and a $^{187}Re/^{188}Os$ ratio of 348 (Peucker-Ehrenbrink and Jahn, 2001; Sun et al., 2003). TSRS—Os isotopic composition of riverine sediment from Tien Shan mountains calculated at 287 Ma (Hattori et al., 2003), which agrees with TSUC estimates. Also shown is estimate of Os isotopic composition of bulk lower continental crust (BLC; Saal et al., 1998) at 287 Ma. Nominal Os_i from arsenopyrite at Muruntau of 0.37 could have a crustal prehistory of only ~40 m.y., based on typical crustal and mantle Re-Os characteristics (C).

require a major crustal component in the ore system, but they also reveal input from a juvenile source, presumably the mantle, suggesting that mantle-to-crust metal and volatile transfer was important in the formation of the Muruntau deposit. This signature, which is heavily masked via interaction with crust, has gone undetected by other isotope systems, including Nd (Kempe et al., 2001), Sr (Kostitsyn, 1994), and Pb (Chiaradia et al., 2006). As such, a juvenile source for gold at Muruntau (Graupner et al., 2006) must be considered. If this is a valid interpretation, emplacement of granitoid rocks proximal to Muruntau, now known to overlap with the timing of gold mineralization, may be evidence of this influx of juvenile material to the crust. Some intrusions have relatively unradiogenic initial Sr isotope ratios (e.g., Sardarin granodiorite, $^{87}Sr/^{86}Sr = 0.7079$; Kostitsyn, 1996) and are interpreted to have formed during interaction of mantle-derived magmas with lower crustal rocks on a regional scale (Kostitsyn, 1996). The juvenile Os and He components in the ore fluids might then have originated by exsolution of fluids from the granitoid magmas, or, alternatively, they may have originated directly from mafic magmas emplaced into the crust. The latter scenario supports the interpretation of Graupner et al. (2006), who invoked a relation-

ship between the generation of gold-bearing fluids and the emplacement of local lamprophyre dikes on the basis of noble gas, C isotope, and halogen data at Muruntau. To further evaluate this hypothesis, robust isotopic ages are required for lamprophyre emplacement, since current dates (ca. 273 Ma) are based on Rb-Sr analyses of hydrothermally altered whole-rock samples (Kostitsyn, 1996) that may reflect disturbance of true crystallization ages by later events.

COMPARISON WITH OTHER OROGENIC GOLD SYSTEMS

The presence of a juvenile Os component is either not present or not resolvable in some other orogenic gold deposits of Phanerozoic age. An important example is the ca. 438 Ma Bendigo deposit of central Victoria, Australia, for which Arne et al. (2001) presented a well-constrained Os_i value of 1.04 ± 0.16 from a Re-Os pyrite-arsenopyrite isochron. This value contrasts with that measured for Muruntau and indicates a predominately crustal derivation of Os for the Bendigo ore system. Morelli et al. (2005) determined a radiogenic Os_i value of 0.83 ± 0.16 for arsenopyrite from the ca. 408 Ma Ovens gold occurrence in the southwestern Meguma terrane, Canada, together with an Os_i value of 0.38 ± 0.16 , much like that for Muruntau, for arsenopyrite from the younger (ca. 380 Ma) Dufferin deposit in the eastern Meguma terrane. Given the paucity of Re-Os data from arsenopyrite in orogenic gold deposits, the significance of these variations in relation to potential gold sources is currently unclear. It is possible that the rocks interacting with the mineralizing fluid during transport in an orogenic gold system may control the resultant Os_i value of the sulfides. Alternatively, the hypothesis that primitive oceanic crust is the ultimate gold source to these deposits (e.g., Bierlein et al., 2006) may explain variable Os_i as variations in the age and/or Re-Os concentrations of this crust could potentially generate such variations.

High-precision Re-Os arsenopyrite geochronology, coupled with Os and He isotopic tracing, provides new constraints for the origin of orogenic gold deposits, such as Muruntau, where the timing of mineralization was ca. 290 Ma, not ca. 275–245 Ma as determined by other methods. The Os_i and He isotope data show that formation of the Muruntau deposit included a component of mantle-derived and/or juvenile metals and fluids. These data are incompatible with models that invoke a purely crustal source for fluids and metals at Muruntau (e.g., Wilde et al., 2001), or those that require mineralization younger than ca. 290 Ma (Kostitsyn, 1996; Wilde et al., 2001). The new Re-Os dates for ore formation overlap the timing of proximal granitoid magmatism, so “intrusion-related” models for ore formation are permitted by these

¹GSA Data Repository item 2007197, Re-Os-He isotopic data for Muruntau arsenopyrite, is available online at www.geosociety.org/pubs/ft2007.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

data (e.g., Wall et al., 2004), although a mantle component is required in magma genesis. Alternatively, if Os, He, and Au in the hydrothermal system have different origins, more complex models involving the coincidence of mantle and crustal magmatism, juvenile and crustal fluids, and favorable structural conditions at ca. 290 Ma, are possible. The use of coupled Re-Os-He isotope analyses in arsenopyrite should provide valuable constraints for the origin of orogenic gold deposits worldwide.

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