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氧化性富金斑岩-矽卡岩矿床中碲、硒、铊富集机制的研究进展*

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摘 要 富金斑岩-矽卡岩矿床目前提供了全球几乎所有的硒、碲及部分铊产量，其中氧化性富金斑岩-矽卡岩矿床可共伴生碲、硒、铊等稀散金属。文章从元素地球化学行为、矿床类型、岩浆作用、赋存状态、稀散金属与铜金关系等方面，总结了氧化性富金斑岩-矽卡岩矿床中有关碲、硒、铊富集机制的研究进展。碲、硒具有亲铁和亲硫的特征，而铊具有亲硫和亲石的双重特征，三者具有不同程度的挥发性。岩浆热液型矿床伴生有碲、硒、铊矿化。基性岩浆的注入和岩浆硫化物熔离可能是氧化性富金斑岩-矽卡岩矿床碲、硒、铊富集的主要岩浆作用。氧化性富金斑岩-矽卡岩矿床中铜和金含量通常呈正相关性，发育有丰富的含碲/硒/铊矿物，但碲、硒、铊与铜、金的关系还不清楚。长江中下游成矿带发育多个氧化性矽卡岩金矿、矽卡岩铜金矿，伴生大规模的碲、硒、铊矿化，且已被综合回收利用，该带是探讨氧化性富金矽卡岩矿床中碲、硒、铊富集机制的理想对象。

关键词 地质学；碲、硒、铊；稀散金属矿床；氧化性富金斑岩-矽卡岩矿床；富集机制

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Recent progress in study of enrichment mechanism of tellurium, selenium and thallium from oxidized gold-rich porphyry-skarn deposits

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Abstract

Gold-rich porphyry-skarn deposits provide almost all of the selenium and tellurium production in the world and part of the thallium production at present. Many oxidized gold-rich porphyry-skarn deposits in the world contain dispersed metals as by-products such as tellurium, selenium and thallium. This paper summarizes the research progress of the enrichment mechanism of tellurium, selenium and thallium in the oxidized gold-rich porphyry skarn deposits from the aspects of element geochemistry, deposit type, magmatism, mode of occurrence, and the relationship between dispersed metals, and copper and gold. Tellurium and selenium are characterized by siderophile and chalcophile element, and thallium has the dual characteristics of chalcophile and lithophile element. Tellurium, selenium and thallium have different volatilities. Magmatic-hydrothermal deposit is the major type of tellurium, selenium and thallium-bearing deposit. Basic magma injection and magmatic sulfide segrega-

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tion may be the important magmatism for Te, Se, and Tl enrichment in the oxidized gold-rich porphyry-skarn deposits. Copper and gold values in the oxidized gold-rich porphyry-skarn deposits are typically positively correlated, and various tellurium/selenium/thallium bearing minerals are developed; however, the genetical relationship between tellurium, selenium, thallium, copper and gold is not clear. Many oxidized Au skarn and Cu-Au skarn deposits occur in the Middle-Lower Yangtze River Belt, which contain economic recovered tellurium, selenium and thallium as by-products. Thus, this metallogenic belt is an ideal object for investigating the mechanism of tellurium, selenium, and thallium enrichment in the oxidized skarn Au deposits.

Key words: geology, tellurium, selenium, thallium, dispersed metal deposits, oxidized gold-rich porphyry-skarn deposits, enrichment mechanism

战略性矿产资源包括稀有金属、稀土金属、稀散金属和部分稀贵金属,简称“四稀”(翟明国等, 2019),被发达国家称为关键金属(Gunn, 2014; Schulz et al., 2017; Jowitt et al., 2018)。近年来,发达国家相继发布关键金属矿产清单,制定了关键金属矿产战略保障供应安全(毛景文等, 2019a)。在此背景下,关键金属矿产的成矿机制已成为国际矿床学界的研究前沿。作为关键金属的重要组成部分,稀散金属(也称分散元素) 指在地壳中丰度很低,且在岩石中以极为分散为特征的元素,包括镓、锗、硒、镉、铟、碲、铼和铊(涂光炽等,2004)。在所有“四稀”矿床中,稀散金属矿床的成矿机制研究程度最低。

20世纪60年代前,关于稀散金属的研究以俄文文献为主。涂光炽等(2004)系统回顾了稀散金属的研究历史,特别总结了19世纪末至20世纪60年代期间取得的研究进展;少量英文文献阐明了地质作用中镓、铊、镉和铟的地球化学行为(Shaw, 1952a; 1952b; 1957; Anderson, 1953; Mookherjee, 1962)。自21世纪80年代以来,全球发现了一批稀散金属独立矿床,提高了伴生稀散金属矿床综合利用水平,同时掀起了稀散金属相关研究的热潮(Jowitt et al., 2018)。例如:英国和美国地调查局出版了关键金属手册和全球关键金属矿产资源报告(Gunn, 2014; Schulz et al., 2017);美国经济地质学会出版了关键金属矿床特征专辑(Verplanck et al., 2016)。

相比之下,中国稀散金属矿床的研究相对较晚。1959年孟宪民等(1959)提出中国许多有色金属矿床中发育具工业价值的稀散金属富集,建议加强综合利用的研究;1978年《稀有金属知识》编写组编著出版《稀散金属》;1980年在广州召开了全国首届稀散金属学术会议;涂光炽等(2004)以中国西南低温成矿域为对象,开展了稀散金属矿床成矿机制的研究,突破了“分散元素不能形成独立矿床”的传统观念,首次提出了稀散金属可以独立成矿的理论(涂光炽等, 2004)。近年来,中国部署了包括稀散金属在内的关键矿产方面的科研找矿项目。毛景文等(2019b) 系统总结了中国21世纪以来包含稀散金属在内的关键金属矿床找矿勘查和研究进展;温汉捷等(2019)报道了碲、铟、锗、镓超常富集机制的研究进展;李晓峰等 (2019) 研究了铟在不同地质体中的富集规律。

富金斑岩-矽卡岩矿床指Au品位大于0.4 g/t的矿床,包括铜金矿床、金铜矿床和金矿床。其中,前两者是铜、金的主要来源(Sillitoe, 2000)。金除了可以产于与还原性岩浆相关的热液金矿床中外,亦可以共、伴生于与氧化性岩浆有关的斑岩-矽卡岩铜金矿床中(Thompson et al., 1999; Meinert, 2000),大型还原性斑岩铜金矿床的成矿机制尚存在争议(Sun et al., 2015; 申萍等, 2020)。金可以产于不同类型矽卡岩矿床,可利用的95%的金来自斑岩-矽卡岩铜金矿床、矽卡岩金矿床,这2类矿床统称为富金矽卡岩矿床(Meinert, 1989)。根据含矿岩浆和热液矿物组合差异,富金矽卡岩矿床可分为还原性和氧化性2类,它们的成矿机制类似于斑岩矿床,氧化性矽卡岩金矿床含矿岩浆氧逸度高($f(O_2)$>FMQ+2),以共、伴生铜、碲、硒矿化为特征;而还原性矽卡岩金矿以富铋(Bi>Te(+S, Se))、金品位高和黑铋金矿+自然铋组合为特征 (Meinert, 2000; Ciobanu et al., 2010),铋熔体捕获金的模式较好地解释了矽卡岩矿床中金的富集机制 (Cockerton et al., 2012; Zhou et al., 2017)。据统计,富金斑岩-矽卡岩矿床提供了目前全球几乎所有的硒和碲产量(John et al., 2016),氧化性富金斑岩-矽卡岩矿床常共、伴生碲、硒、铊等稀散金属(谢桂青等, 2019)。但是,这类矿床中关于碲、硒、铊富集机制的研究却相对薄弱。文章从元素地球化学行为、

矿床类型、岩浆作用、赋存状态、稀散金属与铜金的关系等方面,总结了氧化性富金斑岩-矽卡岩矿床中碲、硒、铊富集机制的研究进展。

1 碲、硒、铊地球化学行为

碲、硒、铊具有不同的地球化学行为(刘英俊等,1984),碲、硒具有亲铁和亲硫的特征,而铊具有亲硫和亲石的双重特征(Shaw, 1952a; 1952b; 1957; Sindeeva, 1964; Cook et al., 2009)。w(Te)在地核、地幔和上地壳的值分别为0.85×10^{-6}、0.009×10^{-6}和0.003×10^{-6},地球上96%的碲在地核中(McDonough, 2014; Palme et al., 2014; Goldfarb et al., 2017)。上地壳中w(Tl) (0.9×10^{-6}) 明显高于地核(0.03×10^{-6}) 和地幔(0.0041×10^{-6})(McDonough, 2014; Palme et al., 2014; Rudnick et al., 2014)。这些特征是含碲矿床富集系数大于含铊矿床的主要原因,矿石中碲富集程度最高可达地壳丰度的10^6倍(涂光炽等, 2004)。因此,自然界含碲矿床的数量远大于含硒和含铊矿床(刘家军等, 2020)。

地幔部分熔融形成了超基性岩浆,其w(Te)、w(Se)主要受控于超基性岩浆中硫化物的比例(Hattori et al., 2002)。换言之,硫化物比例是影响铜镍硫化物矿床含碲、硒的主要原因,也可能是斑岩铜矿床中碲含量高的原因(温汉捷等, 2019)。然而,全球多数斑岩-矽卡岩铜金矿床中w(Te)与硫化物含量无明显正相关性(Goldfarb et al., 2017)。地幔包体和地壳岩石中的w(Tl)和w(Pb)呈正相关性,岩浆热液过程中铊主要呈类质同象替代钾、铷进入云母和钾长石,如锂云母的w(Tl)最高达95×10^{-6} (Shaw, 1952a; 1952b; 1957;刘英俊等, 1984; Rader et al., 2018)。全球多个氧化性富金斑岩-矽卡岩矿床同时富集碲、硒、铊,如美国Bingham超大型铜金矿床中平均w(Te)和w(Se)分别为4.8×10^{-6}、12×10^{-6},w(Tl)最高达31.6×10^{-6} (Austin et al., 2010; Fitzpayne et al., 2018)。但是,对氧化性含矿岩浆演化过程中碲、硒、铊的地球化学行为研究尚属空白。

地质作用中碲、硒、铊具有不同程度的挥发性,由强向弱依次为硒、碲、铊(Saunders et al., 2012)。夏威夷岛中自然硫含硒高达5.18% (Stillings, 2017),Kilauea火山1983~1985年喷发10吨硒和2吨碲(Greenland et al., 1986),新西兰北方岛热泉中有高达1000×10^{-6}的铊(Goldfarb et al., 2016)。硒的挥发性大于碲,是美国西部浅成低温金矿床中西硒、东碲分带的重要原因(Saunders et al., 2012)。浅成低温金矿床中碲、硒主要呈气相迁移成矿(Cooke et al., 2001; Zhai et al., 2018),高品位金碲矿石与氧化性岩浆流体密切相关(Keith et al., 2020)。氧化性富金矽卡岩矿床常共、伴生铜矿化,发育Cu-Au→Au-Cu→Au分带 (Zhao et al., 1999),而伴生的碲、硒、铊是否也存在分带性尚未有相关研究。

2 岩浆热液型碲、硒、铊矿床

相比铜等大宗金属矿床,包括碲、硒、铊在内的稀散金属矿床具有"稀、伴、细"的特征(翟明国等, 2019)。前人对含碲、硒矿床成因类型的开展了一些研究,提出不同分类方法。如:Lindgren(1933) 编著矿床学教材提到金-硒化物脉和金-碲化物脉,主要为浅成低温金矿床。Sindeeva(1964) 将含碲、硒的矿床分为岩浆型、火山岩型、热液型和表生型4种。Dill (2010) 将含碲、硒的矿床分为块状硫化物矿床、斑岩-浅成低温矿床和沉积矿床。刘家军等(2020) 将Au-(Ag)-Te-Se成矿系统划分为浅成低温金银矿床、造山金矿床、卡林-类卡林矿床、(偏)碱性侵入岩金矿床、斑岩(铜)金矿床、矽卡岩(铜)金矿床和块状硫化物矿床。Goldfarb等(2017) 提出含碲的矿床分为铜镍铂族元素硫化物矿床、铁氧化铜金矿床、块状硫化物矿床、斑岩铜金矿床、矽卡岩金矿床、浅成低温金银矿床、造山金矿和(类)卡林金矿床,其中铁氧化物铜金矿床、造山金矿床和(类)卡林金矿床中可见碲化物,目前,这些矿床中的碲均未回收利用(Goldfarb et al., 2016; John et al., 2016)。最新资料表明,富碲的造山型金矿床和(类)卡林金矿床与深部的岩浆密切相关(Muntean et al., 2011; Spence-Jones et al., 2018)。

关于硒矿床的成因分类,涂光炽等(2004) 认为硒矿床有岩浆、火山、热液、沉积4种类型。Stillings (2017) 认为含硒的矿床分为铜镍硫化物矿床、含硒化物的热液矿床、沉积型矿床,另外有辉锑矿型含硒矿床(温汉捷等, 2019)。目前这些矿床中硒也均未回收利用(Goldfarb et al., 2016; John et al., 2016)。此外,部分卡林型金矿可共、伴生硒矿化,如中国西秦岭拉尔玛-邛莫金硒矿床(Liu et al., 2000)。

全球共有40余处含铊矿床,品位最富的铊矿床——马其顿Alchar大型金铊锑矿床,其产出的红铊矿粗粒晶体用于太阳中微子探测器(Amthauer et al., 2012)，最近资料表明，马其顿 Allchar 金铊锑矿床属于与侵入岩相关的铊矿床(Palinkaš et al., 2018)。格陵兰 Ilimaussaq 地区发育岩浆热液型铊矿床(Hettmann et al., 2014)。据报道，中国的江西城门山矽卡岩铜金矿床伴生铊矿化(中国有色金属工业协会，2014)。综上所述，大规模的碲、硒、铊矿化以共、伴生形式产于岩浆热液型矿床中。

3 岩浆作用对碲、硒、铊富集的制约

全球金-(银)-碲-硒成矿系统中碲、硒的主要来源是岩浆岩(刘家军等, 2020)。前苏联专家 Sindeeva (1964) 首次出版关于碲/硒矿床的英文专著，书中认为含碲、硒的矿床与岩浆岩密切相关。在所有碲、硒矿床中，浅成低温金矿床碲、硒的富集机制研究程度最高 (Kelley et al., 2016)，特别是金 (银) 碲矿床 (Cooke et al., 2001)和金 (银) -碲化物(硒化物) 矿床 (Cook et al., 2009)，但目前尚未从浅成低温金矿床中回收利用碲(Kelley et al., 2016)。目前全球可回收利用的碲、硒主要来自氧化性斑岩-矽卡岩铜金矿床 (John et al., 2016)，如乌兹别克斯坦 Almalyk 斑岩铜金矿集区有 1098 吨碲(w(Te)$=7\times10^{-6}\sim16\times10^{-6}$) 和 13228 吨硒($w(Se)=3\times10^{-6}\sim117\times10^{-6}$)(U.S. Geological Survey, 2011; Pašava et al., 2010)。对地幔和下地壳包体及含矿岩浆岩的最新研究表明，后俯冲环境氧化性岩浆相关的斑岩铜金矿中碲的来源是交代岩石圈地幔还是新生下地壳还存在争议 (Holwell et al., 2019; Hou et al., 2019)。

最新研究表明，马其顿 Allchar 金铊锑矿床和中国湖北竹林塘金铊矿床均与隐伏侵入岩体密切相关 (Xie et al., 2017; Palinkaš et al., 2018)；美国内华达和华南西部含铊的卡林型金矿系统是隐伏岩体的末端产物 (Muntean et al., 2011; Hu et al., 2017)；格陵兰 Ilimaussaq 地区热液铊矿床与碱性杂岩体密切相关 (Hettmann et al., 2014)；长江中下游多个矽卡岩铜金矿床伴生大规模铊矿化 (谢桂青等, 2019)，如城门山矿床铊资源/储量为大型 (赣西北地质大队内部资料)。但是，对含铊矿床与岩浆作用的关系研究较少。

全球富金斑岩-矽卡岩矿床的铂族元素含量相对较高(He et al., 2017)，钯、铂、碲、硒元素共同富集，碲钯矿($PdTe_2$) 等铂族元素矿物与黄铜矿、斑铜矿共生(Economou-Eliopoulos et al., 2017)，矿石中 w(Pd)和 w(Te)或 w(Se)呈正相关关系，如乌兹别克斯坦 Almalyk 矿集区斑岩铜金矿床(w(Pd)与 w(Se)相关系数为 0.95) 和保加利亚 Elatsite 斑岩铜金矿床(w(Pd)与 w(Te)相关系数为 0.96)(Tarkian et al., 2003; Pašava et al., 2010)。九瑞和大冶矿集区多个氧化性矽卡岩铜金矿床中碲、硒、钯共同富集，如铜绿山矽卡岩铜金矿床中存在碲钯矿，w(Pd)与 w(Au)、w(Cu)、w(Te)具有一定的正相关性(谷湘平等, 1993; 王敏芳等, 2010)。基性岩浆的注入和岩浆硫化物熔离是斑岩铜金矿床中铂族元素富集的主要岩浆作用(Tarkian et al., 2003; Economou-Eliopoulos et al., 2017; Park et al., 2019)。大洋多金属结核中碲、铊资源量分别是全球陆地的 1.6 倍和 6.5 倍(Hein et al., 2020)，在洋陆俯冲过程中，这些稀散金属进入交代岩石圈地幔，地幔的部分熔融和分离结晶形成了富稀散金属的斑岩-矽卡岩矿床的含矿岩浆 (Cook et al., 2009)。

4 碲、硒、铊赋存状态

据 Mindat 网站检索结果，国际矿物学会迄今已批准的含碲、硒、铊矿物分别达 185、136、77 种。前人对浅成低温金矿床中碲/硒化物开展了较深入的研究，如乌兹别克斯坦 Kochbulak 和 Kairagach 浅成低温热液型金矿床中发现有 30 余种碲化物(碲金矿、碲银矿、辉碲铋矿等)、硒化物(硒铅矿、硒铋矿等) 和碲硒化物(Plotinskaya et al., 2006)。还原条件下形成的热液金矿床以富铋和贫碲、硒(Bi>Te(+S, Se)) 及黑铋金矿+自然铋+磁黄铁矿组合为特征；而在氧化条件下则以富碲、硒和贫铋(Te(+S, Se)> Bi) 及金碲化物+自然碲组合为特征(Ciobanu et al., 2010)，表明氧化性富金斑岩-矽卡岩矿床具有丰富的含碲、硒矿物。但是，目前通过部分氧化性富金斑岩-矽卡岩矿床研究显示，其碲、硒元素赋存状态以含碲/硒矿物和类质同像象形式为主(John et al., 2016)，如保加利亚 Elatsite 斑岩铜金矿中有碲银矿、碲钯矿、碲铜矿和碲金银矿等碲化物及硒铅矿、硒银矿和硒铋银矿等硒化物(Tarkian et al., 2003)；美国 Pebble 超大型斑岩铜金矿床可见碲金矿、碲金银矿 (Gregory et al., 2013)。碲化物、硒化物同时产出的现象在全球富金斑岩-矽卡岩矿床并不多见，如鸡冠咀矽卡岩金矿同时有碲铋矿、碲银矿、硒铅矿和硒银矿 (张伟等, 2014)。红铊矿是含铊矿床最主要的矿石矿物，在(类)卡林型金矿和部分镍矿中发现少量红铊矿(Anthony et al., 1990)；马其顿 Allchar 和湖北竹林塘与侵

入岩相关的远端低温金铊矿床中有粗粒红铊矿(1~2 cm) (Xie et al., 2017; Palinkaš et al., 2018)；而格陵兰Ilimaussaq与碱性岩有关的铊矿床中以硫铊铁铜矿、硫锑铊矿等含铊矿物为主(Hettmann et al., 2014)。

除上述含碲/硒/铊矿物，氧化性富金斑岩-矽卡岩矿床中是否存在其他碲/硒/铊矿物相，如纳米颗粒，目前还未有研究。在意大利铁矿石热液黄铁矿和长江中下游香泉铊矿床中均发现了纳米铊颗粒(Fan et al., 2014; George et al., 2019)。

5　碲、硒、铊与铜、金的共生组合关系

富金斑岩-矽卡岩矿床中w(Cu)与w(Au)通常呈正相关性(Meinert, 1998; Sillitoe, 2000)，但对伴生碲、硒、铊的分布规律及其与铜、金的共生组合关系研究相对较少。Lindgren(1933) 的矿床学教材和Sindeeva(1964) 的碲、硒矿床的专著均未涉及到矽卡岩稀散金属矿床。涂光炽等(2004) 提出了稀散金属可以独立成矿的新理论，主要侧重于低温矿床。国际地科联IGCP486项目(2003-2008年)利用碲/硒化物组合探讨岩浆热液作用中金的富集机制，提出铋碲熔体捕获金的模式(Cook et al., 2009)，但未涉及到矽卡岩矿床伴生碲、硒矿化的成矿规律。

富金斑岩-矽卡岩矿床中碲、硒、铊与铜、金的共生组合关系存在争议。有学者研究认为矿石中w(Te)与w(Cu)基本没有相关性(Goldfarb et al., 2017)，如斑岩铜金矿中绢云母化带的w(Te)最高，而钾化带的w(Cu)最高 (Halley et al., 2015)，美国Kalamazoo斑岩铜矿的浅部150~600 m贫铜矿体有最高的w(Te) (Chaffee, 1982)。也有学者认为矿石中w(Te)和w(Cu)呈弱正相关性，如希腊Skouriés斑岩铜金矿中碲和铜含量相关系数为0.54 (Eliopoulos et al., 1991)。富金斑岩-矽卡岩矿床中w(Se)和w(Cu)可能存在正相关性，如乌兹别克斯坦Kalmakyr和保加利亚Elatsite铜金矿w(Se)和w(Cu)的相关系数分别为0.82和0.84 (Tarkian et al., 2003; Pašava et al., 2010)。斑岩-浅成低温铜金矿的热液黄铁矿中w(Te)、w(Cu)、w(Au)均与温度呈反比(Deditius et al., 2014; Keith et al., 2018)。铊是热液金矿床的重要找矿元素(Ikramuddin et al., 1983)，包裹体原位分析数据表明，岩浆流体中w(Tl)与w(Rb)呈正相关(相关系数为0.70)，w(Tl)和w(Cu)呈弱负相关性(Audétat et al., 2019)。斑岩铜金矿高级泥化蚀变岩的w(Tl)最高，不同于富铜的钾化带(Halley et al., 2015)，智利Collahuasi斑岩铜金矿床不同蚀变带中w(Tl)和w(Cu)呈负相关，w(Tl)主要受钾长石分解蚀变控制(Baker et al., 2010)。另外，已有公开资料显示，全球最大的斑岩铜矿带——南美氧化性岩浆相关的超大型斑岩铜矿床中很少伴生碲、硒 (Sindeeva,1964; Goldfarb et al., 2017)，其原因尚不清楚。

6　结　语

中国是全球碲、硒、铊产量最多的国家 (U.S. Geological Survey, 2020)，但资源安全不容乐观，如中国碲需求年增长80%，预测2023年出现供不应求(姜含璐等, 2016)。中国碳酸盐岩面积占全球碳酸盐岩总面积的25%，发育全球最多的矽卡岩矿床，斑岩-矽卡岩铜金矿床数量也较多，多集中于长江中下游成矿带(Chang et al., 2019; Yang et al., 2019)。该带同样集中发育了重要的金矿类型——矽卡岩金矿(陈衍景等, 2004)。已探明多个大中型矽卡岩铜金矿、金矿床(常印佛等, 2017)，是中国“矽卡岩矿床成矿理论”的发源地。该成矿带中24个富金矽卡岩矿床均为氧化性，以含矿岩体富三价铁 (Fe_2O_3/($FeO + Fe_2O_3$)> 0.5)、出现钙铁榴石+透辉石矿物组合、共、伴生铜矿化为特征，明显不同于北美还原性矽卡岩金矿床(Zhao et al., 1999)。成矿带中多个氧化性矽卡岩金矿、矽卡岩铜金矿伴生大规模的碲、硒、铊矿化(谢桂青等, 2019)，如城门山碲资源储量5571吨(中国矿床发现史江西卷编委会, 1996)，是全球公开资料中最大的的碲矿床。已有相关企业对这些矿床中碲、硒进行综合利用，如大冶有色金属集团和江西铜业集团公司每年分别冶炼出35吨碲和197吨硒、55吨碲(内部报告)。全球对氧化性矽卡岩铜金矿与金矿中伴生碲、硒、铊含量和成矿特征的差异研究基本空白，以九瑞矿集区丰山矿田为例，建立了氧化性矽卡岩铜金碲矿+远接触带低温金铊矿床组合模型(Xie et al., 2019; Han et al., 2020)。因此，长江中下游成矿带是探讨氧化性富金矽卡岩矿床中碲、硒、铊富集机制的理想对象，中国学者有望做出重要贡献。

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