

胶东玲珑金矿区碲化物形成条件及其对金富集过程的约束^{*}

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Abstract Jiaodong gold province has proved gold reserves of more than 5000t, which is the most important gold concentration area in China. Linglong gold district is located in the northern section of Zhao-Ping fault in the northwest of Jiaodong Peninsula, and it has the typical characteristics of a gold-bearing quartz vein deposit. Some of the ore sections in this mining area produce tellurium-bismuth enriched gold ore, therefore, it is an ideal object to study the relationship of the genesis of ore deposits and the extreme enrichment mechanism of gold with the enriched tellurium-bismuth. Through microscopic observation, scanning electron microscope analysis (SEM) and electron probe micro-analysis (EPMA), the mineral assemblages and formation mechanism of tellurium-bismuth enriched gold ore in Linglong gold district were studied in detail for the first time. The gold minerals in Linglong mining area are mainly silver-bearing native gold, and the gold fineness is high with an average of 894. Five kinds of tellurides co-occurring with native gold have been found, namely, tetradyomite, tellurobismuthite, hessite, volynskite and melonite, among which the tetradyomite is the most abundant. According to the study of mineral assemblages and analysis of physical and chemical conditions, the range of tellurium fugacity in the ore-forming fluid is determined to be $-12.6 < \log f_{\text{Te}_2} < -9.1$, while the sulfur fugacity range of $-13.6 < \log f_{\text{S}_2} < -9.6$. The enrichment of tellurium and bismuth indicates they are formed by a deep mantle source, and to be more specifically, the tellurium and bismuth may have been derived from the subduction of the paleo-Pacific plate. The rich concentration of large grains of native gold in Linglong gold district is closely related to tellurium and bismuth, and the precipitation of silver telluride improves the fineness of native gold.

Key words Tellurides; Native gold; Enrichment mechanism; Linglong gold district; Jiaodong gold province

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摘要 胶东地区探明金储量超过 5000t,是我国最重要的金矿集区。玲珑金矿区位于胶东西北部招-平断裂北段,具有典型含金石英脉矿床的特征。矿区部分矿段产出富碲铋化物的明金矿石,是研究富碲铋化物金矿床成因和金超级富集机制的理想对象。通过显微岩相学观察、扫描电镜及电子探针分析,首次对胶东玲珑金矿区富碲铋化物明金矿石的矿物组合和形成机制进行了较为详细的研究。玲珑金矿区含金矿石中的金矿物主要为含银自然金,成色整体较高,平均为 894。共发现了五种与自然金共生的碲化物,分别是辉碲铋矿、碲铋矿、碲银矿、碲铋银矿和碲镍矿,其中辉碲铋矿含量最多。通过矿物共生组合研究及物理化学条件分析,确定成矿流体的碲逸度范围为 $-12.6 < \log f_{\text{Te}_2} < -9.1$,硫逸度范围为 $-13.6 < \log f_{\text{S}_2} < -9.6$ 。碲、铋元素的富集指示了深成慢源的信息,碲、铋等物质可能来源于古太平洋板块俯冲。玲珑金矿区大颗粒自然金的超级富集与碲、铋等元素密切相关,且银碲化物的产出提高了自然金的成色。

关键词 碲化物;自然金;富集机制;玲珑金矿区;胶东金成矿省

中图法分类号 P618.51; P618.83

胶东地区目前是中国最重要的黄金产区,拥有超过 5000t 的黄金资源 (Deng *et al.*, 2020b)。胶东金矿床赋存于前寒武纪变质基底中,主要分为浸染状细脉-网脉型(“焦家型”)和石英-硫化物脉型(“玲珑型”)矿床 (Deng *et al.*, 2020a)。焦家型金矿床产于区域性断裂的蚀变带中,玲珑型金矿床分布在区域性断裂的次级断层中 (Yang *et al.*, 2016b)。经过几十年的研究,关于胶东地区金的来源、超级富集机制以及矿床成因模式仍然存在激烈的争论。胶东金矿因其矿床的线性构造分布、矿化类型、围岩蚀变、流体成分和稳定同位素组成等 (Goldfarb and Santosh, 2014) 与造山型金矿特征 (Goldfarb *et al.*, 2019; Qiu *et al.*, 2020a) 相似,一般被归类为造山型金矿。由于胶东金矿床的构造环境和变质背景相对于传统造山型金矿成矿模式 (Groves *et al.*, 1998) 具有独特性,故很难将其纳入经典造山型金矿。胶东金矿应划分为一类独特的胶东型金矿 (Zhai and Santosh, 2013),或一种独特的造山型金矿 (Goldfarb and Santosh, 2014),即胶东型造山型金矿 (Deng *et al.*, 2020b)。玲珑金矿区位于胶东西北部,招远-平度断裂带北部,以石英-硫化物脉为特征的玲珑型金矿床而闻名于世,整个矿区预计超过 1000t 以上的黄金资源量 (Qiu *et al.*, 2020b)。玲珑金矿区部分矿段产出高品位的明金矿石,大颗粒自然金的超级富集机制是备受瞩目的科学问题。

在热液金矿床中,铋-硫族化合物(硫族主要为 S 和 Te)常与金紧密共生。铋、碲矿物在国内外金矿床中均有发现,例如罗马尼亚 Sacarimb (Cook and Ciobanu, 2004),美国 Golden Sunlight (Spry *et al.*, 1997),菲律宾 Acupan (Cooke and McPhail, 2001) 等,以及中国的河北东坪 (Cook *et al.*, 2009a) 等。许多研究表明铋-碲熔体在金的运移和沉淀机制中起着关键作用,可作为热液中金的关键吸附剂 (Cabri, 1965; Frost *et al.*, 2002; Törmänen and Koski, 2005; Ciobanu *et al.*, 2006; Plotinskaya *et al.*, 2006; Tomkins *et al.*, 2007; Tooth *et al.*, 2008, 2011; Cook *et al.*, 2009a, b; Voudouris *et al.*, 2013)。并且由于碲化物对硫逸度、碲逸度、pH 值、温度等条件的高度敏感性,使其成为限制金沉淀时物理化学条件的重要指标。因此,对碲化物的研究有助于我们了解金的成矿过程及成矿时的物理化学条件。

玲珑金矿区明金矿石中存在大量与自然金共生的碲化物。本文通过显微岩相学观察、扫描电镜及电子探针分析,对玲珑金矿区碲化物的矿物组合特征及化学成分等进行研究;在此基础上,确定了碲化物的种类及成分特征,探讨共生矿物的成矿条件,并对成矿物质来源以及自然金的富集机制等问题进行了论述。本文是关于玲珑金矿区自然金共生碲化物的首次详细报道,在查明碲化物种类及特征的同时,阐明了碲化物产出的地质意义。

1 区域地质背景

胶东半岛位于华北克拉通东南缘,西部以岩石圈尺度的郯庐断裂带为界 (Deng *et al.*, 2018) (图 1a),北临渤海,东部和南部被黄海所包围 (图 1b)。胶东半岛被认为是三叠纪扬子板块和华北板块碰撞拼合后形成的 (Liu *et al.*, 2006),被五莲-烟台断裂分成东南部的苏鲁地体和西北部的胶北地体 (魏瑜吉等, 2020),胶北地体可进一步分为胶北地块和胶莱盆地 (何登洋等, 2020; 图 1b)。胶东金矿床大多位于胶莱盆地北部,受 NNE-NE 向断裂控制 (Zhang *et al.*, 2020)。胶东地区广泛分布着前寒武纪基底变质岩,主要包括胶北地块的以 TTG (奥长花岗岩-英云闪长岩-花岗闪长岩) 片麻岩为主的新太古代胶东群,以变质沉积岩为主的元古宙景山群、粉子山群和蓬莱群 (Tang *et al.*, 2008) 以及苏鲁地体的三叠纪超高压变质岩 (图 1b)。

胶东地区中生代侵入岩十分发育,主要包括玲珑花岗岩、郭家岭花岗闪长岩、艾山花岗岩以及广泛分布的镁铁质岩脉。玲珑花岗岩以玲珑黑云母花岗岩和寨家河二长花岗岩为主,LA-ICP-MS 测得锆石 U-Pb 年龄主要在 166 ~ 149Ma (Zhao *et al.*, 2019),玲珑花岗岩来源于新太古代下地壳的部分熔融 (Yang *et al.*, 2018)。郭家岭花岗岩主要由斑状花岗闪长岩和石英二长岩组成,锆石 U-Pb 年龄显示其侵位时间为 133 ~ 126Ma (Yang *et al.*, 2014b),由下地壳前寒武纪变质基底部分熔融形成 (Hou *et al.*, 2007),并在上升过程中受到上地壳的混染 (Wang *et al.*, 2014)。艾山花岗岩主要出露在胶东半岛东部,由未变形的碱性花岗岩组成,锆石 U-Pb 年龄显示艾山花岗岩主要侵位于 120 ~ 108Ma (Tang *et al.*,

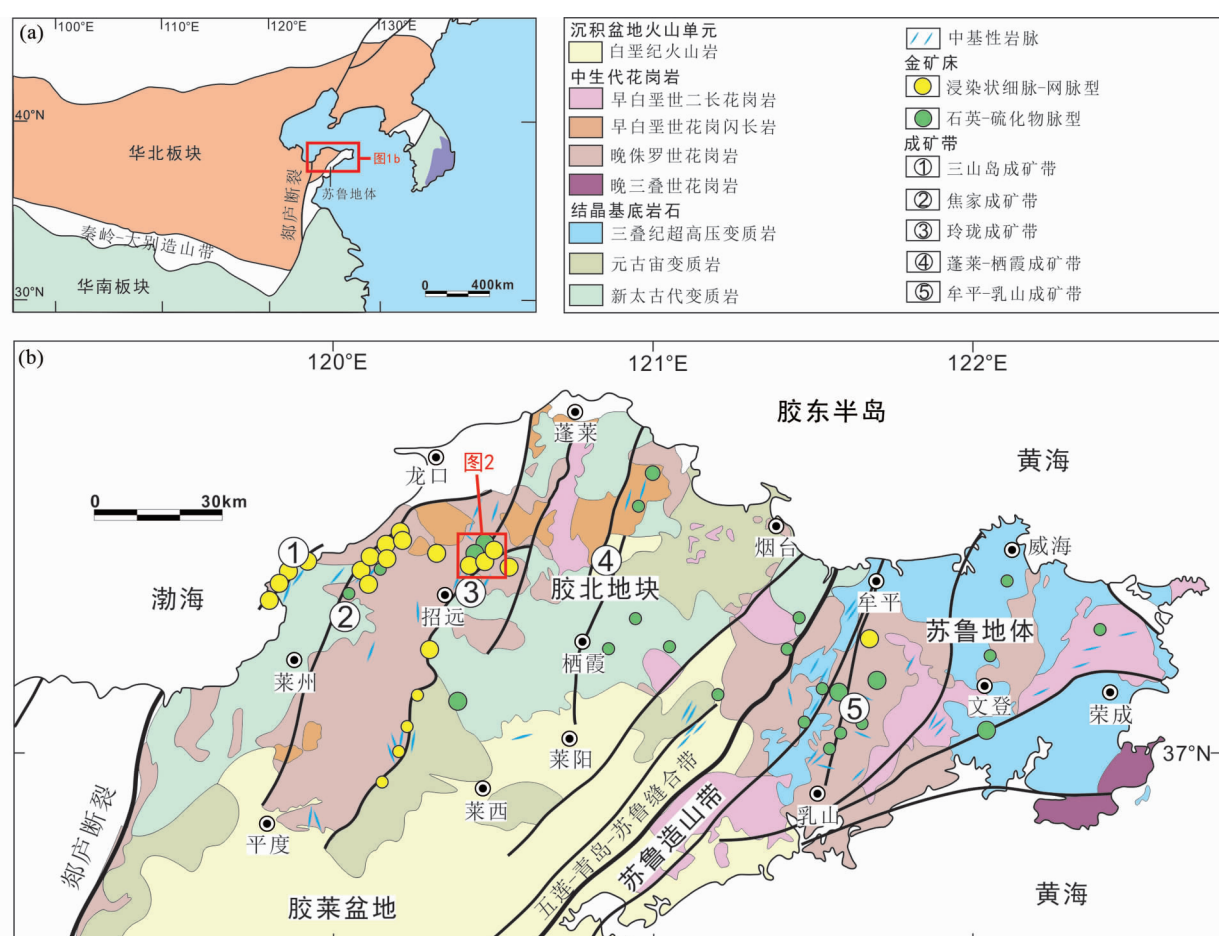


图1 胶东大地构造位置简图(a)和胶东金成矿省区域地质图(b) (据 Zhang *et al.*, 2020 修编)
Fig.1 Simplified geologic map of the tectonic situation of Jiaodong (a) and geological map of Jiaodong gold province (b)
(modified after Zhang *et al.*, 2020)

2014),具有壳幔混合成因特点(Charles *et al.*, 2013)。胶东半岛的镁铁质岩脉分布广泛,但体积通常较小,SHRIMP 锆石 U-Pb 年龄显示基性岩脉侵位时间在 130 ~ 110Ma (Deng *et al.*, 2017),主要来源于岩石圈地幔的低程度部分熔融(Deng *et al.*, 2017)。玲珑花岗岩和郭家岭花岗岩为胶东地区金矿床的主要赋矿围岩(Yang *et al.*, 2014a)。

胶东地区主要存在 EW 向和 NNE-NE 向两组构造体系(图 1b)。EW 向构造主要为古老基底褶皱(Sai *et al.*, 2020),其形成与中生代华北板块与扬子板块陆陆碰撞造成的南北向挤压作用有关(杨立强等,2014; Goldfarb *et al.*, 2021)。NNE-NE 向区域断裂及其次级断裂是胶东半岛的主要控矿构造(Guo *et al.*, 2013;图 1b)。其中郯庐断裂和五莲-烟台断裂是区域一级断裂,NNE-NE 向的二级和三级断裂被认为是郯庐断裂和五莲-烟台断裂的次级断裂(Goldfarb *et al.*, 2001)。自西到东主要有五条断裂带,依次为三山岛-仓上断裂带、焦家断裂带、招远-平度断裂带、蓬莱-栖霞断裂带和牟平-乳山断裂带(赛盛勋和邱昆峰,2020),区域内金矿床的分布严格受这些断裂控制(Deng *et al.*, 2020c; 图 1b)。

2 玲珑金矿区地质特征

玲珑金矿区位于胶东半岛西北部,招远-平度断裂带北段,以石英-硫化物脉为特征的玲珑型金矿而闻名。矿区由西山、东山、九曲、大开头、台上、罗山、阜山、东风等金矿床组成(图 2),金资源总量超过 1000t,属于世界级超大型金矿区。矿区出露的地层主要为新太古代胶东群变质岩和第四纪沉积物。新太古代胶东群变质岩主要出露于矿区以东,也有一些呈透镜状分布于中生代花岗岩中,主要由黑云斜长片麻岩和斜长角闪岩组成(孙华山等,2016)。矿区内中生代岩浆岩十分发育,主要为晚侏罗世玲珑花岗岩和早白垩世郭家岭花岗岩闪长岩。晚侏罗世玲珑花岗岩可分为玲珑黑云母花岗岩和栾家河二长花岗岩(Yang *et al.*, 2012),在破头青断裂两侧分布广泛(图 2)。早白垩世郭家岭花岗岩通常产于矿区西北部。矿区内中基性岩脉十分发育,由煌斑岩、闪长玢岩、石英闪长玢岩组成(Wen *et al.*, 2015),在空间上与金矿体有局部联系。

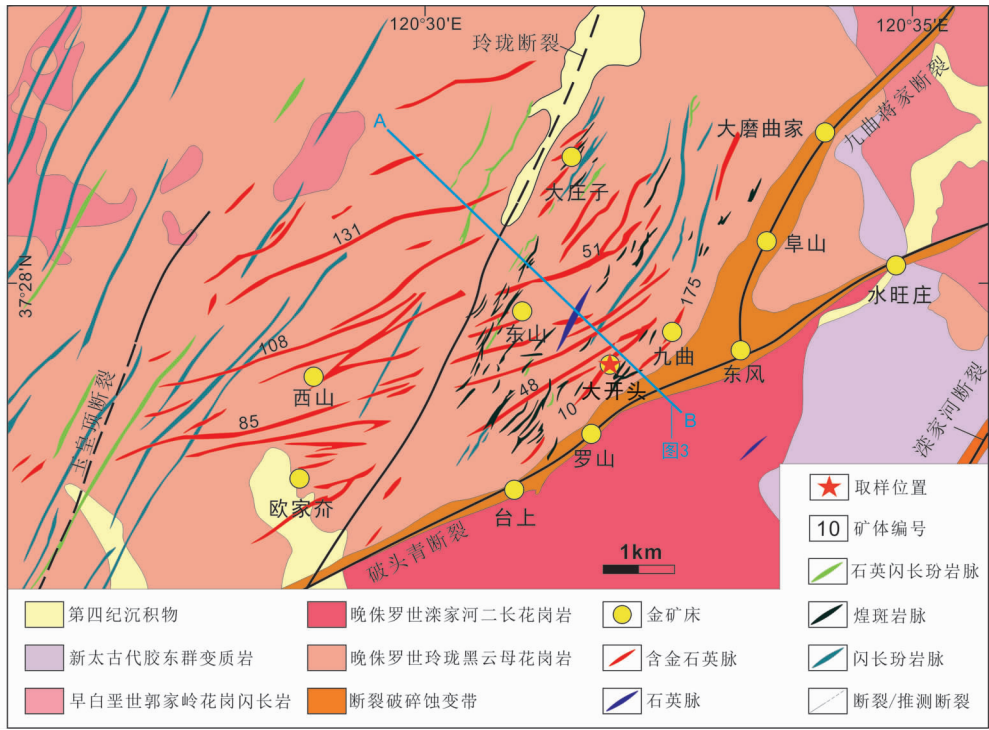


图2 玲琅金矿区地质简图(据 Guo *et al.* , 2020 修编)
Fig.2 Sketch geologic map of Linglong gold district (modified after Guo *et al.* , 2020)

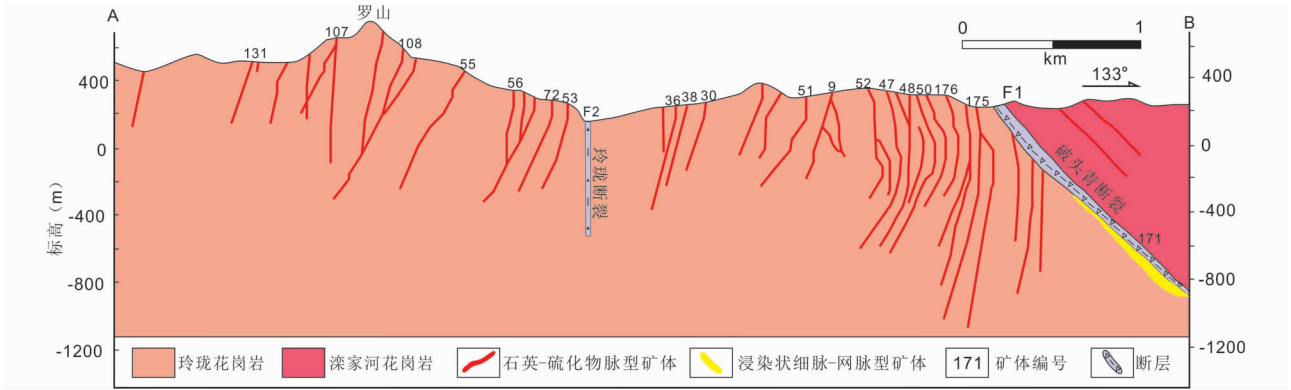


图3 玲琅金矿区剖面图(据 Wen *et al.* , 2015 修编)
Fig.3 Geological profile crossing Linglong gold district (modified after Wen *et al.* , 2015)

矿区内主要发育三条断裂构造,包括破头青断裂、玲琅断裂和九曲蒋家断裂(图2)。破头青断裂属于招远-平度断裂的北段,位于矿区东南部,沿玲琅花岗岩和滦家河花岗岩接触带分布。破头青断裂为成矿前或成矿期形成的主要控矿断裂带,走向为 $50^{\circ}\sim 80^{\circ}$,倾向SE,倾角 $30^{\circ}\sim 60^{\circ}$,控制着台上、罗山、东风和水旺庄等超大型浸染状细脉-网脉型金矿床。沿断裂带发育花岗质碎屑岩和断层泥,断裂带周围有宽阔的热液蚀变带(Guo *et al.* , 2017)。九曲蒋家断裂带走向 35° 左右(申俊峰等,2013),倾向SE,倾角 40° 左右,由位于破头青断裂下盘的一系列次级断裂组成。玲琅断裂走向 $20^{\circ}\sim 30^{\circ}$,倾向SE和NW,倾角 $65^{\circ}\sim 85^{\circ}$,为成矿后断裂,切割玲

琅金矿区中部和破头青断裂。玲琅断裂沿断裂发育花岗质碎屑岩、角砾岩、糜棱岩和少量的断层泥,热液蚀变作用相对较弱(Guo *et al.* , 2017)。矿区内的二级断裂长度一般为数百至数千米,宽度一般为数米至数十米,走向NNE-NEE,倾向NW和SE,倾角 $50^{\circ}\sim 75^{\circ}$,是矿区内控制中基性岩脉和含金石英-硫化物脉产状的主要构造(Guo *et al.* , 2020),产出有东山、西山、九曲、阜山、大开头等玲琅型石英-硫化物脉金矿床。

玲琅矿区内的矿体主要产于玲琅花岗岩和滦家河花岗岩岩体中(张祖青等,2007;Li *et al.* , 2008),玲琅花岗岩中热液蚀变较发育,主要有钾长石化、绢英岩化、硅化、碳酸盐化

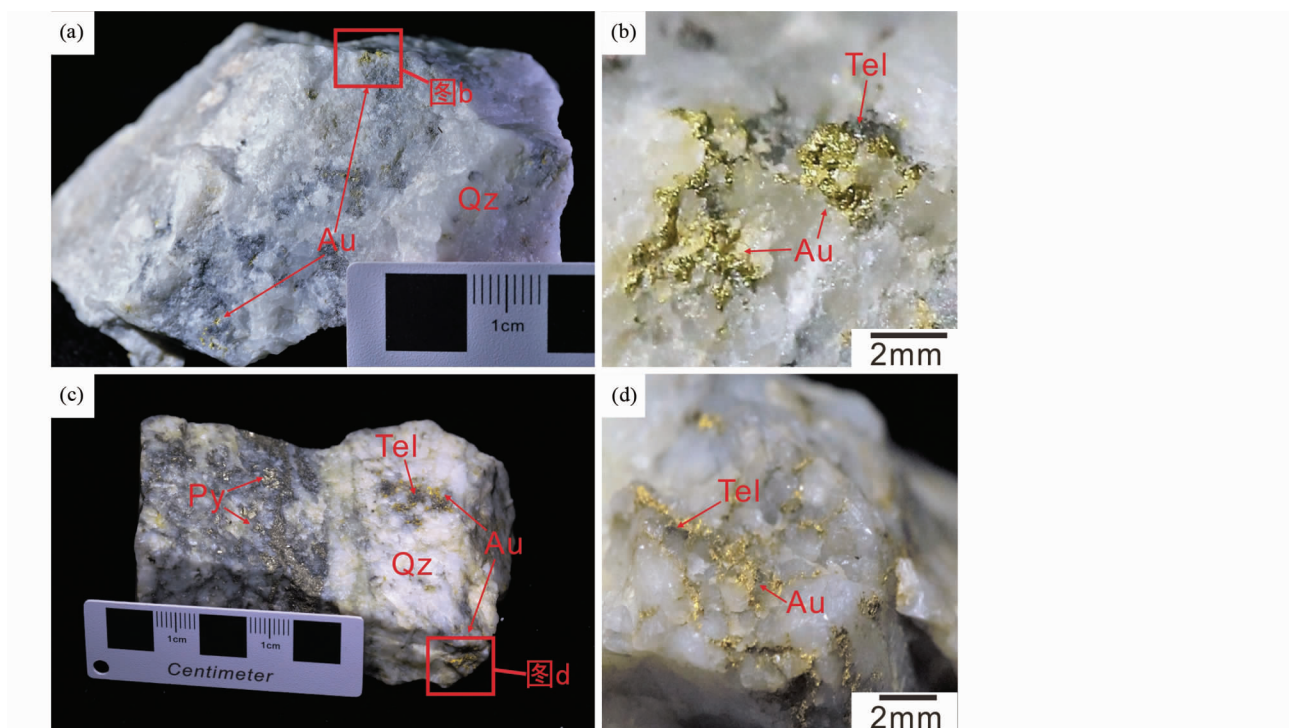


图4 玲珑金矿区含金石英-硫化物脉样品照片

Au-自然金; Tel-碲化物; Qz-石英; Py-黄铁矿

Fig. 4 Photographs of gold-bearing quartz-sulfide vein samples from Linglong gold district

Au-native gold; Tel-tellurides; Qz-quartz; Py-pyrite

等, 蚀变带一般沿矿脉两侧呈对称分布(申俊峰等, 2013)。矿区内已探明金矿脉超过 500 条, 具有经济价值的矿脉约 30 条, 主要为 9、36、47、51、55、56、108 和 175 号矿脉等(图 2、图 3), 这些脉状矿体一般走向 $30^{\circ} \sim 75^{\circ}$, 倾向 NW, 延伸几百米到几千米, 宽度从几十厘米到几米不等。其中 108 号矿脉是石英-硫化物脉型金矿的最具代表性矿体, 矿脉长约 5500m, 走向 $45^{\circ} \sim 65^{\circ}$, 倾向 NW, 倾角 $50^{\circ} \sim 70^{\circ}$, 是玲珑矿区已知的最大石英-硫化物脉矿体, 由宽度 0.3 ~ 3m 不等的含金石英-硫化物脉组成, 周围发育钾长石蚀变和局部绢英岩蚀变, 蚀变宽度在 1 ~ 8m 之间(Guo *et al.*, 2020)。含金石英-硫化物脉的品位通常从几克/吨到十几克/吨不等, 最高可达数百克/吨。主要的矿石矿物为自然金、银金矿和黄铁矿, 其次是黄铜矿、方铅矿和闪锌矿。主要的脉石矿物有石英、绢云母、长石、方解石和绿泥石等。

玲珑金矿区成矿作用可分为 4 个成矿阶段: 黄铁矿-石英阶段、石英-黄铁矿阶段、石英-多金属硫化物阶段和石英-碳酸盐岩阶段(范宏瑞等, 2005)。黄铁矿-石英阶段为成矿早阶段, 基本不含金。石英-黄铁矿阶段和石英-多金属硫化物阶段是金的主成矿阶段。石英-碳酸盐岩阶段为成矿晚阶段偶见少量的自然金产出。

3 样品描述与分析方法

含金的高品位石英-硫化物脉样品采自玲珑矿区大开

头矿段, 为石英-黄铁矿阶段的样品。矿石手标本可见自然金呈浸染状或不规则树枝状集合体赋存于石英-硫化物脉中, 自然金与暗色碲化物共生(图 4)。对 12 个代表性矿石样品进行切面磨制标准探针片进行研究, 矿石探针片在廊坊市地科勘探技术服务有限公司完成。

选取的探针片经表面喷碳处理, 增强导电性后进行扫描电镜和 X 射线能谱及电子探针分析。扫描电镜-能谱分析(SEM-EDS)在核工业北京地质研究院地质矿产研究所岩石矿鉴定实验室完成。使用的仪器为 TESCAN VEGA3 型扫描电子显微镜, 元素面分布(EDS-Mapping)分析采用 X 射线能谱仪, 工作电压为 15kV, 工作距离为 12mm。

电子探针测试工作在山东省地质科学研究院自然资源部金矿成矿过程与资源利用重点实验室完成, 仪器型号为日本电子公司生产的(JEOL) JXA-8230, 所用标准样品均为加拿大 Astimex 系列金属和矿物标样, 具体如下: 金(Au)、银(Ag)、黄铁矿(S、Fe)、方铅矿(Pb)、闪锌矿(Zn)、赤铜矿(Cu)、毒砂(As)、辰砂(Hg)、硒化铋(Bi、Se)、镍黄铁矿(Ni)、辉碲矿(Sb)、钴(Co)等。波谱分析所用加速电压为 20kV, 电流 20nA, 束斑直径 $1\mu\text{m}$ 。实验室温度 22°C 、湿度 30%。相对误差 0.01%。

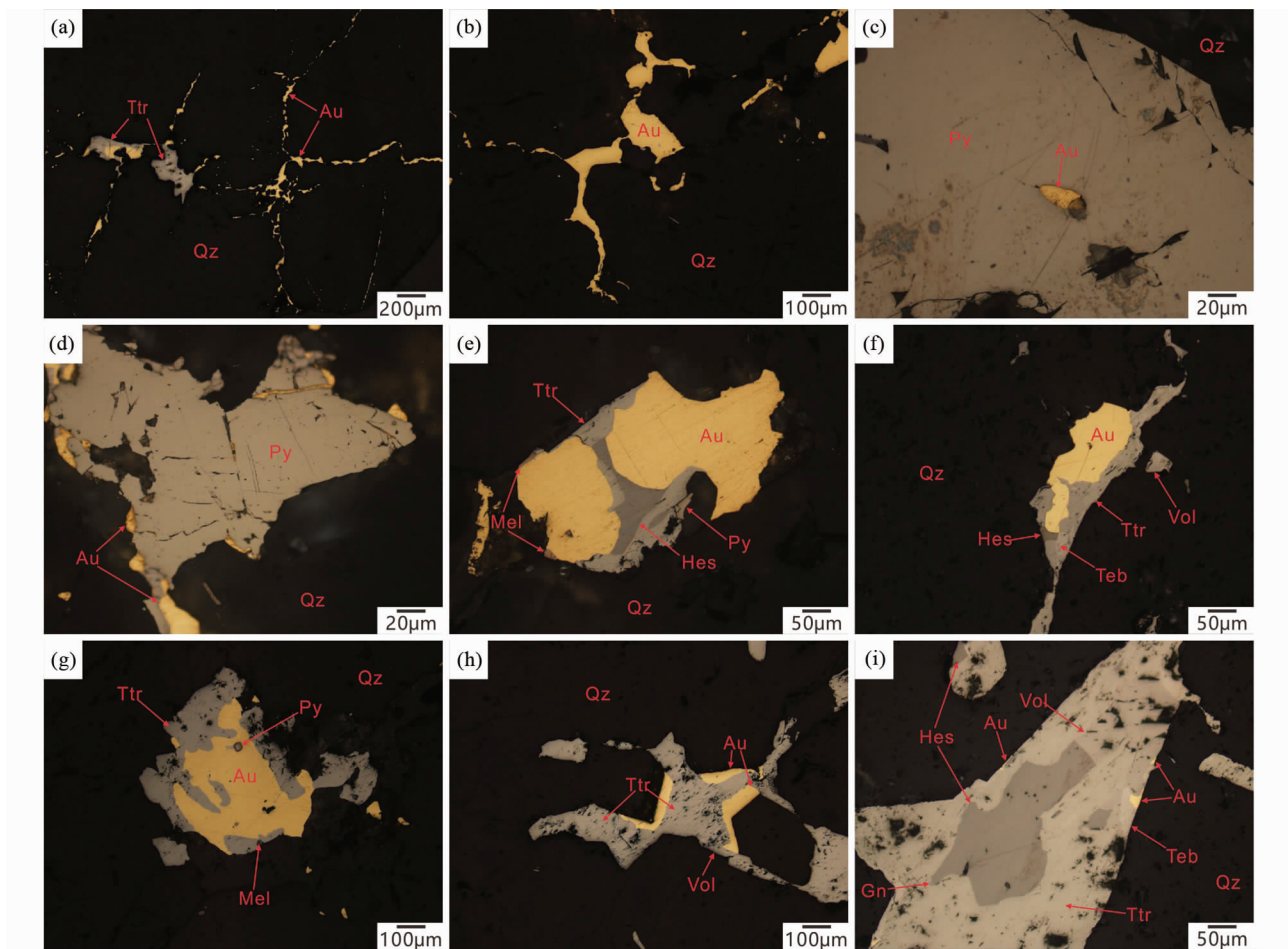


图5 玲珑金矿区自然金赋存状态及共生矿物组合显微照片

(a) 自然金呈细脉状分布于石英中; (b) 石英中不规则状自然金; (c) 黄铁矿中的包体金; (d) 自然金分布于黄铁矿裂隙及黄铁矿-石英间隙中; (e-i) 自然金及碲化物的共生关系。Ttr-辉碲铋矿; Teb-碲铋矿; Mel-碲镍矿; Hes-碲银矿; Vol-碲铋银矿; Gn-方铅矿

Fig. 5 Reflected light photomicrographs showing native gold occurrence and symbiotic mineral assemblages in Linglong gold district (a) veinlet gold in quartz; (b) irregular gold in quartz; (c) inclusion gold in pyrite; (d) native gold is distributed in pyrite fractures and in pyrite-quartz intersections; (e-i) symbiosis between native gold and tellurides. Ttr-tetradymite; Teb-tellurobismuthite; Mel-melonite; Hes-hessite; Vol-volynskite; Gn-galena

4 分析结果

4.1 自然金矿物学和地球化学

自然金呈金黄色,反射率明显高于碲化物和黄铁矿、方铅矿,粒径变化较大,从数微米到数毫米不等。自然金通常与碲化物共生,形成平滑的共结边结构(图5)。自然金主要有三种赋存状态:(1)裂隙金:自然金呈不规则粒状和脉状,赋存于石英和黄铁矿的裂隙中(图5a-b,d);(2)包体金:自然金多呈不规则粒状、浑圆状赋存于黄铁矿中(图5c),少数赋存于石英中;(3)粒间金:多呈不规则状赋存于矿物之间的空隙中,如黄铁矿和石英的间隙(图5d)。

电子探针成分分析显示自然金中 Au 含量为 88.93% ~ 91.06%, Ag 含量为 9.38% ~ 11.83% (表1)。对自然金电子探针数据进行统计,采用目前流行的 $Au/(Au + Ag) \times 1000$

的计算方法来确定金成色,玲珑矿区自然金成色较高,平均成色为 894。根据金银系列矿物的分类,玲珑金矿区自然金属于含银自然金。

4.2 碲化物矿物学和地球化学

根据显微鉴定及电子探针成分分析(表2),共发现了五种碲化物,包括辉碲铋矿、碲铋矿、碲银矿、碲铋银矿和碲镍矿(图5),为玲珑金矿区首次较为详细的碲化物报道。其中辉碲铋矿含量最多,分布最广泛。碲化物与自然金共生,为主成矿阶段产物。

(1)辉碲铋矿:反射色呈铅灰色,与自然金共生,形成平滑共结边结构。本研究测得辉碲铋矿中铋元素含量为 58.69% ~ 61.72%,碲元素含量为 34.53% ~ 35.79%,硫元素含量为 3.83% ~ 4.43%,含有微量的金、银、铁等元素。与标准辉碲铋矿(Bi_2Te_2S : Bi 59.21%, Te 36.26%, S 4.53%)

表 1 自然金电子探针分析结果(wt%)

Table 1 Compositions of native gold detected by EMPA (wt%)

样品号	Au	Ag	Te	Bi	Fe	Co	Ni	Cu	Zn	S	Hg	Cd	Se	Total	金成色
20LL07-8-B-8	91.06	9.73	0.08	0.71	0.01	—	—	—	—	—	0.10	0.06	0.03	101.77	903
20LL10-1-8	90.90	9.38	0.03	0.85	—	—	—	0.06	—	—	—	0.06	0.04	101.32	906
20LL10-1-9	90.69	9.83	0.01	0.80	—	—	—	—	0.05	—	—	0.04	—	101.42	902
20LL02-1-B4	88.93	11.66	0.02	0.76	0.02	—	—	—	—	—	—	0.08	—	101.48	884
20LL02-1-A10	89.09	11.83	0.01	0.76	—	—	—	—	0.03	0.02	—	0.12	—	101.85	883
20LL04-5-2	89.51	10.55	0.04	0.64	—	0.02	—	—	0.03	0.01	—	0.08	—	100.88	895
20LL04-5-6	90.50	10.35	0.05	0.73	—	0.01	—	—	—	—	0.01	0.06	—	101.71	897
20LL07-4-2	88.99	11.01	0.02	0.82	—	0.03	—	—	0.03	—	0.20	0.11	0.03	101.23	890
20LL07-4-3	89.10	10.97	0.10	0.83	0.01	—	0.01	—	0.03	—	0.14	0.11	—	101.33	890
20LL07-4-6	89.49	11.10	0.11	0.78	—	—	0.01	—	—	—	—	0.05	—	101.54	890
20LL07-4-7	89.02	11.40	0.09	0.84	—	0.04	—	0.05	—	—	—	0.07	0.03	101.54	886
20LL07-4-12	90.16	10.59	0.09	0.75	—	—	0.01	—	—	—	—	0.16	—	101.75	895
20LL07-4-19	89.80	10.47	0.05	0.77	—	—	—	0.02	0.04	0.02	0.07	0.05	0.01	101.31	896
20LL12-2-21	89.05	11.19	0.08	0.70	0.35	—	0.02	—	0.03	0.02	—	0.07	0.02	101.53	888
20LL12-2-22	89.54	11.17	0.03	0.71	—	—	—	—	—	—	0.07	—	0.01	101.54	889
20LL12-2-27	89.31	11.26	0.05	0.79	0.02	0.01	—	—	—	—	—	0.05	0.04	101.53	888
20LL7-4-21	90.39	9.42	—	0.72	—	0.03	0.01	0.04	—	0.01	0.06	0.09	—	100.75	906
20LL7-4-22	90.43	9.90	0.08	0.83	—	—	—	0.01	—	—	0.52	—	0.04	101.82	901
20LL7-4-26	90.36	10.04	0.07	0.94	—	—	0.02	0.04	—	—	0.26	0.10	0.01	101.85	900

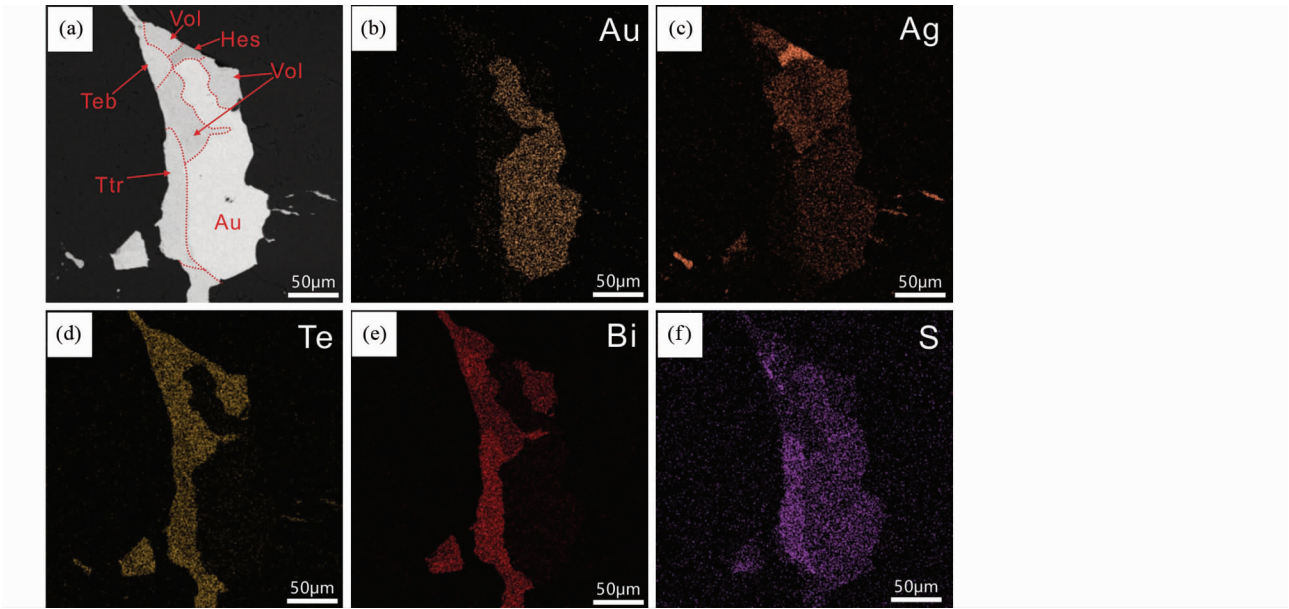


图 6 自然金共生碲化物元素扫面图
(a) 自然金及碲化物 BSE 图像; (b-f) Au、Ag、Te、Bi、S 元素 X 射线能谱扫面图

Fig. 6 X-ray mapping of telluride minerals associated with native gold
(a) BSE image of native gold and tellurides; (b-f) X-ray mapping of Au, Ag, Te, Bi, S elements

对比,相对贫碲、硫,富铋。辉碲铋矿为样品中分布最为广泛,含量最高的碲化物。

(2) 碲铋矿:反射色呈浅灰白色,反射率略高于辉碲铋矿,与辉碲铋矿及自然金共生。本研究测得碲铋矿中铋元素含量为 49.35% ~ 53.88%,碲元素含量为 45.52% ~ 47.23%,含微量的金、银等元素。与标准的碲铋矿(Bi_2Te_3 ;

表 2 碲化物电子探针分析结果(wt%)

Table 2 Compositions of telluride minerals detected by EMPA (wt%)

矿物种类	样品号	Au	Ag	Te	Bi	Fe	Co	Ni	Cu	Zn	S	Hg	Cd	Se	Total
辉铋碲矿 (Bi ₂ Te ₂ S)	20LL07-6-B-7	—	0.06	34.71	59.39	0.02	—	0.01	—	—	4.34	—	0.06	0.07	98.66
	20LL07-6-A-1	—	0.11	34.74	59.99	—	—	—	0.01	—	4.22	—	—	0.04	99.12
	20LL07-6-A-2	—	0.07	34.83	60.29	0.02	0.01	—	—	—	4.20	0.01	—	0.03	99.46
	20LL07-8-B-2	—	0.34	35.72	58.98	0.01	—	—	0.01	0.07	3.83	0.02	—	0.05	99.02
	20LL07-8-A-5	0.05	0.31	35.26	60.78	0.01	—	0.02	0.02	—	4.31	0.08	0.02	—	100.85
	20LL07-8-A-6	0.01	0.20	35.21	60.82	0.02	—	—	—	—	4.43	—	0.06	0.01	100.76
	20LL10-1-6	—	0.14	35.22	61.46	—	—	—	0.01	—	4.31	—	—	0.01	101.16
	20LL10-1-13	0.01	0.29	35.17	61.39	—	0.01	0.01	—	—	4.33	0.13	0.01	0.03	101.38
	20LL02-1-A3	—	0.03	35.79	61.72	—	—	0.01	—	—	4.28	0.04	—	0.01	101.88
	20LL04-5-1	—	0.07	34.57	60.32	—	0.01	—	—	—	4.26	0.03	—	0.10	99.34
	20LL04-5-3	0.02	0.06	35.63	61.33	0.02	0.03	—	0.01	—	4.42	0.06	—	0.03	101.61
	20LL04-5-4	0.07	0.01	35.23	61.28	0.05	—	0.03	0.01	—	4.29	—	—	0.02	100.98
	20LL04-5-7	0.01	0.02	35.36	61.31	—	—	0.03	—	—	4.30	0.14	0.02	0.01	101.20
	20LL04-5-8	—	0.07	35.18	60.56	—	—	0.02	—	—	4.33	—	—	0.02	100.17
	20LL04-5-9	0.01	0.06	35.13	60.76	0.02	0.02	0.03	—	0.04	4.35	—	—	0.06	100.49
	20LL04-5-10	0.02	0.05	34.99	60.96	0.03	—	0.02	—	0.01	4.41	0.07	—	0.04	100.59
	20LL04-5-12	—	0.08	34.93	60.58	—	—	0.01	0.02	—	4.28	—	0.02	0.05	99.98
	20LL04-5-17	0.02	0.03	35.07	60.34	0.01	—	—	—	—	4.28	—	—	0.08	99.83
	20LL07-4-1	—	0.06	34.82	60.16	—	—	—	0.04	—	4.13	0.03	—	—	99.24
	20LL07-4-5	0.01	0.05	34.53	59.90	0.01	—	—	0.06	—	4.18	—	—	—	98.74
碲铋矿 (Bi ₂ Te ₃)	20LL07-4-8	—	0.15	34.98	58.69	—	0.02	—	—	—	4.15	0.05	0.05	0.01	98.09
	20LL07-4-11	0.02	0.09	34.49	59.74	0.03	—	0.04	—	0.01	4.17	0.12	0.04	—	98.74
	20LL7-4-30	—	0.23	34.87	60.68	—	—	0.06	—	0.04	4.25	—	0.06	0.07	100.26
	20LL7-4-31	0.11	0.34	34.49	59.42	—	—	—	0.05	0.06	4.24	—	0.04	—	98.74
	20LL07-6-B-5	—	0.06	46.14	52.71	0.02	—	0.05	0.05	0.01	0.03	—	0.01	0.05	99.13
	20LL07-8-B-1	—	0.12	46.39	53.44	—	—	—	—	—	0.01	0.03	—	0.06	100.05
	20LL07-8-B-7	—	0.39	45.52	52.42	—	0.04	0.01	0.01	—	—	0.01	—	—	98.40
	20LL07-8-B-10	—	0.22	46.37	53.28	0.04	—	0.01	0.02	—	—	—	—	0.06	100.01
	20LL07-8-A-9	0.08	2.30	45.57	51.43	—	0.01	0.03	—	—	0.02	—	—	0.01	99.44
	20LL07-8-A-10	0.04	3.63	44.86	49.35	0.03	0.01	0.04	0.06	—	0.06	—	0.04	0.05	98.16
碲铋银矿 (AgBiTe ₂)	20LL02-1-B3	—	0.06	47.23	53.70	—	—	—	0.05	0.05	—	—	—	0.06	101.14
	20LL02-1-A5	—	0.03	46.10	53.88	—	—	0.03	—	0.06	0.01	0.05	—	0.02	100.18
	20LL02-1-A11	0.02	—	45.97	52.28	0.01	0.02	—	0.04	0.04	0.02	0.06	—	0.03	98.48
	20LL04-5-16	0.11	0.03	46.19	52.51	0.02	—	—	0.04	—	—	—	0.03	—	98.93
	20LL07-6-B-6	—	10.85	47.38	42.66	0.03	—	—	—	—	—	—	0.07	0.02	101.01
	20LL07-6-A-5	0.01	10.77	47.70	43.07	—	0.03	0.04	0.03	—	0.03	—	0.07	0.01	101.74
	20LL07-8-B-11	0.01	18.50	44.18	38.74	—	—	—	—	0.01	—	—	0.08	0.03	101.54
	20LL07-8-A-3	0.19	19.14	43.86	37.78	—	—	—	—	—	0.01	—	0.07	0.06	101.11
	20LL07-8-A-4	0.03	18.71	44.03	38.02	—	—	0.02	—	—	—	—	0.05	0.04	100.90
	20LL07-6-B-1	0.06	63.21	37.52	—	—	—	—	—	—	0.01	0.13	0.37	—	101.32
碲银矿 (AgTe ₂)	20LL07-6-B-2	0.18	63.18	37.48	0.06	0.10	0.03	—	—	—	0.03	—	0.30	—	101.34
	20LL07-6-B-6	0.03	60.00	40.05	0.11	0.01	—	0.01	—	0.01	0.03	—	0.24	0.05	100.52
	20LL07-6-B-8	0.04	60.36	40.80	0.07	—	0.01	0.03	0.01	—	0.02	0.02	0.21	—	101.56
	20LL07-6-B-10	0.06	62.85	37.47	0.03	0.04	—	—	0.03	—	0.04	—	0.30	0.05	100.87
	20LL07-6-A-6	0.14	63.50	37.44	0.07	—	—	—	0.03	—	0.03	0.01	0.36	0.02	101.60

续表 2

Continued Table 2

矿物种类	样品号	Au	Ag	Te	Bi	Fe	Co	Ni	Cu	Zn	S	Hg	Cd	Se	Total
碲银矿 (AgTe ₂)	20LL07-6-A-7	0.05	63.25	37.37	0.09	0.01	—	—	0.02	0.01	0.03	—	0.32	—	101.16
	20LL07-6-A-13	0.03	63.43	37.12	—	—	—	—	0.30	—	0.16	—	0.41	0.08	101.53
	20LL07-8-B-5	0.29	63.21	37.40	0.02	0.01	0.01	0.01	0.04	0.03	0.06	—	0.31	0.04	101.42
	20LL07-8-B-12	0.03	63.63	37.69	—	0.02	—	0.02	—	—	0.02	—	0.32	—	101.72
	20LL07-8-A-1	0.12	63.05	37.07	0.04	0.01	—	0.03	—	0.05	0.07	0.06	0.31	—	100.80
	20LL07-8-A-2	0.19	63.55	37.62	—	0.03	0.01	—	0.01	—	0.03	—	0.31	0.03	101.77
	20LL07-8-A-7	0.11	59.52	35.60	—	—	—	0.01	0.01	—	0.53	—	0.26	0.03	99.25
	20LL07-8-A-8	0.05	62.76	37.10	0.04	—	—	0.01	0.07	—	0.07	0.01	0.38	—	100.67
	20LL10-1-5	0.21	63.40	37.57	0.04	—	—	—	—	—	0.07	0.08	0.36	0.05	101.80
	20LL7-4-24	0.22	63.63	37.58	—	—	—	—	0.02	0.03	0.04	—	0.30	0.03	101.86
	20LL7-4-33	0.34	63.28	36.78	—	—	0.03	—	0.04	0.02	0.13	—	0.30	0.03	100.94
	20LL7-4-34	0.31	63.06	37.09	0.06	—	0.01	—	0.05	0.01	0.10	—	0.35	—	101.05
碲镍矿 (NiTe ₂)	20LL10-1-1	—	0.01	81.32	—	—	—	18.18	0.05	—	0.03	0.05	—	0.07	99.74
	20LL10-1-2	0.02	—	82.06	—	—	—	18.24	0.03	—	—	—	—	0.02	100.37
	20LL10-1-3	—	0.08	81.01	0.02	—	—	18.29	0.02	—	0.01	—	0.02	—	99.44

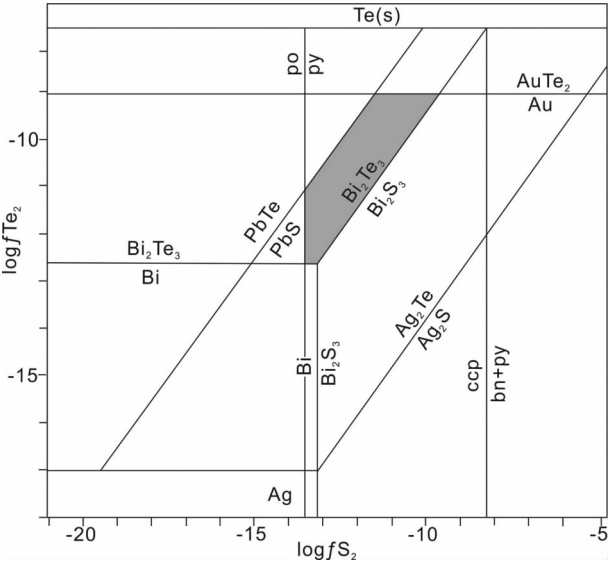


图 7 260℃ 条件下 $\log f_{\text{Te}_2}$ - $\log f_{\text{S}_2}$ 相图 (据 Afifi *et al.*, 1988)

po-磁黄铁矿;bn-斑铜矿;ccp-黄铜矿

Fig. 7 $\log f_{\text{Te}_2}$ vs. $\log f_{\text{S}_2}$ phase diagram at 260℃ (after Afifi *et al.*, 1988)

po-pyrrhotite; bn-bornite; ccp-chalcopyrite

Bi 52.20%,Te 47.80%)对比,相对贫碲。

(3)碲银矿:反射色呈暗灰色,与自然金及其他碲化物共生。本研究测得碲银矿中银元素含量为 59.52% ~ 63.63%,碲元素含量为 35.60% ~ 40.80%,含有微量的金、镉等元素。与标准碲银矿(Ag₂Te:Ag 62.86%,Te 37.14%)对比,除个别样品外,轻微富银。

(4)碲铋银矿:反射色呈浅暗灰色,反射色相较于辉碲铋矿较暗,介于辉碲铋矿和碲银矿之间,常与碲银矿共生(图 6)。本研究测得碲铋银矿中碲元素含量为 43.86% ~ 47.70%,铋元素含量为 37.78% ~ 43.07%,银元素含量为 10.77% ~ 19.14%。与标准碲铋银矿(AgBiTe₂:Te 44.61%,Bi 36.53%,Ag 18.86%)对比,相对富铋,个别样品中贫银。

(5)碲镍矿:相对于其他碲化物含量较低,仅在个别样品中发现,反射色呈淡玫瑰色。本研究测得碲镍矿中碲元素含量为 81.00% ~ 82.06%,镍元素含量为 18.18% ~ 18.29%。与标准碲镍矿(NiTe₂:Te 81.30%,Ni 18.70%)对比,轻微富碲贫镍。

5 讨论

5.1 碲化物形成条件

碲逸度、硫逸度和温度对碲化物的形成起了至关重要的作用。在一定的碲逸度条件下低温有利于碲化物的形成,在相同的温度下高碲逸度对碲化物形成有利(涂光炽,2000;刘家军等,2013)。碲化物对成矿流体的硫逸度、碲逸度、pH 值、温度等条件的高度敏感性,可以作为限制金沉淀时物理化学条件的重要指标(Afifi *et al.*, 1988;McPhail, 1995;Cook and Ciobanu, 2004;Tombros *et al.*, 2010),有助于我们了解金成矿时的物理化学条件。

玲珑金矿区明金矿石中的自然金和碲化物产出于石英-黄铁矿阶段,该阶段流体包裹体均一温度平均为 260℃(李碧乐等,2009;Wen *et al.*, 2015; Yang *et al.*, 2016a; Guo *et al.*, 2017)。结合矿物共生组合和成矿温度,可对碲化物形成时的硫逸度和碲逸度条件进行计算。成矿流体的碲逸度

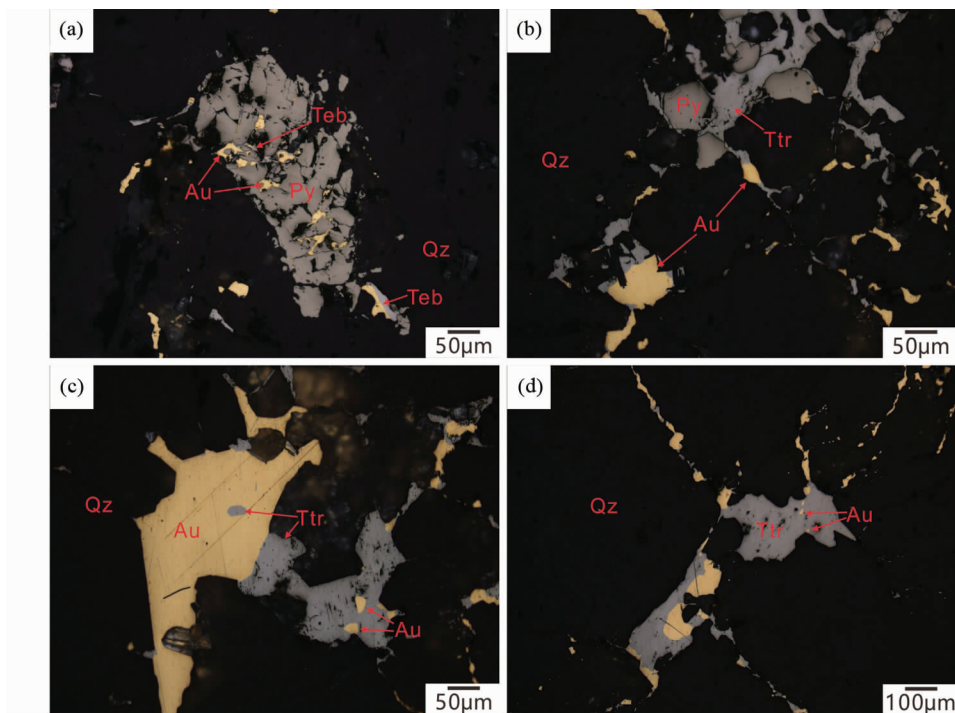


图8 自然金共生碲化物及黄铁矿接触关系显微照片

(a) 自然金及碲化物分布于黄铁矿裂隙中; (b) 碲化物溶蚀黄铁矿; (c, d) 自然金和碲化物的固溶体分离结构

Fig. 8 Reflected light photomicrographs of contact relationship between native gold co-occurrence tellurides and pyrite

(a) native gold and tellurides are distributed in the cracks of pyrite; (b) tellurides corrode pyrite; (c, d) solid solution separation structure of native gold and tellurides

和硫逸度范围通过 Afifi *et al.* (1988) 建立的 $\log f_{\text{Te}_2}$ - $\log f_{\text{S}_2}$ 图解来约束。根据与自然金共生的碲银矿、碲铋矿、辉碲铋矿、碲镍矿、碲铋银矿、及少量黄铁矿和方铅矿等矿物, 而没有出现碲金矿、碲铅矿和磁黄铁矿等矿物, 初步判定玲珑金矿区碲化物形成时期的碲逸度的范围为 $-12.6 < \log f_{\text{Te}_2} < -9.1$, 硫逸度的范围为 $-13.6 < \log f_{\text{S}_2} < -9.6$ (图7)。White *et al.* (1957) 根据实验得出自然金中银含量和温度的关系式, Afifi *et al.* (1988) 基于此关系式提出可通过共生的碲银矿和银金矿来计算成矿流体的碲逸度, 其公式为: $\log f_{\text{Te}_2} = (1/4.576T) \{-50197 + 16.32T - 18.302T \log X_{\text{Ag}} + 4(1 - X_{\text{Ag}})^2 [5650 - 1600(1 - X_{\text{Ag}}) - 1.375T]\}$, 其中 X_{Ag} 为 Ag 在金固溶体中的质量分数, T 为开氏温度。应用该公式, 要求体系中除碲银矿外, 不能出现其他的金银系列碲化物, 以排除早期析出的矿物相在低温状态下分解出碲金矿、碲金银矿等金银系列碲化物的情况。玲珑金矿区除碲银矿外, 未发现与自然金共生的其他碲金银系列矿物, 可应用该关系式。根据玲珑金矿区自然金中 Ag 的质量分数(表1)和成矿温度 260℃, 大致可计算得出成矿流体碲逸度 $\log f_{\text{Te}_2} = -9.52$, 与 $\log f_{\text{Te}_2}$ - $\log f_{\text{S}_2}$ 图解得到的碲逸度范围较为一致。

通常在一定的碲逸度条件下, 低温有利于碲化物的形成。在相同的温度下, 高 $f_{\text{Te}_2}/f_{\text{S}_2}$ 对碲化物形成有利。在玲珑金矿区石英-硫化物脉矿体中, 仅在石英-黄铁矿阶段发现碲化物大量产出。自然金及共生碲化物赋存于石英及黄铁矿

裂隙及间隙中(图8), 自然金及碲化物的形成晚于黄铁矿。推测是由于黄铁矿的形成, 消耗了热液体系中的硫, 硫逸度下降, $f_{\text{Te}_2}/f_{\text{S}_2}$ 升高, 导致碲化物形成。

5.2 成矿物质来源与金富集机制

铋为亲地核地幔元素, 在地壳中丰度较低 (0.16×10^{-6}) (Rudnick and Gao, 2014)。玲珑金矿区大量碲、铋矿物的出现, 指示成矿物质源区可能需要有较高的碲和铋含量。研究表明玲珑金矿区与矿体时空关系较为密切的基性岩脉及围岩花岗岩中铋含量也很低(胡宝群等, 2014), 与地壳中铋的丰度相近。因此, 玲珑金矿区铋矿物的大量富集可能指示成矿物质具有深成源区。碲同样是亲地核地幔元素, 在地壳中丰度极低 (5×10^{-9}) (Wedepohl, 1995), 与碲相关的热液矿床多与地幔或者深部岩浆活动有着密切的联系 (Mao *et al.*, 1995; 毛景文和魏家秀, 2000)。此外 Hein *et al.* (2003) 分析了大量洋底沉积物中的铁锰结核发现其中碲的含量具有极高的异常值 ($0.06 \times 10^{-6} \sim 205 \times 10^{-6}$)。据估计洋底铁锰结核中的碲资源量可达全球陆地的 1.6 倍 (Hein *et al.*, 2020)。结合其他稳定同位素等证据, 本文认为洋壳俯冲过程中, 富碲的洋底沉积物部分熔融可能是玲珑金矿区富碲的重要原因 (Cook *et al.*, 2009b; Harris *et al.*, 2013)。这与 Goldfarb and Santosh (2014) 提出的胶东地区金矿床成矿物质来源与

表 3 胶东地区金矿床中自然金成色

Table 3 Gold fineness of gold deposits in Jiaodong gold province

矿床	矿床类型	金成色(平均)	样品数	资料来源
焦家	浸染状细脉-网脉型	830	11	胡换龙和范宏瑞,2018
		641	—	Yang <i>et al.</i> , 2011
		670 ~ 827	—	宋明春等,2011
		675	31	杜心君和郑若惠,1989
三山岛	浸染状细脉-网脉型	749	123	Peng <i>et al.</i> , 2021
		718	8	杜心君和郑若惠,1989
七里山	浸染状细脉-网脉型	815	12	曾庆文等,2020
新立	浸染状细脉-网脉型	661	4	梁亚运等,2015
新城	浸染状细脉-网脉型	687	15	杜心君和郑若惠,1989
马塘	浸染状细脉-网脉型	685	31	杜心君和郑若惠,1989
灵山沟	石英-硫化物脉型	772	16	杜心君和郑若惠,1989
阎家疃	石英-硫化物脉型	805	8	张琪彬等,2018
旧店	石英-硫化物脉型	744	46	李洪奎等,2015
宋家庄	(浸染状-石英脉)过渡型	744	20	海东婧,2013
谢家沟	(浸染状-石英脉)过渡型	844	73	邓军等,2007
玲珑	石英-硫化物脉型	894	36	本文

古太平洋板块俯冲作用有关的观点一致。

玲珑金矿区流体包裹体研究表明,其成矿期流体性质具有富 CO₂、中温、低盐度 H₂O-CO₂-NaCl ± CH₄ 体系的特征(李碧乐等,2009; Wen *et al.* , 2015; Yang *et al.* , 2016a; Guo *et al.* , 2017)。在这类性质流体中,金最有可能以金硫络合物如 Au(HS)₂⁻ 的形式运移(Pokrovski *et al.* , 2009; Williams *et al.* , 2009; Phillips and Powell, 2010)。由于物理化学条件变化,造成 Au(HS)₂⁻ 络合物的失稳导致金沉淀。但是 Au(HS)₂⁻ 在流体中的溶解度较低,流体中金络合物赋金能力是有限的,要形成明金尺度的大颗粒金是极其困难的,难以解释玲珑金矿区明金矿石中大颗粒自然金的成因。玲珑金矿区明金样品中的自然金普遍与碲-铋矿物紧密共生,自然金与碲-铋矿物形成平滑共结边或固溶体分离结构(图 8),Te、Bi 等元素与金的成矿过程是密切相关的。金虽然有较高的熔点(1064℃),但在有低熔点亲硫元素(LMCE)的参与下,他们所形成的多金属熔体往往具有较低的熔点(Frost *et al.* , 2002)。以 Bi 元素为例,自然铋具有低熔点(271℃),并且 Bi 熔体对 Au 具有极强的吸附能力。在含约 13% Au 的金-铋熔体中,其熔点可低至 241℃(Törmänen and Koski, 2005)。在 Au-Ag-Te 体系中,当熔体中含有 50% Au、15% Ag、和 35% Te 时,共晶熔体的熔点低至 300℃左右(Cabri, 1965)。玲珑金矿区石英-黄铁矿阶段成矿温度或可满足体系中多金属熔点的要求。并且在较低氧逸度或富含 CH₄ 还原性热液体系中,铋熔体可与载金流体共存,而金可有效地从热液流体分配进入到铋熔体相中(Tooth *et al.* , 2008)。热力学模拟实验表明,含有约 20 × 10⁻⁹ 的金热液流体可与含 42% 金的金-铋熔体共存。即使在含有 0.2 × 10⁻⁹ 金的热液

中,也将与含有约 5% 金的熔体共存(Tooth *et al.* , 2011)。因此,即便金在流体中不饱和,在含有铋等 LMCE 存在的情况下,也可以形成有经济开采价值的矿床,可见 LMCE 熔体的赋金能力远超过含金热液流体的赋金能力(Ciobanu *et al.* , 2009)。玲珑金矿区的成矿物理化学条件均满足低熔点多金属熔体的要求,结合观察到的与自然金共生的大量碲铋矿物,意味着金-银-碲-铋熔体是导致大颗粒可见金富集的关键。

玲珑金矿区明金矿石自然金成色较高,平均为 894(表 1),通过对比发现玲珑金矿区自然金成色要略高于胶东其他金矿床自然金成色(表 3)。研究中发现大量与自然金共生的碲银矿、碲铋银矿,可能是由于含银碲化物的形成消耗了体系中的银含量(图 6),提高了自然金的成色。

综上,玲珑金矿区碲、铋的来源与地幔具有较密切关系,与金共生的碲铋矿物大量出现显示了成矿物质深成、幔源的成因信息。碲、铋等元素在金的超级富集机制方面起到了不可忽视的作用,玲珑金矿区的大颗粒自然金的富集可能与金及碲、铋等元素形成低熔点多金属熔体有关。含银碲化物消耗了体系中 Ag 的含量,提高了自然金的成色。

6 结论

(1)玲珑金矿区明金矿石中的金矿物为含银自然金,主要以裂隙金、粒间金、包体金形式赋存,金成色整体较高,平均为 894。共发现了五种与自然金共生的碲化物,分别是辉碲铋矿、碲铋矿、碲银矿、碲铋银矿和碲镍矿,其中辉碲铋矿含量最多。

(2) 碲、铋元素的大量富集,揭示了成矿物质来源具有深成幔源的信息,胶东地区碲、铋等物质来源与古太平洋板块俯冲相关。碲化物形成时成矿流体中碲逸度的范围为 $-12.6 < \log f_{\text{Te}_2} < -9.1$, 硫逸度的范围为 $-13.6 < \log f_{\text{S}_2} < -9.6$ 。

(3) 玲珑矿区大颗粒自然金的富集,与碲、铋等元素结合形成低熔点金属熔体有关,含银碲化物的形成提高了自然金的成色。

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