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云南金顶超大型铅锌矿床沥青 Re-Os 法测年及地质意义*

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Abstract Oil-gas reservoirs and metal deposits often co-exist in many sedimentary basins, and their genetic relations have been paid much attention. The Jinding Zn-Pb deposit, Yunnan, China, is so far the largest Zn-Pb deposit in China, the youngest and only giant Zn-Pb deposit hosted in continental sedimentary rocks in the world. Bitumen and heavy oil were often observed in the Jinding Zn-Pb deposit. It has been a subject of debate whether the bitumen formed before or after the Zn-Pb mineralization, making it difficult to evaluate and the genetic relationship between the bitumen and Zn-Pb mineralization. The bitumen in the Jinding Zn-Pb ores hosted in the breccia-bearing sandstones and sandy breccias of the Paleocene Yunlong Formation has been dated by the Re-Os method in this paper, and an isochron age of 68 ± 5Ma (MSWD = 9.2, n = 6) has been obtained. Therefore, the oil-gas reservoir in the Jinding ore district was formed earlier than the Zn-Pb mineralization. The hydrocarbons in the reservoirs may have provided the condition for the production of reduced sulfur required for Zn-Pb mineralization through thermal chemical reduction of sulfates. The formation of the oil-gas reservoir and the Zn-Pb deposit may have been a continuous geologic process, and the oil-gas reservoir was one of the basic conditions of Zn-Pb mineralization in the Jinding ore district. The oil-gas reservoir was destructed by the mineralization process.

Key words Zinc-lead ores hosted in the sandy breccias; Bitumen; Re-Os dating; The Jinding giant Zn-Pb deposit; Lanping, Yunnan

摘 要 油气藏与金属矿床在世界许多沉积盆地内共存，油气成藏与金属成矿的动力学关系备受关注。云南兰坪金顶产有中国目前最大铅锌矿床，也是世界上唯一陆相沉积岩容矿、且形成于新生代的超大型铅锌矿床。矿床中常见沥青、重油等有机质，它们的形成早于或晚于铅锌硫化物成矿存在明显分歧，限制了对油气成藏与铅锌成矿关系的认识。本文针对金顶超大型矿区以古新统云龙组含砾砂岩和砂砾岩为主岩铅锌矿石中沥青，开展了 Re-Os 法同位素测年，获得 68 ± 5Ma 的等时线年龄(MSWD = 9.2, n = 6)，指示金顶古油气成藏形成于古新世，先于铅锌硫化物大规模成矿；烃类物质具有通过热化学还原硫酸盐提供铅锌成矿所需硫化氢的客观条件；油气成藏与铅锌成矿在云南金顶矿区很可能是一个先后发生的连续地质过程，成藏为成矿奠基，成矿伴随着油气藏的破坏。

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关键词 砂砾岩容矿铅锌矿石;沥青;Re-Os 同位素测年;金顶超大型铅锌矿床;云南兰坪
中图法分类号 P597.3

1 引言

世界各地沉积岩容矿的铅锌矿床中多见有机质,它们与金属成矿的关系倍受关注(Dianar and Sureau, 1990;Chi *et al.*,1995;Sicree and Barnes,1996;Disnar,1996;谢树成等,1997;Bartrick and Andrew,1997;Pangenberg and Macko,1998;殷鸿福等,1999;Spangenberg *et al.*,1999;Ulrich *et al.*,1999;Gusfkiewicz and Kwiecinska,2001; Fallick *et al.*,2001;薛春纪等,2002a, 2007a, 2009;朱弟成等,2003;Southam and Saunders,2005;高永宝等,2008a;顾雪祥等;2010)。云南金顶铅锌矿床是目前中国最大铅锌矿床,也是世界上唯一陆相沉积岩容矿、且形成于新生代的超大型铅锌矿床(Xue *et al.*, 2000,2003),代表了沉积岩容矿铅锌矿床的一种新类型(Kyle and Li,2002;Xue *et al.*,2004,2006,2007c;薛春纪等,2007b);矿区矿石、围岩及矿物流体包裹体中多见有机质(胡明安,1989a;薛春纪等,2002a, 2007a, 2009;王大锐和张抗,2003;常象春和张金亮,2003;付修根,2004;Xue *et al.*,2007c;高永宝等,2008a),是认识有机质与金属成矿关系的重要对象。

对金顶铅锌矿床的成因研究存在"同生沉积-后期改造层控矿床"(白嘉芬等,1985)、"中低温非岩浆热液成矿"(高广立,1989)、"同生沉积-变形叠加成矿"(吴淦国和吴习东,1989)、"喷气(热液)沉积成矿"(赵兴元,1989)、"岩溶成矿"(胡明安,1989b)、"壳幔流体混合成矿"(尹汉辉等,1990;王京彬和李朝阳,1991;Xue *et al.*,2000,2003,2006,2007c;薛春纪等,2002a, b, 2007b;Chi *et al.*,2007;Wang *et al.*, 2010)等颇多分歧。从有机质的岩相学、有机地球化学和油气成藏条件分析,推测矿区铅锌成矿前曾存在油气藏形成(薛春纪等,2007a, 2009;高永宝等,2008a),并认为油气对硫酸盐的热化学还原形成大量硫化氢,从而导致铅锌硫化物大量沉淀成矿(高永宝等,2008b;薛春纪等,2009);但有学者依据矿区有细脉状产出的沥青等特点,认为有机质是铅锌硫化物成矿后运移到矿区的(Leach, 2010,个人交流),很可能与金属成矿关系不大。本文对金顶铅锌矿床中的沥青开展了 Re-Os 法同位素测年,试图为探索金顶有机质与金属成矿关系提供新的依据。

2 地质背景和矿床地质

金顶铅锌矿床产在西南三江中段兰坪盆地内。兰坪盆地隶属昌都-思茅微板块,东侧以金沙江断裂带与扬子板块相接,西侧以澜沧江断裂带与保山地块毗邻;在古特提斯基础上沉积了中-新生界海相、陆相碳酸盐岩、火山碎屑岩和碎屑岩建造,地层中有多个陆相膏盐层,存在多个沉积间断(薛春纪等,2002a,b;)。受古金沙江洋和古澜沧江洋相向俯冲以及印度-欧亚板块碰撞影响,兰坪盆地印支期具有残留海性质,燕山期是拗陷盆地,喜马拉雅期属走滑拉分盆地(Xue *et al.*,2003,2004)。板内构造体制下的深大断裂和岩浆活动、地幔扰动和地幔流体上涌、地层中不整合及壳幔相互作用等是金顶铅锌成矿的基本地质背景(Xue *et al.*, 2004, 2007c)。

金顶铅锌矿田先后大致经历了中-新生界沉积、推覆构造、局部穹隆以及穹隆破裂等地质过程(薛春纪等,2002b;Xue *et al.*,2007c),油气成藏和金属成矿可能伴随推覆构造、穹隆化和热液活动先后发生(薛春纪等,2007a, 2009;高永宝等,2008a)。矿区发育多个推覆构造,较老地层多被推覆到较新地层之上;局部穹隆化使推覆构造面和它上下的地层发生变形,形成金顶穹隆(吴淦国和吴习农,1989;薛春纪等,2002b)。穹隆核心由中白垩统虎头寺组(K_2h)灰色砂岩和古新统云龙组(E_1y)红色砂砾岩正常层序、即原地系统构成,围绕穹隆核心向外围依次为下白垩统景星组(K_1j)灰色细砂岩、中侏罗统花开左组(J_2h)杂色泥岩和粉砂泥岩、上三叠统麦初箐组(T_3m)碳质泥岩和三合洞组(T_3s)碳质灰岩等倒转层序构成的外来系统(图 1)。铅锌硫化物后生热液矿化主要发生在主推覆构造面上下的 K_1j 砂岩和 E_1y 砂砾岩中,矿体呈板状、脉状,主要集中在穹隆近核部(图 1)。典型矿石结构是闪锌矿、方铅矿、黄铁矿等硫化物热液矿物交代碎屑岩中钙质胶结物形成的胶结结构,硫化物矿物它形微晶-细晶状,后生热液成矿特征明显。成矿温度为 100 ~ 250℃,成矿深度 1.0km 左右(薛春纪等,2002b)。

3 矿区有机物质

金顶矿区岩石和矿石中有机质及古油气藏遗迹多见(薛春纪等,2007a, 2009;高永宝等,2008a)。T_3s 中沥青灰岩多见,夹有炭质泥灰岩及炭质泥岩薄层,沥青含量在 1% ~25% 不等(云南省地质局第三地质大队,1984[①]),它们富炭色黑,有机质多为含泥沥青和干酪根,显微镜下集合成黑色条带和层纹。矿区 T_3m 中常见有黑色碳质泥岩和碳质泥灰岩,含炭化植物碎片。J_2h 中发育黑色碳质泥岩。矿区 K_1j 砂岩基本全层发生铅锌硫化物矿化,矿石和矿化砂岩经常会嗅到石油味;打开标本时,常会观察到石油从某个集中点向四周扩散,并且同时嗅到浓烈的汽油味;K_1j 矿化砂岩中也常见到黑色有机物质斑点团块状、浸染状分布,显微镜下有机质个体不

① 云南省地质局第三地质大队. 1984. 云南省兰坪县金顶铅锌矿详细勘探地质报告

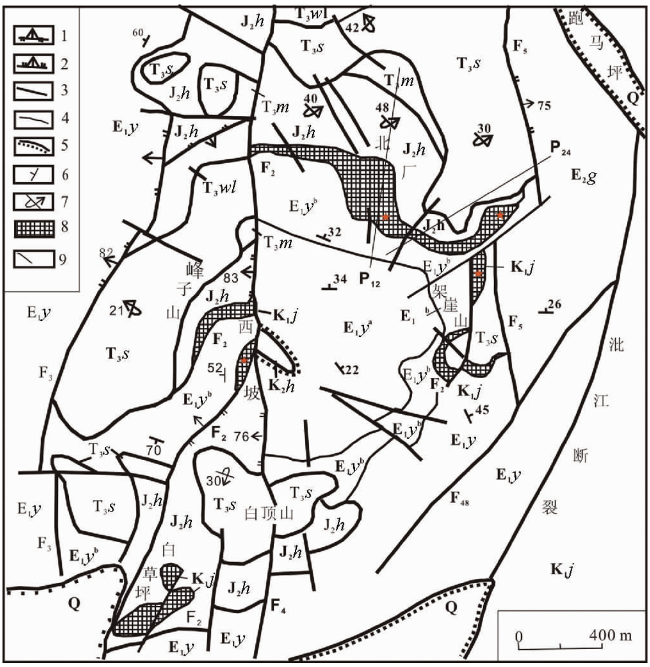

图 1 金顶超大型铅锌矿床地质图(据薛春纪等,2002b)

Q-第四系；E_2g-始新统果朗组岩屑石英砂岩；E_1y-古新统云龙组；E_1y^a-云龙组下段粉砂泥岩；E_1y^b-云龙组上段砾岩和砂岩；K_2h-中白垩统虎头寺组石英砂岩及粉砂岩；K_1j-下白垩统景新组砂岩；J_2h-中侏罗统花开左组粉砂质泥岩；T_3m-上三叠统麦初箐组粉砂-细砂岩；T_3wl-上三叠统挖鲁八组泥岩河粉砂岩；T_3s-上三叠统灰岩；1-逆冲推覆断裂；2-正断层；3-性质不明断层；4-地质界线；5-不整合界面；6-正常岩层产状；7-倒转岩层产状；8-铅锌矿体；9-勘探线及编号；五角星-铅锌硫化物矿石中沥青采样和观察点

Fig. 1 The geological map of the Jinding Zn-Pb ore deposit (after Xue *et al.*, 2002b)

Q-Quaternary; E_2g-Eocene Guolang Formation sandstone and siltstone; E_1y-Paleocene Yunlong Formation; E_1y^a- Lower member of Yunlong Formation siltstone and mudstone; E_1y^b- Upper member of Yunlong Formation breccia and sandstone; K_2h-Middel Cretaceous Hutousi Formation sandstone; K_1j-Lower Cretaceous Jingxing Formation sandstone; J_2h-Middle Jurassic Huakaizuo Formation siltstone and mudstone; T_3m-Upper Triassic Maichuqing Formation shale and siltstone; T_3wl-Upper Triassic Waluba Formation; T_3s-Upper Triassic Sanhedong Formation limestone; 1-thrust fault; 2-normal fault; 3-indeterminate fault; 4-geological boundary; 5-unconformit; 6-normal stratigraphic sequence; 7-reversed stratigraphic sequence; 8-Zn-Pb orebody; 9-exploratory line and number; Star-sample location and viewpoint of the bitumen in Zn- and Pb-sulfide minerals

定形,浸染状、团块状分布并与闪锌矿伴生;K_1j 砂岩钙质胶结物常被闪锌矿等硫化物矿物交代,并且伴生有沥青。

金顶矿床所在区域的 E_1y 中不含明显有机物质,而矿区 E_1y 含砾细砂岩岩层上部发生铅锌硫化物矿化,多黑色或深色,更常嗅到的石油气味比在 K_1j 矿化砂岩中更加浓烈。野外新打开岩、矿石标本时,嗅到的石油气味使人感到刺激难忍。在角砾岩型矿石的空洞格架或裂隙内发现黑褐色粘稠原油物质(重油),具有石油气味,新打开的岩、矿石新鲜面可见黑褐色重油从空洞中慢慢渗出(图 2c, d)。角砾岩型矿石内空洞或晶洞及其附近常见黑色玻璃状(脆)沥青(图 2a, b),它们很可能是石油等有机物质在后来成矿作用热影响下成熟的产物。矿区架崖山矿段露天采场多处可见古油气藏遗迹,有机质热成熟现象显著。从区域到矿区,不同形式和成熟度的有机物质可能是原始有机物质降解、生烃、运移、储集、成藏、热成熟、裂解及构造热流体破坏影响等不同阶段的产物(薛春纪等,2007a;高永宝等,2008a),成藏应早于金属成矿作用(薛春纪等,2009)。

尤其在以云龙组(E_1y)含砾砂岩和砂砾岩为主岩铅锌硫化物矿石的角砾格架和空洞中,沥青和重油常集中产出(图 2),这些有机质的岩相学和有机地球化学特征已专门论述铅锌硫化物矿体主岩、即云龙组(E_1y)含砾砂岩和砂砾岩中角砾次棱角状,分选差,主要成分是来自上三叠统三合洞组(T_3s)碳质灰岩、沥青灰岩和麦初箐组(T_3m)泥灰岩、碳质泥岩,少部分为中侏罗统花开左组(J_2h)杂色泥岩和粉砂泥岩;填隙物为含岩屑含长石石英细砂岩。铅锌硫化物矿化发生在填隙物、即含岩屑含长石石英细砂岩中,同样表现为闪锌矿、方铅矿、黄铁矿等硫化物热液矿物交代细砂岩中方解石胶结物形成胶结结构的后生热液成矿特征,含砾砂岩和砂砾岩中角砾常被交代成不规则边界(薛春纪等,2002b)。

4 样品和 Re-Os 法测年

本次用于 Re-Os 法测年的沥青样品采自金顶铅锌矿区架崖山矿段露天采坑底部(图 1),为以云龙组(E_1y)含砾砂岩和砂砾岩为主岩铅锌硫化物矿石角砾格架中的沥青(图 2a, b)。野外采样中,先选择采坑底部新剥露出的较新鲜矿体部位,使用木质工具逐粒采集沥青 3 ~ 5g,用拉边封口塑料袋存放样品;实验室内将样品用木质工具稍作粉碎后,在双目实体显微镜下针对 0.5 ~ 1.0mm 沥青颗粒逐粒检查其纯度,排除有连生或混入其他杂质矿物的颗粒,使样品沥青纯度达到 99.5% 以上;然后,在超声波中清洗样品,以除去沥青样品表面吸附的粉尘等杂质,以备测年。由于没有能够获得足够量纯度较高的重油样品(图 2c, d),本次没有对铅锌硫化物矿石角砾空洞中的重油开展测年工作。

准确称取 0.2g 备好的沥青样品,将其转入 Carius 管中,加入氧化剂(3mL 盐酸,5mL 硝酸,1mL 双氧水)和稀释剂,在 200℃封闭溶样 24h(李超等,2011)。用蒸馏法分离 Os(杜安道等,1994),用丙酮萃取法分离 Re(李超等,2009)。Re、Os 的制备液在 HR-element2-ICP-MS 分析,分析结果见表 1。Re、Os 含量的不确定度包括样品和稀释剂称量误差、同位素组成误差、稀释剂的标定误差、质谱分析的分馏校正误差和

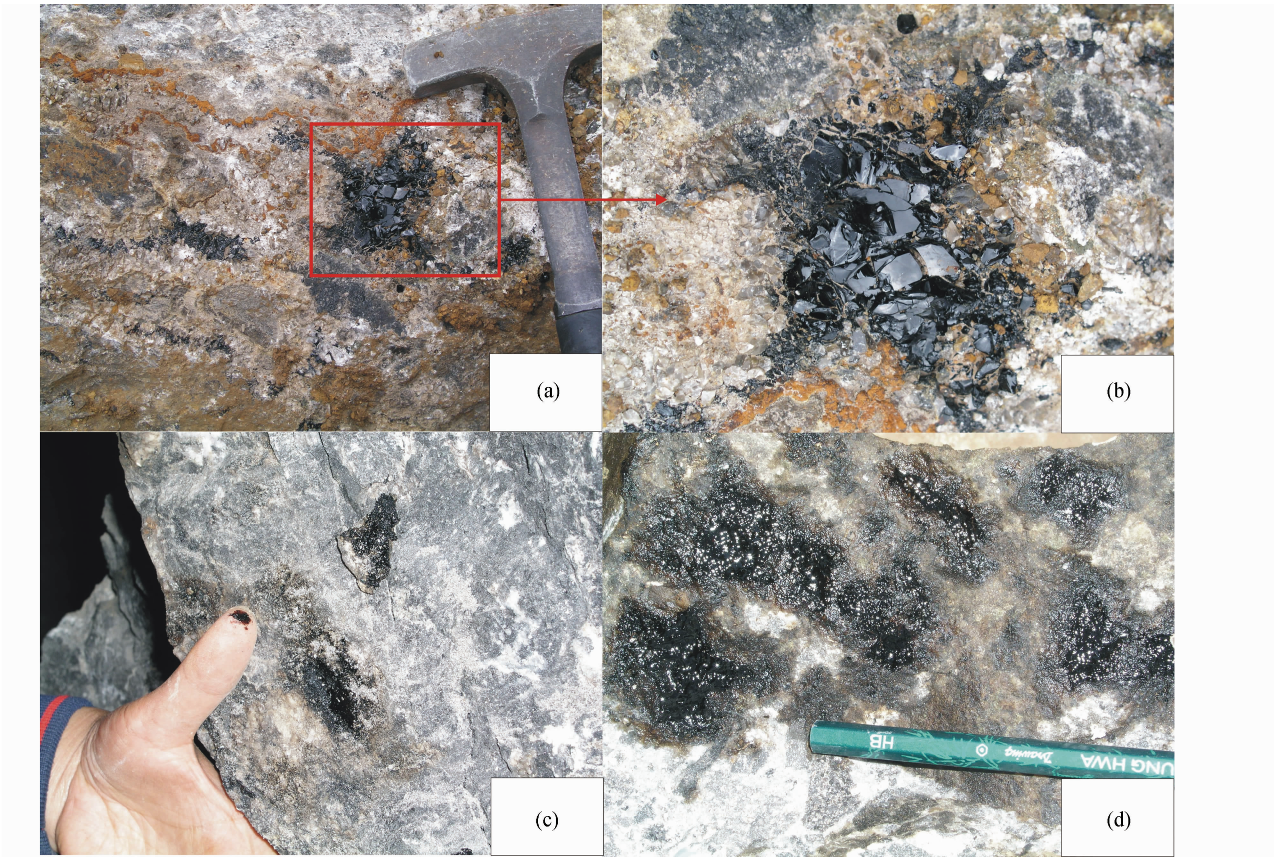

图 2 金顶矿区铅锌硫化物矿化含砾砂岩和砂砾中岩角砾格架内的沥青(a、b)和空洞内的重油(c、d)

Fig. 2 the bitumen (a, b) in the grilles and the heavy oil (c, d) in the cavity of breccia-bearing sandstone and sandy breccia bearing Zn- and Pb-sulfide minerals, Jinding

表 1 金顶铅锌矿石中沥青的 Re、Os 同位素组成分析结果数据

Table 1 The analysis results of Re-Os isotopic compositions of the bitumen in the Jinding zinc-lead ores

样品号	样重(g)	Re($\times10^{-9}$)		普通 Os($\times10^{-9}$)		^{187}Os($\times10^{-9}$)		^{187}Re/^{188}Os		^{187}Os/^{188}Os	
		测定值	不确定度	测定值	不确定度	测定值	不确定度	测定值	不确定度	测定值	不确定度
JYS-11	0.201	71.30	0.6	0.1299	0.0063	0.1164	0.0112	2651	131	6.886	0.739
JYS-1	0.200	131.9	1.2	0.1027	0.0023	0.1424	0.0013	6208	148	10.66	0.23
JBC-19	0.207	542.8	6.5	1.027	0.008	0.9646	0.0084	2553	37	7.219	0.042
JYS-2	0.200	404.4	4.6	0.0487	0.0036	0.3094	0.0030	40085	3013	48.80	3.62
JYS-3	0.204	340.9	5.1	0.0583	0.0010	0.2727	0.0026	28233	656	35.93	0.62
JYS-4	0.143	89.31	0.81	0.1161	0.011	0.1028	0.001	3715	360	6.802	0.657

注:由国家地质实验测试中心分析,分析者高炳宇、李超、杜安道;用蒸馏法分离 Os,用丙酮萃取法分离 Re,在 HR-element2-ICP-MS 完成 Re、Os 同位素组成分析

待分析样品同位素比值误差。整个流程的空白平均值 Re 为 4pg,普通 Os 为 0.3pg,^{187}Os 为 0.03pg。空白相对样品中 Re、Os 含量,可以忽略不计。沥青样品中 Re 的含量在 $71.30\times10^{-9}\sim404.4\times10^{-9}$ 之间,普通 Os 和 ^{187}Os 的含量分别为 $0.0487\times10^{-9}\sim1.027\times10^{-9}$ 和 $0.1028\times10^{-9}\sim0.9646\times10^{-9}$。

金顶铅锌硫化物矿石中沥青的 Re、Os 同位素组成分析数据(表 1)用 isoplot 软件(Ludwig, 2003)处理,获得金顶沥青 Re-Os 同位素等时线(图 3)年龄为 68±5Ma,^{187}Os/^{188}Os 初始比值为 4±1,MSWD=9.2。

5 讨论

沉积岩中的有机质多与海相还原环境有关。海水中 Re 在氧化条件下以 ReO_4^- 形式存在,溶解度大,容易迁移;而在富有机质沉积岩形成的还原环境,海水中 ReO_4^- 被还原成较

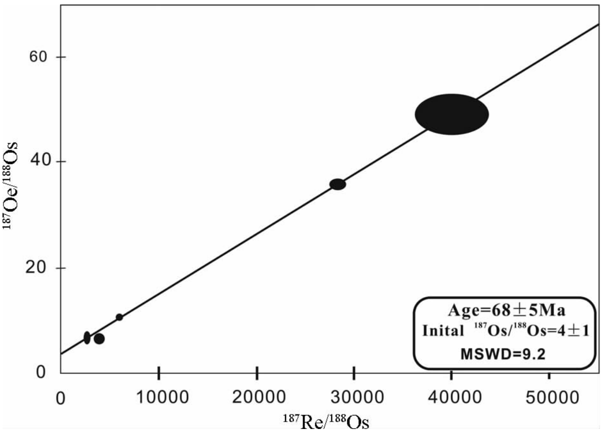

图 3 金顶铅锌矿石中沥青的 Re-Os 等时线

Fig. 3 The Re-Os isochron of the bitumen in the Jinding zinc-lead ores

难溶解的组分被有机物吸附(Bruland,1983)。海水中 Os 在氧化条件下以 $HOsO_5^-$ 形式存在,溶解度大,易于迁移;而在还原环境下,以活动性很弱的低价形式存在,因此在富有机质的还原沉积环境中,高价态的 Os 被还原富集(Peucker-Ehrenbrink and Ravizza,2000;Yoshiro *et al.*,2007)。由于 Re、Os 在富有机质的岩石中富集,并且这些富有机质的岩石常形成于较还原环境,Re-Os 同位素体系能够保持较好封闭性,使 Re-Os 同位素体系近年有效地应用于富有机质地质样品。

油页岩、黑色页岩、炭质泥岩、富有机质灰岩等构成的黑色岩系在沉积的过程中,有机质能吸附、富集海水中的 Re、Os 元素,故其沉积过程就是其中 Re-Os 同位素体系封闭计时的过程;黑色岩系 Re-Os 同位素年龄反映的是黑色岩系的成岩年龄,初始 $^{187}Os/^{188}Os$ 反映的是黑色岩系沉积时海水的 $^{187}Os/^{188}Os$ 值。原油、沥青、油砂等多由黑色岩系在一定条件下经过热降解和热裂解衍生而成,它们的形成往往经过了生烃、运移、圈闭等有机质富集过程;此过程中 Os 同位素达到平衡,Re-Os 同位素体系会重置和重新计时(李超等,2010);相对于黑色岩系,这些有机质样品更富集有机质,Re 和 Os 含量和放射成因同位素较高,Re-Os 同位素年龄记录的是烃源岩生烃后,烃类运移或圈闭的年龄(Creaser *et al.*,2002),初始 $^{187}Os/^{188}Os$ 指示含烃流体的来源(Selby *et al.*,2005)。

油气勘查、有机岩相学和地球化学研究表明,兰坪盆地沉积柱中,上三叠统三合洞组(T_3s)碳质灰岩和麦初箐组(T_3m)碳质泥岩是主要烃源岩(胡明安,1989a;王大锐和张抗,2003;常象春和张金亮,2003;付修根,2004;高永宝等,2008a;薛春纪等,2007a, 2009)。金顶矿区云龙组为陆相含砾砂岩和砂砾岩,虽然其中较多角砾为上三叠统三合洞组(T_3s)碳质灰岩和麦初箐组(T_3m)碳质泥岩,具有一定的生烃能力,但研究认为金顶穹隆中大量油气显示和遗迹说明大部分烃类物质是二次运移到穹隆内富集的(Xue *et al.*,2007c;高永宝等,2008a;薛春纪等,2009)。本次针对金顶矿区古新统云龙组铅锌硫化物矿化含砾砂岩和砂砾岩角砾格架空隙中沥青,Re-Os 法测得等时线年龄 68±5Ma,应代表烃类流体运移至云龙组内富集成藏的时代,它在误差范围内与云龙组地层的沉积成岩时代相一致。兰坪盆地新生代陆相沉积主要与盆地中央南北走向的比江断裂自新生代以来强力活动形成的走滑拉分盆地有关,这个断裂具有较大的切割深度(尹汉辉等,1990;薛春纪等,2002b;Xue *et al.*,2004),为盆地主要烃源岩生烃后向上运移提供了地质条件。兰坪盆地古新统主要沿比江断裂的西侧分布,云龙组含砾砂岩和砂砾岩成岩之后,矿区发生了推覆和穹隆构造,随后烃类流体沿比江断裂从下部运移注入,穹窿构造对油气起到了很好的圈闭保护作用。

本次测年沥青样品所代表的古油气藏与其储层、即古新统云龙组属同时代;金顶铅锌硫化物矿化以云龙组油气储层为主岩,发生显著的热液交代后生成矿作用(白嘉芬等,1985;吴淦国和吴习东,1989;胡明安,1989b;尹汉辉等,1990;薛春纪等,2002b; Chi *et al.*,2007),反映油气成藏先于铅锌成矿。油气藏内有机质对硫酸盐的热化学还原产生的大量硫化氢为铅锌硫化物沉淀奠定了还原硫化学物质基础(Xue *et al.*,2003,2006,2007c;高永宝等,2008b),很可能沿比江断裂从深部注入金顶油气藏的金属离子与硫化氢快速集中反应导致铅锌硫化物大量沉淀而成矿(Xue *et al.*,2006,2007c;薛春纪等,2009),油气成藏与铅锌成矿很可能是先后连续发生的动力学过程。

6 结论

金顶超大型矿床以古新统云龙组含砾砂岩和砂砾岩为主岩铅锌矿石中沥青的 Re-Os 等时线年龄为 68±5Ma,指示金顶古油气成藏时代。

金顶古油气藏形成于古新世,先于铅锌硫化物大规模成矿,烃类物质具有通过热化学还原硫酸盐提供铅锌成矿所需硫化氢的客观条件。

油气成藏与铅锌成矿在云南金顶矿区很可能是一个先后发生的连续地质过程,成藏为成矿奠基,成矿伴随着油气藏的破坏。

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