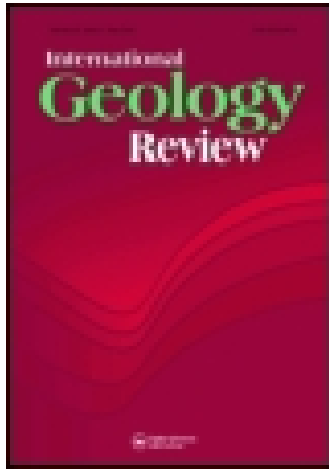


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Au-Se Paragenesis in Cambrian Stratabound Gold Deposits, Western Qinling Mountains, China

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Abstract

Cambrian stratabound gold deposits in the western Qinling Mountains are hosted in a siliceous formation composed of black chert and carbonaceous slate. Studies have shown that Se is sufficiently high in abundance either in wall rocks or in gold ores as to reach economic grade. Locally, some independent Se ore bodies (Se >500 ppm) can be delineated. In gold ores, Se is present mainly as separate minerals, or as isomorphous solutions in sulfides. In addition, Se is positively correlated with Au. What is more important is that in the gold ores, native gold is commonly intimately intergrown with selenium minerals. On the basis of this unique phenomenon, the authors hypothesize that gold and selenium may be transported by Au-S-Se or Au-Se complexes. The co-enrichment of Au and Se is attributed mainly to the boiling of ore fluids and their mixing with shallow-seated oxygen-bearing water.

Introduction

THE CAMBRIAN STRATABOUND gold deposits in the southern subzone of the western Qinling Mountains include the Laerma (Edu) and the Qiongmo gold deposits and the Yaxiang gold ore occurrence (Fig. 1). They constitute a new type of gold deposit in China that possesses unique geological characteristics. In the mid-1980s, the Northwest Sichuan Geological Party of Sichuan Provincial Bureau of Geology and Mineral Resources and the No.3 Geological Party of the Gansu Provincial Bureau of Geology and Mineral Resources began surveying and exploring those gold deposits. In their study of the Cambrian gold deposits, the authors found not only a variety of independent selenide minerals and Se-bearing minerals (Liu and Zheng, 1992; Liu Jiajun et al., 1992, 1993, 1995, 1996, 1998a-1998d, 1999, 2000), but also observed strong enrichment of Se in the ores (Liu and Zheng, 1993a; Liu Jiajun et al., 1997a). Moreover, locally some separate Se ore bodies can even be delineated (Se >500 ppm). The strong enrichment of Se in the gold deposits poses a challenge to the traditional conception that dis-

persed elements cannot be involved in mineralogenesis, and it is also a clear indication that a new type of Au-Se paragenetic mineralization exists (Liu and Zheng, 1993b). This provides the basis for a combined study of enrichment and metallogenesis of dispersed elements (the conditions required for Se enrichment and gold metallogenesis) so as to explore the mechanism of Au-Se paragenesis. Meanwhile, its discovery is of great significance to the comprehensive utilization of the abundant mineral resources in this region and in promoting the development and utilization of, and research on, selenium in the western Qinling Mountains and analogous areas of China.

The Problem

Selenium is a dispersed element and is not easily enriched to such an extent as to be of commercial importance. The occurrence of selenides has been reported from four major types of hydrothermal deposits: "telethermal" selenide vein-type deposits, unconformity-related deposits, sandstone-hosted uranium deposits, and epithermal Au-Ag deposits in

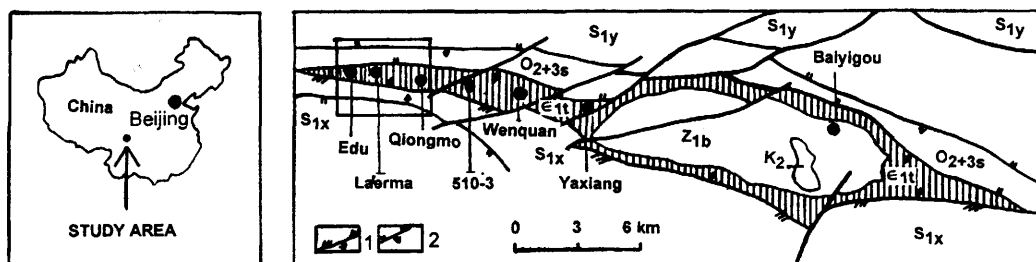


FIG. 1. Regional geological sketch map of the Cambrian stratabound gold deposits, western Qinling Mountains (based on the Northwest Geological Party of Sichuan Provincial Bureau of Geology and Mineral Resources [Northwest Geological, 1990], with slight modifications). Legend: K₂ = Upper Cretaceous; S_{1y} = Lower Silurian Yangchanggou Formation; S_{1x} = Lower Silurian Xiadi Formation; O_{2-3s} = Middle-Upper Ordovician Sulimutang Formation; ε_{1t} = Cambrian Taiyangding Formation; Z_{1by} = Sinian Baiyigou Formation; 1 = reverse fault; 2 = thrust fault.

subaerial volcanic environments (Simon et al., 1997). As reported in the literature worldwide, hydrothermal gold deposits containing selenides are mostly restricted to epithermal Au-Ag deposits that are genetically related to continental volcanism, for instance, the Koryu (Shimizu et al., 1998), Kushikino (Mukaiyama and Izawa, 1964; Shikazono et al., 1990), Hishkari (Izawa et al., 1990; Shikazono et al., 1990), Chitose, and Yatani mines (Shikazono et al., 1990) of Japan; the Sleeper (Saunders et al., 1988; Saunders, 1993) and Jarbridge mines (Hewitt, 1968) of Nevada; the Silver City mine (Petruk et al., 1974; Ramdohr, 1980) of Idaho, and the Republic mine (Full and Grantham, 1968) of Washington, all in the United States; the Torquay mine (Paar et al., 1998) of England; the Great Barrier Island (Ramsay and Kobe, 1974) and Camoola mines (Main et al., 1972) of New Zealand; the Salida (Kieft and Oen, 1973) and Redzhang Lebong (Sindeeva, 1964) mines of Indonesia; the Guanajuato mine (Petruk and Owens, 1974) of Mexico; the Providencia mine (Paniuga, 1995) of Spain; as well as many Au-Ag deposits in the Circum-Pacific and Mediterranean metallogenic belts, such as the Prasolovskoye, Kunashir Island, and Kuril Island deposits (Nekrasov and Lunin, 1987; So et al., 1995), all of which can be taken as the typical examples. Nevertheless, those ore deposits bear no Se-rich mineralized bodies, although the concentrations of Se are somewhat elevated.

In their studies of the Cambrian stratabound gold deposits in western Qinling, the authors found that large numbers of separate Se minerals occur in the gold deposits (Liu and Zheng, 1992; Liu Jiajun et al., 1992, 1993, 1995, 1996, 1998a, 2000); moreover, some Se-rich mineralized bodies (Liu and Zheng, 1993a; Liu et al., 1997a) also are present. In

the Se-bearing gold ores, the contents of Se are generally within the range of 30 to ~200 ppm, with a maximum of 7700 ppm (Liu et al., 1997a). Compositional ranges in the various investigated deposits are listed in Table 1.

Generally speaking, selenide minerals are chiefly concentrated in Ag-rich and telluride-rich epithermal Au-Ag deposits, mostly associated with continental volcanism. However, the Cambrian stratabound gold deposits in the western Qinling Mountains are unrelated to continental volcanism (Liu Jiajun et al., 1997b, 1998b), and, on the other hand, Ag anomalies and occurrences of tellurium minerals are lacking (Zheng et al., 1994). Such strong enrichment of Se in the gold deposits is unusual. What would cause such a strong enrichment? What role did selenium play in the process of gold mineralization? Why are Au and Se paragenetically associated? These questions cannot be explained in terms of traditional hypotheses.

Geology of the Gold Deposits

The Cambrian stratabound gold deposits are located in the terminal part of western Baiyi anticline in the southern subzone of the western Qinling Mountains (Fig. 1). The deposits are hosted in the Cambrian Taiyangding Group siliceous formation, consisting of black chert and slate, and are obviously controlled by stratigraphic, lithologic, and structural characteristics. The stratabound nature of the gold deposits is well accepted (Zheng et al., 1994).

More than 80% of the gold ore bodies occurring in the siliceous formation are hosted in chert, and they are generally distributed in broken zones along

TABLE 1. Contents of Gold and Selenium in Gold Deposits¹

Sampling locality	No. of samples	Contents (ppm)				Remarks
		Au		Se		
		Range	Average	Range	Average	
Yaxiang prospecting line 7	11	0.005–0.50	0.125	0.52–36.30	10.34	Weakly mineralized area
Yaxiang prospecting line 8	14	0.007–0.17	0.010	0.00–6.40	3.27	Weakly mineralized area
Wenquan prospecting line 101	100	0.003–0.05	0.008	0.20–37.90	4.50	Unmineralized area
Qiongmo prospecting line 15	10	0.002–0.02	0.009	1.40–19.50	7.37	Weakly mineralized area
Qiongmo prospecting line 11	8	0.017–0.24	0.054	1.10–11.50	6.70	Weakly mineralized area
Qiongmo prospecting line 7	79	0.002–7.88	0.634	1.11–65.47	16.32	Strongly mineralized area
Qiongmo prospecting line 0	5	0.010–0.21	0.104	0.00–14.90	8.12	Strongly mineralized area
Laerma prospecting line 137	21	0.002–1.25	0.218	1.75–47.77	10.73	Weakly mineralized area
Laerma prospecting line 109	20	0.150–68.02	6.252	1.20–1202.0	185.66	Strongly mineralized area
Laerma prospecting line 106	84	0.016–3.45	0.547	2.50–48.20	17.92	Strongly mineralized area
Laerma PD109 tunnel	13	0.160–2.88	0.935	0.53–1100.0	145.69	Strongly mineralized area
Laerma drill hole ZK1071	4	0.220–6.40	2.460	13.10–22.50	15.23	Strongly mineralized area
Laerma drill hole ZK1072	8	0.350–4.10	2.121	2.10–807.00	104.70	Strongly mineralized area
Laerma drill hole ZK109	4	0.280–1.63	0.620	9.80–165.40	61.05	Strongly mineralized area
Qiongmo OPI tunnel	12	0.067–26.43	5.101	6.80–315.98	165.23	Strongly mineralized area
Qiongmo PD8 tunnel	3	0.017–0.08	0.048	15.29–49.81	6.25	Weakly mineralized area
Qiongmo PD48 tunnel	3	0.044–1.02	0.396	41.10–7700	2612.03	Strongly mineralized area
Qiongmo PD11 tunnel	58	0.003–6.09	0.335	0.79–222.00	43.45	Strongly mineralized area
Qiongmo drill hole ZK3-1	2	0.047–0.05	0.048	2.10–2.60	2.35	Weakly mineralized area
Qiongmo drill hole ZK7-1	47	0.003–16.70	1.510	1.40–83.50	22.20	Strongly mineralized area
Qiongmo drill hole ZK7-2	39	0.001–1.39	0.242	3.40–232.00	31.10	Strongly mineralized area
Qiongmo drill hole ZK7-3	56	0.010–3.31	0.962	2.90–91.60	13.85	Strongly mineralized area
Other areas in Laerma	3	0.140–3.03	1.095	28.70–91.60	68.83	Strongly mineralized area
Other areas in Qiongmo	6	0.042–0.53	0.181	5.30–100.00	29.50	Weakly mineralized area
Average/total	610	0.001–68.02	0.745	0.00–7700	42.08	

¹Some of the data were provided by Liu Xinhua and Zhou De'an (1991).

the bedding. The ore bodies are stratoid and lenticular in form, and their mode of occurrence is consistent with that of the country rocks. The ore minerals and element associations are very complicated. Up to now, more than 80 mineral species have been identified, including sulfides of Cu, Pb, Zn, As, Mo, Ni and Sb, U-bearing minerals, and a large number of oxide minerals (Liu, 1996). The common minerals include native gold, tiemannite, clausenthalite, pyrite, marcasite, stibnite, quartz, bar-

ite, dickite, etc., but native selenium, telluride, and selenides of Ag and Cu have not been found in the gold ores. The authors have identified a variety of selenides—for example tiemannite, clausenthalite, antimonoselite, sederholmite, kullerudite, and Ni-As-S-Se mineral phases. The diversity of mineral species in the ores and the extensive occurrence of selenium minerals are the outstanding features of this type of gold deposit (Liu and Zheng, 1993b).

According to the differences of ore host rocks, three types of ore can be distinguished: (1) chert-type gold ores; (2) slate-type gold ores; and (3) dacite-porphyry-type gold ores. The first type includes stratiform chert ores and silicified chert ores, which is the principal type in the gold deposits, accounting for 80% of the total amount of ores. In these ores the content of Au varies greatly, with the maximum up to 66.0 ppm, but mostly within the range of 1.0 to 7.0 ppm. The content of Au in the slate-type ores generally varies between 2.0 ppm and 5.0 ppm. The dacite-porphyry-type ores are common in the Qiongmo mining district. The content of gold in this type of ore is extremely variable, with the maximum up to 41.57 ppm. In addition to gold, separate orebodies of Cu, U, Mo, Sb, V, and Zn are delineated locally in the ore deposits. What is particularly important is that in their study of the ore deposits, the authors found a strong enrichment of Se, In, and PGE (the platinum-group elements) (Liu and Zheng, 1993a; Liu Jiajun et al., 1997a, 1998c; Zheng et al., 1991). Moreover, selenium ore bodies can be delineated locally. As a result, the commercial value of the gold deposits is expected to be greatly enhanced.

Wall-rock alterations in the ore deposits include silicification, stibnitization, pyritization, baritization, dickitization, and decoloration, with silicification predominant. Most gold ore bodies are hosted in the most strongly silicified rocks. Decoloration is commonly accompanied by silicification. Its outstanding feature is that the color of the rocks tends to change from greyish black and dark grey to greyish white, as organic matter is eliminated. The other types of wall-rock alteration are of limited distribution. Generally speaking, wherever the wall-rock alteration is strong, the ores are relatively complicated chemically, and the degree of mineralization is relatively high; wherever wall-rock alteration is weak, the mineralization is indistinct.

As viewed from the crosscutting relationship of veins, ore fabrics, and mineral paragenetic associations, formation of the Laerma and Qiongmo gold deposits evidently involved two metallogenic stages: (1) an early metallogenic stage, represented by the association of pyrite–marcasite–chalcopyrite–bornite–native gold–quartz; and (2) the main metallogenic stage, represented by the association of stibnite–selenide–native gold–quartz–barite. Gold mineralization occurred mainly between late Meso-

zoic and early Cenozoic time (49–137 Ma) (Liu Jiajun et al., 1998d).

Mechanism of Au-Se Paragenesis

Theoretical basis

It is well known that selenium is located in the sixth main group of the fourth period in the Periodical Table of Elements, belonging to the S-Se-Te series. Because of the similarities of selenium and sulfur in crystal chemistry and geochemical properties (e.g., ionic radius, lattice energy coefficient, ionic potential, etc.), selenium is readily accommodated in crystal structure sites of sulfides, and is present isomorphously. In addition to other conditions, temperature is one of the principal factors controlling the behavior of selenium in hydrothermal solutions. Experimental evidence (Liu Yingjun et al., 1987) suggests that under high-temperature conditions, the range in which S and Se are present isomorphously increases. Sulfide minerals that crystallized at moderate to high temperatures are relatively enriched in selenium, whereas selenium is scarce and dispersed in the sulfides crystallized at low temperatures. Under this circumstance, if there is more selenium than sulfur, formation of separate minerals of selenium will be favored. In hydrothermal processes, selenium could be widespread in the dispersed form or as independent minerals in geological bodies during the hydrothermal stage. That is to say, almost all sulfide ore deposits of hydrothermal origin and some gold deposits would contain selenium. As to the stratabound gold deposits, hydrothermal precipitation is one of the necessary prerequisites of gold mineralization. Although gold in the stratabound gold deposits is not abundant in sulfide crystal structures, it is commonly present in some sulfides as fine, disseminated particles or inclusions.

On the other hand, Au and Se are both siderophile elements and show insignificant differences in some properties, for example electronegativity (Au 2.3 eV, Se 2.4 eV), and covalent radius (Au 0.134 nm, Se 0.1076 nm). Au and Se are both quite mobile in hydrothermal solutions, and hence are easily transported. Both elements can combine to form selenides of gold, such as fischerite (Ag_3AuSe_2), kurilite ($(\text{Au,Ag})_2(\text{Te,Se,S})_2$), penzhinite ($(\text{Ag,Cu})_4\text{Au}(\text{S,Se})_4$) and petrovskite ($(\text{AuAg}(\text{S,Se}))$).

Transport forms of Au and Se

As described previously, the Cambrian stratabound gold deposits are very high in Se content, in addition to several kinds of selenide minerals, indicating that a special metallogenic environment prevailed during formation of the deposits. In fact, they were formed under moderate- to low-temperature conditions characterized by low sulfur fugacity (depletion in reduced sulfur), and SO_4^{2-} occurring as the dominant form of sulfur. The occurrence of a variety of selenide minerals in the deposits, especially the frequent intergrowth of native gold and selenide minerals (Fig. 2), and the positive correlation between Au and Se in the gold ores provide strong evidence suggesting that during hydrothermal mobilization and transport, selenium and gold share much in common with respect to geochemical behavior.

As far as the types of anion groups complexing with gold are concerned, it is known that there are 22 kinds of complexes of Au with oxygen, sulfur, and tellurium in the oxygen-sulfur-selenium-tellurium group system (Liu, 1996), but the complexing of Au with Se has not been reported previously. As for the coordination ligands of complexes, Se and S are extremely similar in geochemical behavior (Cotton and Wilkinson, 1988). In addition, Se and S are identical in electron configuration and similar in chemical properties. The molecular configuration of H_2Se is similar to that of H_2S , both being designated by the angle type. The bond angle of H-Se-H is about 90° , slightly smaller than that of H-S-H. This is probably because the decrease of electronegativity of the elements leads to the polarization of bond-combining orbits, thus allowing the two elements to approach one other more closely. Like H_2S , the decomposition of H_2Se leads to the release of hydrogen ions, and Se complexes with heavy metals to form selenides. But the acidity and reductibility of H_2Se are both stronger than those of H_2S . Studies of the geochemical behavior of Se in hydrothermal solutions (Dyachkova and Khodakovskiy, 1968; Yamamoto, 1976) have indicated that, at temperatures higher than 200°C , H_2S and H_2Se are the predominant aqueous Se and S species in an S- and Se-bearing aqueous solution, with the activity ratio of $m_{\text{H}_2\text{Se}}/m_{\text{H}_2\text{S}}$ being close to that of $m_{\text{S}^{2-}}/m_{\text{Se}^{2-}}$; at temperatures lower than 150°C , HSe^- is the dominant aqueous Se species in a Se-bearing solution, with $m_{\text{H}_2\text{Se}}/m_{\text{H}_2\text{S}}$ not being approximated by $m_{\text{S}^{2-}}/m_{\text{Se}^{2-}}$.

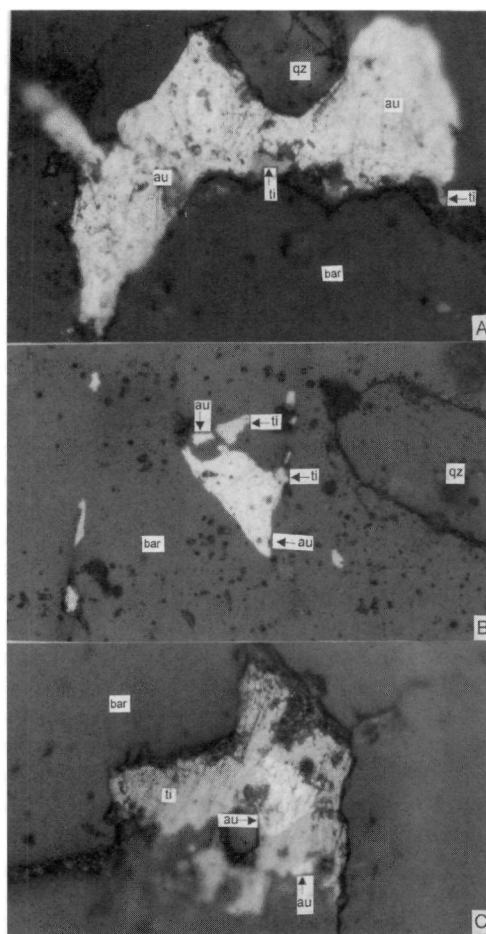


FIG. 2. A, B, and C. Coexistence of native gold with tiemannite. Polished, (-), $\times 400$, $\times 200$, and $\times 400$, respectively. Abbreviations: au = native gold; ti = tiemannite; qz = quartz; bar = barite.

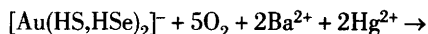
Experimental results have also demonstrated that pH and oxygen fugacity ($f\text{O}_2$) have a great influence on the activity ratio of $m_{\text{S}^{2-}}/m_{\text{Se}^{2-}}$ (Dyachkova et al., 1968; Yamamoto, 1976). Therefore, in strongly reducing Au-bearing solutions, gold is transported mainly in the form of $\text{H}[\text{Au}(\text{HS})_2]$ (Liu, 1996). Under this circumstance, H_2S , HS^- and S^{2-} in the complex $\text{H}[\text{Au}(\text{HS})_2]$ may be replaced by H_2Se , HSe^- , and Se^{2-} , respectively to form the Au-S-Se complex $\text{H}[\text{Au}(\text{HS}, \text{HSe})_2]^{0}$ or $[\text{Au}(\text{HS}, \text{HSe})_2]^{-}$, or even finally to form the Au-Se complex $[\text{HAu}(\text{HSe})_2]^{0}$ or $[\text{Au}(\text{HSe})_2]^{-}$. Once the stability field required for this kind of complexes is destroyed, such Au-S-Se complexes will dissociate, and Au will coexist or intergrow with selenide min-

erals, or may even form selenides of Au. The intimate coexistence of native gold with a number of selenide minerals in the gold deposits indicates that gold is transported in the form of an Au-S-Se complex.

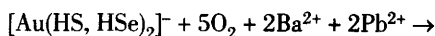
Coprecipitation and enrichment of Au and Se

Reflecting a significant difference in the oxidation potentials of S and Se, separation of these elements is believed to occur normally in an oxidizing environment. Sulfur is easily oxidized, with $E^0 = -0.45$ eV, as opposed to $E^0 = 0.74$ eV for Se. So, under reducing conditions, Se is present isomorphously in the sulfide crystal structure. But in a relatively strongly oxidizing environment, Se is present mainly as separate selenide minerals. If the decomposition and precipitation of Au-S-Se complexes is effected by way of the extensive production of sulfides, thus diminishing the concentrations of H_2S in ore fluids, Se could, in a random manner, enter the sulfides to replace sulfur to form Se minerals. The extensive occurrence of independent selenide minerals in the gold deposits, and the fact that Se finds its way into the structure of sulfides via isomorphous substitution demonstrates that gold is transported in the form of Se complexes, and this may be a reasonable explanation for the strong co-enrichment of Au and Se.

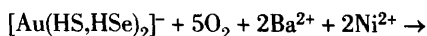
In the studied Cambrian stratabound gold deposits, the co-enrichment of Au and Se is attributed mainly to boiling of ore fluids and their mixing with shallow-seated oxygen-bearing water (Liu, 1996). The reaction formulae can be written as follows:



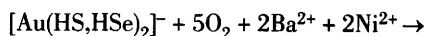
Au (native gold) + 2BaSO₄ (barite) + 2HgSe (tiemannite) + 2H₂O



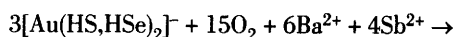
Au (native gold) + 2BaSO₄ (barite) + 2PbSe (clausthalite) + 2H₂O



Au (native gold) + 2BaSO₄ (barite) + 2NiSe (sederholmite) + 2H₂O



Au (native gold) + 2BaSO₄ (barite) + NiSe₂ (kullerudite) + 2H₂O



3Au (native gold) + 6BaSO₄ (barite) + 2Sb₂Se₃ (antimonelite) + 6H₂O.

So, in the ore fluids, Au and Se can be transported together and co-precipitated, leading to the paragenetic enrichment of Au and Se.

Geological Implications

The crustal abundance of Se is a thousand times lower than that of S, but larger than that of Te (Liu Yingjun et al., 1987; Simon et al., 1997). Because Se and S are very similar in crystal chemistry and in some geochemical properties—for instance, ionic radius (S²⁻ 0.184 nm, Se²⁻ 0.191 nm), lattice energy coefficient (S²⁻ 1.15, Se²⁻ 1.10), and ionic potential (S²⁻ -1.09 eV, Se²⁻ -1.05 eV) (Liu Yingjun et al., 1987)—and moreover, Se, unlike Te, belongs to the strongly chalcophile elements, it is easy for Se to replace S in sulfides, but difficult to form selenides. This may explain why Se independent minerals are scarcer than sulfides and tellurides in nature.

As reported up to now in the literature, gold deposits that contain selenides are, for the most part, closely associated with continental volcanism, and represent epithermal Au-Ag deposits that contain large amounts of tellurides and Ag-bearing selenides. However, the Cambrian stratabound ore deposits are unrelated to continental volcanism; they also lack silver anomalies and contain no tellurides. Instead, large numbers of separate selenide minerals and Se-bearing minerals, as well as Se mineralized lodes, are present. These phenomena indicate that the gold deposits were formed in a special ore-forming environment, as has been clearly demonstrated by research on the physical and chemical conditions of formation of the gold deposits (Liu Jiajun et al., 1997a), i.e., the involvement of Se- and Au-rich fluids in gold mineralization under moderate to low temperatures (142–269°C) and low-pressure (9–30 MPa) conditions.

Gold in the ore-forming fluids was transported in the form of S-Se-Au complexes. At early stages of the metallogenesis, both fO_2 and fSe_2 were relatively low, and fS_2 was relatively high, with $fSe_2/fS_2 < 1$ favoring the precipitation of sulfides. Under this circumstance, Se in the ore-forming fluids could not be separated completely from S through its precipitation, and therefore, the minerals formed were mostly Se-bearing. During the main stage of metallogenesis, with the precipitation of sulfides, fSe_2/fS_2 increased correspondingly, with fO_2 being relatively high. Higher fO_2 hampered the entry of Se into the

sulfides, thus favoring the formation of selenides. Particularly, the oxidation of S into sulfate radical ions enhanced the effective separation of Se from S, leading to the formation of Se minerals in large amounts.

The control of the oxidizing environment on the separation of S and Se, and on the transport of Se and deposition of selenide minerals from hydrothermal fluids has been discussed in detail by Tischendorf (1959, 1969) in terms of Eh, and by Simon and Essene (1996) and Simon et al. (1997) in terms of $fO_{2(g)}$ and $fS_{2(g)}$. These variables are critical to an understanding of the aqueous geochemistry of Se and in order to discuss the mechanism of Au-Se paragenesis in the Cambrian gold deposits. So the following two prerequisites must be satisfied for the paragenetic enrichment of Au and Se: (1) an abundant source of Au and Se; and (2) a relatively oxidizing environment.

Does the described Au-Se paragenesis mean that Se could be used as an indicator element of the Cambrian stratabound gold deposits? Up to now, a variety of elements have been proposed as indicator elements for hydrothermal gold deposits, such as Sb, As, Cd, Tl, Mn, Hg, Ba, B, and the halogens (Brookins, 1986; Warren, 1986). In our studies of the gold deposits, we have also proposed that Se can be used as one of the indicator elements of stratabound gold deposits (Liu and Zheng, 1994). Nevertheless, more research is required to fully understand the role of Se as a tracer, the mechanism of Au-Se paragenesis, and the role of Se in gold mineralization.

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