



邯郸煤田岩浆侵蚀区9号煤中黄铁矿赋存特征及成因机制

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邯郸煤田岩浆侵蚀区9号煤中黄铁矿赋存特征及成因机制

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摘要:以邯郸煤田受岩浆侵入作用明显的云驾岭煤矿9号煤为研究对象,综合运用光学显微镜观察、带能谱的电子探针(EPMA-EDS)、X射线衍射仪(XRD)、X射线荧光分光仪(XRF)等多种分析测试方法,分析了9号煤中全硫、形态硫的含量以及煤中的矿物富集特征,研究了9号煤中全硫及黄铁矿的赋存状态,探讨了9号煤中硫的来源,以及不同形态黄铁矿的成因机制。研究结果表明,邯郸煤田云驾岭煤矿9号煤为高硫煤,硫元素含量在0.61%~7.12%,其中硫化物硫(含量1.18%~4.90%)为煤中全硫的主要形态,有机硫(含量0.56%~2.16%)次之,硫酸盐硫(含量0.01%~0.06%)含量甚微。云驾岭煤矿9号煤沉积于海陆相过渡环境,海水入侵为9号煤层提供了丰富硫源,燕山期岩浆岩侵入体引入的气液物质使云驾岭9号煤中全硫含量升高,特别是上部煤分层的全硫含量较整个煤层明显升高。云驾岭煤矿9号煤中黄铁矿微观赋存形态主要包括块状黄铁矿、浸染状黄铁矿和裂隙充填黄铁矿,具有多阶段演化的特点。块状黄铁矿主要形成于早期成岩阶段,燕山期岩浆岩侵入体带来的高温及其气液物质改造了煤中黄铁矿的形态特征,使煤中黄铁矿发生活化并重新结晶为块状部分。黄铁矿受高温影响部分无机硫扩散至周边煤体中并以有机硫的形式固存,提高了上部煤分层的有机硫含量。

关键词:邯郸煤田;岩浆侵入;硫;黄铁矿;成因机制

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Occurrence and genetic mechanism of pyrite in the No. 9 coal seam in magmatic erosion area of the Handan coalfield

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Abstract: The No. 9 coal of Yunjialing Coal Mine in Handan Coalfield, which is subject to obvious magmatic intrusion, is taken as the research object. Optical microscope observation, electron probe energy spectrometry (EPMA-EDS), X-ray diffractometer (XRD), X-ray fluorescence spectrometry (XRF) and other analytical tests were used to analyze the enrichment characteristics of whole sulfur, sulfur forms and minerals, to study the occurrence of sulfur and pyrite in the coals, and to explore the sources of different types of pyrite. The results show that the No. 9 coal of Yunjialing Coal Mine in Handan Coalfield is a high-sulfur coal (0.61% ~ 7.12%), sulfide sulfur is the main form of sulfur in the coal (1.18% ~ 4.90%), followed by organic sulfur (0.56% ~ 2.16%) with a little sulphate sulfur (0.01% ~ 0.06%). The No. 9 coal of Yunhailing coal mine was deposited in the transitional environment of sea and land phases, and seawater intrusion provided abundant sulfur source for the No. 9 coal seam, the gas-liquid materials brought about by neutral magmatism during the Yanshan period elevate the total sulfur content in the No. 9 coal of the Yunjialing Coal Mine, especially the total sulfur content of the upper coal plies are significantly higher than the total sulfur content of the whole coal seam. The microscopic occurrence of pyrite in the No. 9 coal mainly includes massive pyrite, disseminated pyrite and fissure-filled pyrite, and is characterized by multi-stage evolution. Massive pyrite is mainly

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formed in the early diagenetic stage, the high temperature and gas-liquid materials brought about by neutral magmatism during the Yan-shan period modified the morphology of pyrite in the coal, resulting in the activation and recrystallization of pyrite in the coal into a massive fraction. Inorganic sulfur from the high-temperature-affected portion of the pyrite diffused into the surrounding coal body and sequestered as organic sulfur, increasing the organic sulfur content of the upper coal plies.

Key words: handan coalfield; magmatic intrusion; sulfur; pyrite; genetic mechanism

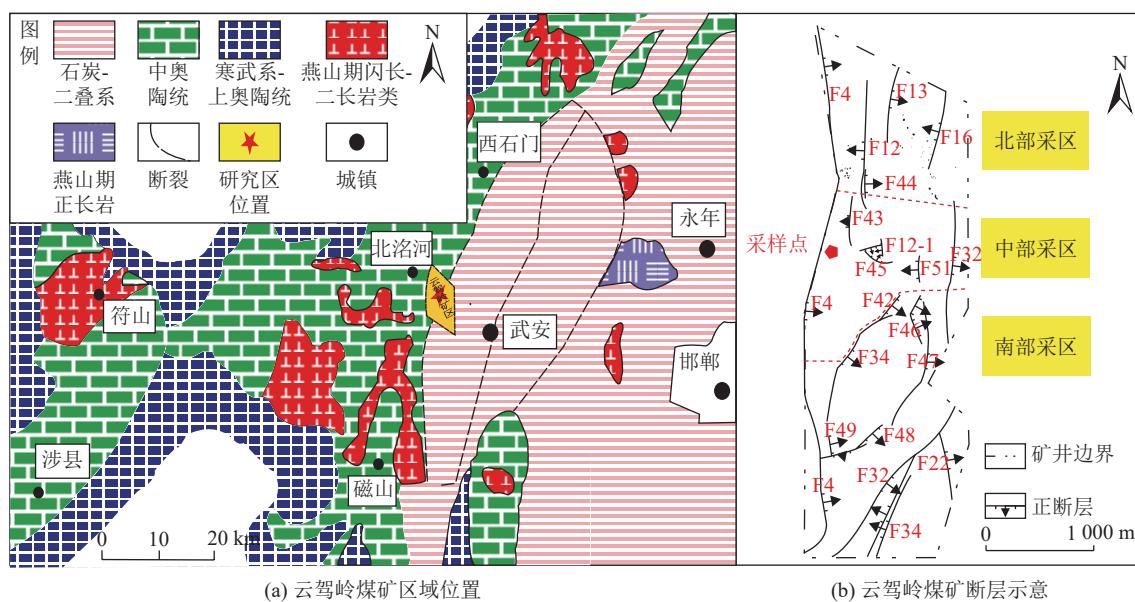
0 引言

黄铁矿是煤中常见的硫化物矿物,与煤中硫的关系密切^[1-5]。煤燃烧产生硫氧化物是造成酸雨的主要原因^[6-8],煤中黄铁矿也会引发诸多环境问题^[9],例如黄铁矿氧化引起的酸性矿山排水问题^[10],对人与环境的危害不言而喻。BERNE^[11]认为煤中黄铁矿的形成包括3个必要条件: SO_4^{2-} 、 Fe^{2+} 和有机质。唐跃刚等^[12]将黄铁矿的演化模式分为2种,即直接沉淀黄铁矿成因演化模式和复杂成因黄铁矿的形成模式。刘大锰等^[13]将华北晚古生代煤中的黄铁矿分为8种类型,即莓球状、自形粒状、鱼子状、块状、均质球形、其他形状、结节状和裂隙充填,并根据黄铁矿的S、Fe含量、S/Fe原子比例及伴生元素等特征,将黄铁矿的成因分为4代:同生-准同生期、早成岩期、晚成岩期和后生阶段。在某些情况下,黄铁矿的形态是重叠的,通常自形体通常是莓球状组合的一部分,后者可以被块状黄铁矿覆盖^[1]。本次研究中运

用原子吸收分光光度计、X射线衍射、偏光显微镜及带能谱的电子探针对云驾岭煤矿采集的9号煤样品进行测试,分析了9号煤中黄铁矿的赋存特征及成因机制,通过对硫化物形态的详细描述,以期更准确地预测各种选煤工艺去除黄铁矿的相对有效性,为邯郸-峰峰矿区煤炭的可选性提供理论支持,提高相关产业的经济与环境效益。

1 地质背景

邯郸煤田位于中国河北省西南部,地质上位于华北克拉通中部。代世峰等^[14]对邯郸煤田的地质背景进行了细致描述。云驾岭煤矿位于邯郸煤田西北部(图1),含煤地层包括石炭系下统本溪组、石炭系上统太原组和二叠系下统山西组。山西组和太原组是研究区主要的含煤地层,由老到新依次为海陆过渡相沉积环境和陆相沉积环境^[15-16],地层柱状如图2所示。



1—石炭-二叠系; 2—中奥陶统; 3—寒武系-上奥陶统; 4—燕山期间长-二长岩类; 5—燕山期正长岩; 6—断裂; 7—研究区位置; 8—城镇

图 1 云驾岭煤矿区域位置及断层示意图(改自张明杰等^[17])

Fig.1 Regional location map and fault diagram of Yunjialing coal mine

燕山期岩浆活动对区内石炭二叠纪煤层产生重要影响,岩浆岩似层状侵入9号煤层上部,9号煤层

局部顶板为侵入岩浆岩,岩浆热液与煤的高温接触使侵入区煤层的变质程度升高,影响了煤中的矿物

组成^[16]。侵入岩浆岩岩石类型为蚀变角闪长玢岩、闪长岩,多表现为碳酸盐化、绿泥石化和绢云母化。区内断层大量发育,以中小型最为发育,几乎均为正断层,以NNE及NE向为主;为深部热液上涌提供了通道。

2 样品采集与测试

样品采自云驾岭煤矿9号煤,共采集18个样品,包括煤样14个,夹矸样2个,顶板与底板样品各1个,夹矸样品、顶板样品和底板样品分别用后缀P、R和F标注。所有采集的样品均用聚乙烯袋包裹,以尽量减少氧化。

将样品进行粉碎筛选,筛选为2组,分别为18~40目(8.8×10^{-4} ~ 3.8×10^{-4} m)和200目(7.4×10^{-5} m)。第一组选用18~40目样品用于制备粉煤光片,使用自动磨抛机研磨和抛光,达到表面光滑无擦痕后,用于偏光显微镜和电子探针鉴定分析;第二组为200目的煤样,用于全硫、形态硫的测定以及XRD测试。

使用偏光显微镜(Leica DM2500P)在反射光下对矿物进行鉴定并拍照,主要在10倍和20倍目镜下进行矿物的鉴定。运用电子探针和能谱仪分析煤中矿物的赋存状态,在背散射模式下进行拍照。

全硫的测定参照国家标准GB 214—1996《煤中全硫的测定方法》,硫酸盐硫、硫化物硫及有机硫的测定参照国家标准GB/T 215—2003《煤中各种形态硫的测定方法》,并利用原子吸收分光光度计测定部分煤中(全硫含量>1%)硫化物硫的含量。

将200目的煤样进行低温灰化后与夹矸和顶底板样品一同进行XRD分析,采用MDI jade5.0和Siroquant软件对样品中矿物组成进行定性与定量分析。

3 煤质特征及煤中主要矿物成分

表1列出了云驾岭煤矿9号煤中全硫、形态硫和 Fe_2O_3 的含量。结果显示9号煤中全硫含量的范围为0.61%~7.12%,平均值为3.27%,根据国家标准GB/T15224.2—2010对煤中硫分分级,云驾岭煤矿9号煤被划定为高硫煤。对煤中全硫含量大于1%的样品进行了形态硫测试。硫化物硫含量范围为0.71%~4.90%,平均值为2.71%,是煤中硫的主要形式;有机硫次之,范围为0.56%~2.16%,平均值为1.07%;硫酸盐硫甚微。全硫在煤层中的纵向含量变化较大,上下部较高,中部靠近夹矸的煤中硫含量较低,沉积旋回韵律明显。煤中全硫和硫化物硫同 Fe_2O_3 (数据由X射线荧光光谱仪测试所得,未发表数据)呈高度正相关(图3),相关系数 R^2 分别为0.93

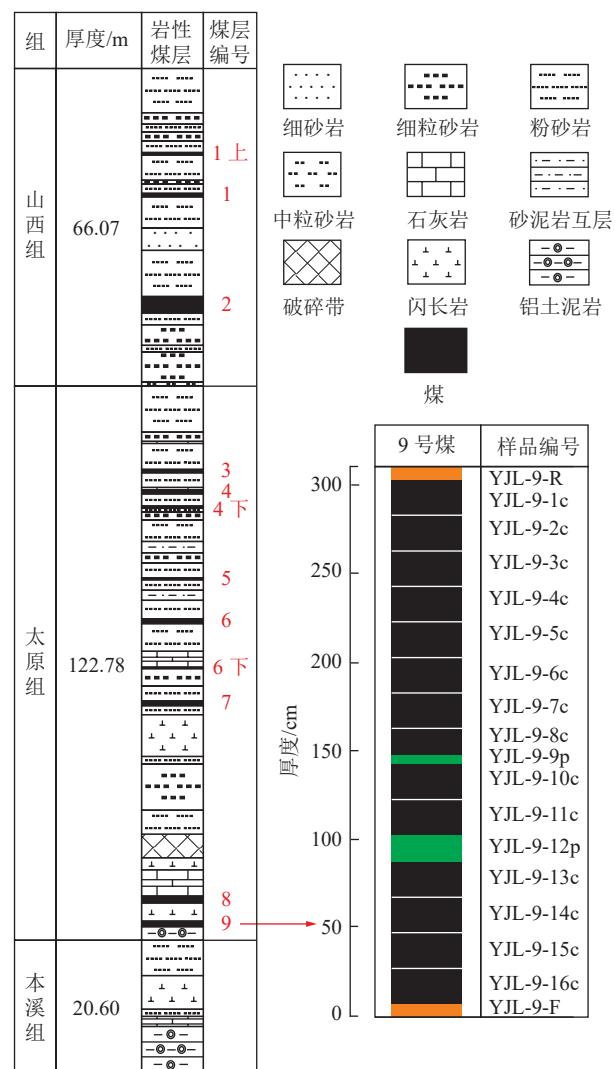


图2 云驾岭煤矿地层柱状
Fig.2 Stratum histogram of the Yunjianling Coal Mine

和0.92,加之相似的垂直变化(图4),表明黄铁矿是煤中硫的主要载体。

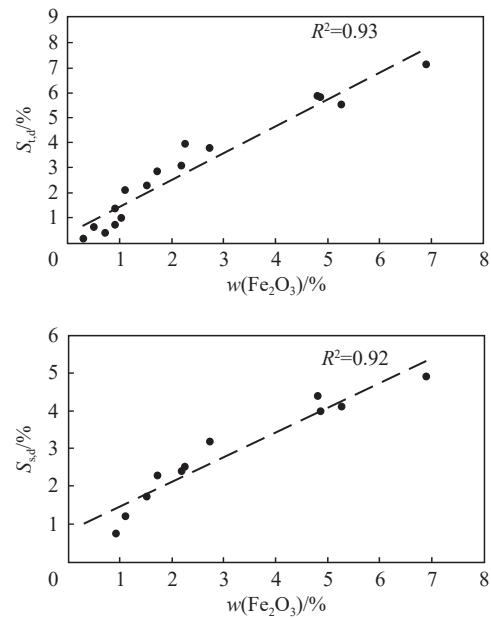
高硫煤通常沉积于受海水影响的环境,原始植物、侵入泥炭沼泽中海水的硫酸盐等控制了煤中硫的含量分布与赋存状态^[18]。除海水外,浅成热液、热液或富含硫酸盐的流体也可能使煤中硫浓度升高^[1,19-21]。

云驾岭煤矿太原组9号煤形成于海岸带环境,海水为煤层提供了大量硫酸根离子。燕山期岩浆岩侵入引入的局部热提高了煤的成熟度,使区域内含煤地层发生了不同程度的变质,同时,岩浆岩侵入体可能引入大量热液使云驾岭9号煤中的全硫含量升高,特别是上部煤层的全硫含量较整个煤层明显升高(图4)。煤中黄铁矿受高温影响部分无机硫扩散至周边煤体中并以有机硫的形式固存^[22-23],提高了上部煤层的有机硫含量。

表2列出煤中主要矿物成分,包括:伊利石、方

表 1 9 号煤全硫、形态硫(部分样品)及 Fe_2O_3 含量Table 1 Total sulfur, morphological sulfur (Partial samples) and Fe_2O_3 content of No. 9 coal

样品编号	含量/%				
	全硫	硫化物硫	硫酸盐硫	有机硫	Fe_2O_3
YJL-9-R	2.57	2.55	0.06	—	6.36
YJL-9-1c	5.50	4.10	0.01	1.38	5.28
YJL-9-2c	7.12	4.90	0.06	2.16	6.91
YJL-9-3c	5.84	4.39	0.05	1.40	4.27
YJL-9-4c	5.80	3.96	0.03	1.82	4.87
YJL-9-5c	3.76	3.16	0.03	0.57	2.74
YJL-9-6c	2.25	1.70	0.02	0.53	1.53
YJL-9-7c	1.36	0.71	—	0.65	0.92
YJL-9-8c	0.70	—	—	—	0.92
YJL-9-9p	0.36	—	—	—	0.73
YJL-9-10c	0.61	—	—	—	0.52
YJL-9-11c	3.95	2.50	0.04	1.41	2.26
YJL-9-12p	0.12	—	—	—	0.31
YJL-9-13c	0.95	—	—	—	1.04
YJL-9-14c	3.06	2.37	—	0.69	2.19
YJL-9-15c	2.82	2.25	0.01	0.56	1.74
YJL-9-16c	2.08	1.18	—	0.90	1.11
YJL-9-F	0.41	—	—	—	0.22

图 3 云驾岭煤矿 9 号煤全硫、硫化物硫与 Fe_2O_3 关系Fig.3 Relationship between total sulfur, sulfide sulfur and Fe_2O_3 of No. 9 coal in Yunjialing Coal Mine

解石、钠长石、黄铁矿以及高岭石; 在部分煤样中还鉴定出铁白云石、白云母和菱铁矿, 但含量均较低。黄铁矿在煤层中普遍发育, 纵向含量分布与煤层中全硫的含量分布相似(图 4)。

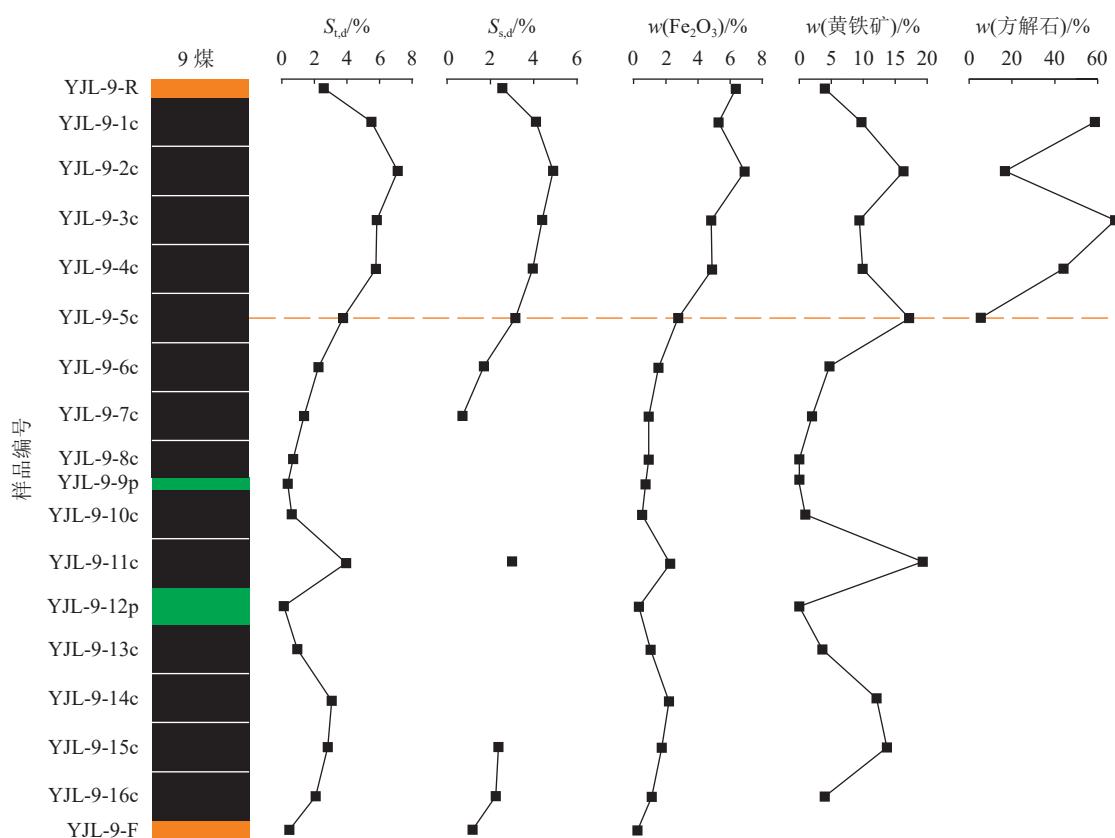
图 4 云驾岭煤矿 9 号煤中全硫、硫化物硫、 Fe_2O_3 、黄铁矿含量以及方解石含量的剖面变化Fig.4 longitudinal distribution of total sulfur, pyrite sulfur, Fe_2O_3 , pyrite and calcite in No. 9 coal of Yunjialing Coal Mine

表2 云驾岭煤矿9号煤中主要矿物成分含量

Table 2 Primary mineral composition of No. 9 coal in the Yunjialing Coal Mine

样品编号	质量分数/%				
	伊利石	方解石	钠长石	黄铁矿	高岭石
YJL-9-R	49.7	—	14.9	4.0	—
YJL-9-1c	4.5	58.7	21.2	9.7	—
YJL-9-2c	44.5	16.7	19.7	16.3	2.0
YJL-9-3c	2.5	68.1	20.1	9.4	—
YJL-9-4c	—	44	46.1	9.9	—
YJL-9-5c	31.1	5.4	42.7	17.2	3.5
YJL-9-6c	62.2	—	21.8	4.7	10.7
YJL-9-7c	45.8	—	11.2	2.0	—
YJL-9-8c	72.8	—	16.7	—	—
YJL-9-9p	63.3	—	30.3	—	—
YJL-9-10c	63.6	—	18.9	1.0	10.9
YJL-9-11c	49.1	—	16.9	19.3	7.0
YJL-9-12p	75.6	—	13.5	—	—
YJL-9-13c	39.9	—	45.4	3.6	5.2
YJL-9-14c	28.2	—	47.6	12.1	4.0
YJL-9-15c	28.4	—	49.9	13.7	4.1
YJL-9-16c	32.7	—	63.2	4.0	—
YJL-9-F	41.7	—	6	—	—

4 煤中黄铁矿显微赋存特征

运用偏光显微镜及带能谱的电子探针对云驾岭煤矿9号煤中黄铁矿进行了观察与分析,煤中黄铁矿微观赋存形态包括块状黄铁矿、浸染状黄铁矿和裂隙充填黄铁矿。其中块状黄铁矿普遍发育,块状黄铁矿形状普遍不规则,粒度变化大(2~1 000 μm),表面光洁或发育孔洞(图5a、图5b),煤层上部黄铁矿表面孔洞较为发育,内部充填矿物或有机质,而下部煤层的黄铁矿表面孔洞明显减少,较为光洁(图6)。浸染状黄铁矿和裂隙充填黄铁矿较少。

5 黄铁矿成因分析

黄铁矿的形成是一个复杂的多阶段过程,硫酸盐、有机质和Fe离子的供应以及有利于细菌活动的厌氧环境是黄铁矿形成的必要条件。不同期次和环境下生成的黄铁矿的形貌、微结构及地球化学特征也都不尽相同^[12-13]。如霉球状、自形晶体和块状黄铁矿通常是在泥炭堆积或早期成岩过程中受海水影响形成的^[24]。后生阶段形成的黄铁矿常以割理状和裂隙填充状的形式出现在煤层中^[4,25]。

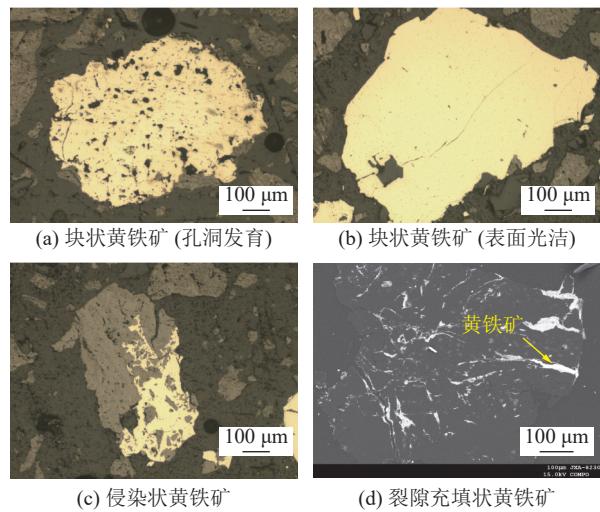


图5 云驾岭煤矿9号煤中黄铁矿微观赋存特征
Fig.5 Microscopic occurrence characteristics of pyrite in No. 9 coal of the Yunjialing Coal Mine

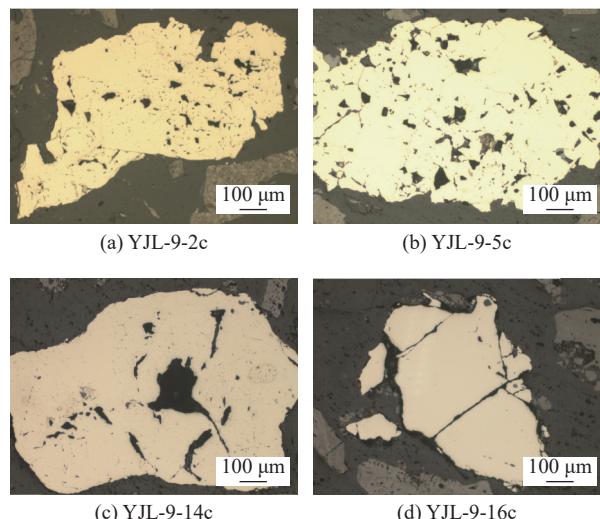


图6 显微镜下煤中不同表面特征的块状黄铁矿
Fig.6 Massive pyrite with different surface characteristics in coal under microscope

邯郸煤田太原组含煤地层沉积于海陆相过渡环境,为煤中黄铁矿的形成提供了适宜的环境。受海水影响的泥炭环境表现出弱还原性,有利于细菌繁殖和活动。来自海水的硫酸盐被细菌代谢,生成H₂S、多硫化物和元素硫,与Fe离子反应形成同生黄铁矿^[1,4,26]。云驾岭煤矿9号煤中普遍发育块状黄铁矿,块状黄铁矿主要形成于早期成岩阶段,在反应时间与物质供应都充足的前提下,随着黄铁矿生成环境介质浓度的增加,黄铁矿数量增加并聚集,最后向连生体及完全连生体方向演化,形成块状^[9-10,24,27]。

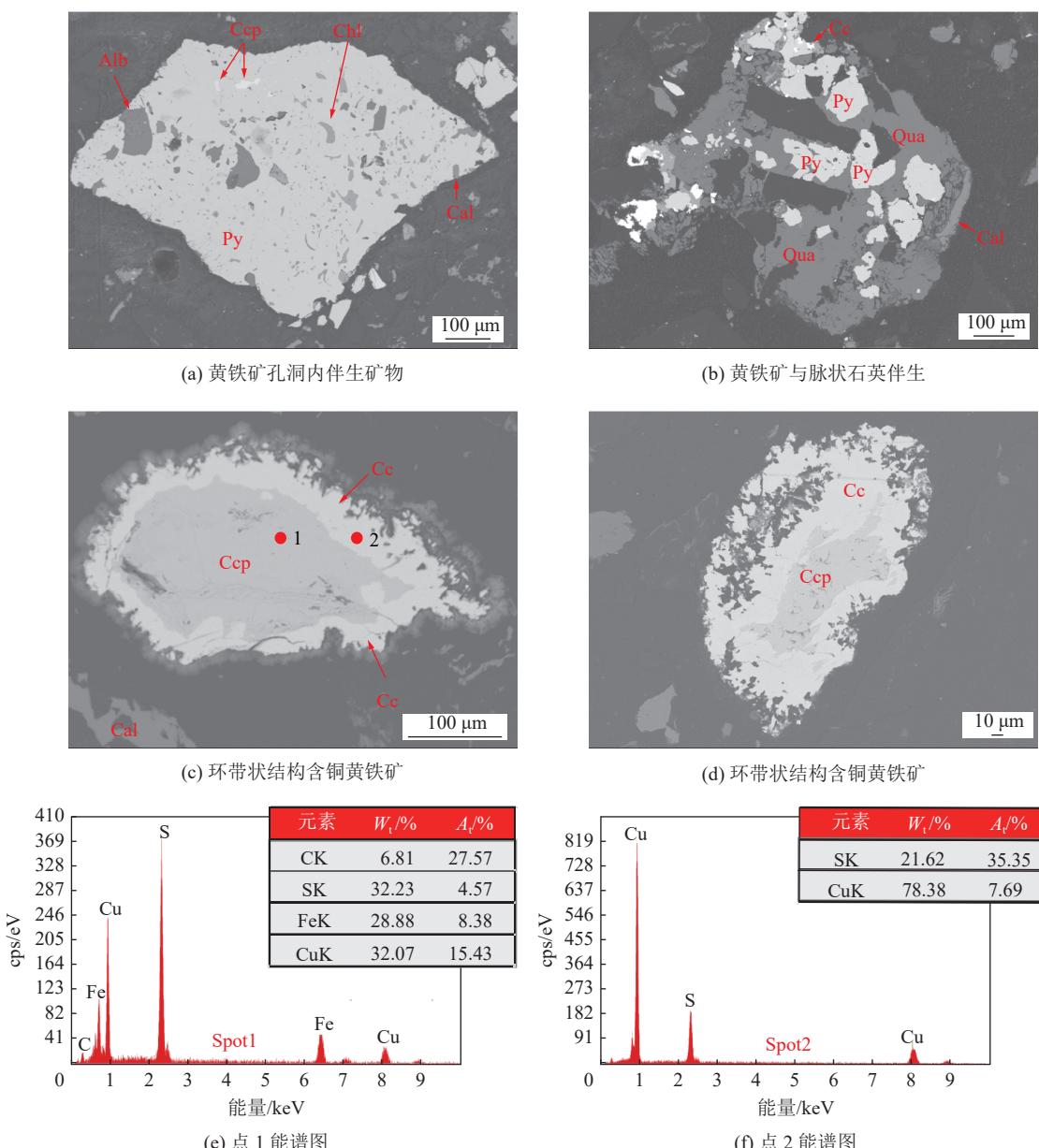
燕山期间岩浆岩似层状侵入到9号煤上部,岩浆岩侵入体引入的热液影响了煤层上部黄铁矿的形成与赋存,导致云驾岭9号煤层上部的黄铁矿表面

孔洞发育, 而下部则较为光洁, 孔洞明显减少(图 6); 热液向下部煤层淋滤过程中, 所携带的 Fe 离子与 S 元素结合形成黄铁矿。岩浆侵入带来的局部热可能使煤中黄铁矿活化并重新结晶为块状^[28-29], 因此在云驾岭 9 号煤中未发现莓球状和自形晶状等同生-准同生阶段形成的黄铁矿。而在同时期沉积的葛泉矿 9 号煤中则普遍发育莓球状和自形晶状黄铁矿^[30]。

黄铁矿的伴生矿物也可以为黄铁矿的形成提供关键信息。在 YJL-9-1c 样品中发现黄铁矿表面孔洞赋存多种后生矿物, 包括方解石、鲕绿泥石、钠长石和黄铜矿(图 7a)。根据 XRD 检测数据得出, 方解石只在煤层上部存在, 呈条带状或充填在黄铁矿表面

孔隙内, 表现为后生成因^[31]。9 号煤上部的侵入岩成分主要为蚀变的角闪长玢岩和闪长岩, 闪长玢岩和角闪石蚀变产生的 Ca、Fe、Mg 离子为方解石和黄铁矿的形成提供了物质来源^[32], 部分 Fe 离子和 Mg 离子结合形成鲕绿泥石充填在黄铁矿孔洞内^[33]; 煤层下部没有检测出方解石可能是热液活动影响逐渐减弱, 活化元素逐渐沉淀的结果^[34-35]; 煤中钠长石受热液蚀变影响析出 Na 的离子随热液迁移至黄铁矿孔洞内再结晶^[36]; 热液中运输的 Cu 离子与黄铁矿发生交代作用, 在黄铁矿表面形成斑点状黄铜矿, 个别为辉铜矿(图 7a、图 7b)。

煤中自生石英可能是由于煤中高岭石与镁铁质



Py—黄铁矿; Qua—石英; Chl—绿泥石; Alb—钠长石; Cal—方解石; Ccp—黄铜矿; Cc—辉铜矿

图 7 煤中黄铁矿伴生矿物及环带结构含铜黄铁矿(电子探针背散射成分像)

Fig.7 associated minerals of pyrite and zonal copper bearing pyrite in coal (electron probe back-scattering component image)

热液作用,高岭石脱硅后形成的,或是由低温热液沉淀形成^[37-38]。云驾岭9号煤中YJL-9-4c样品在电子探针下发现自生脉状石英包裹块状黄铁矿(图7b),自生脉状石英可能是由煤层上部岩浆活动带来的镁铁质热液与煤中高岭石作用,高岭石脱硅后形成的。流体中携带着Fe离子与环境中的H₂S反应形成黄铁矿。

此外,在煤中还发现了环带结构含铜黄铁矿(图7c,图7d),边缘呈破碎、港湾状,能谱分析内部核心为黄铜矿,外部环带矿物铜含量明显高于内部的铜含量,未发现Fe离子,鉴别为辉铜矿。煤中黄铁矿可能有多个矿化阶段的叠加,热液中的铜离子促使黄铁矿生长环带结构发育^[39],边缘呈破碎、港湾状表明形成过程中的物理化学条件具有明显的波动性。

6 结 论

1)云驾岭煤矿9号煤中黄铁矿微观赋存形态包括块状黄铁矿、浸染状黄铁矿和裂隙充填黄铁矿。块状黄铁矿主要形成于早期成岩阶段,多呈不规则状,粒度变化大(2~1 000 μm),表面光洁或发育孔洞。浸染状黄铁矿和裂隙充填黄铁矿表明后生成因,只在煤层上部发现。

2)燕山期岩浆岩侵入注入的热液导致云驾岭9号煤层上部的黄铁矿表面孔洞发育,内部充填矿物或有机质,而下部则较为光洁,孔洞明显减少;热液中所携带的Fe离子与S元素结合形成黄铁矿。岩浆侵入带来的局部热使煤中黄铁矿发生活化并重新结晶为块状。

3)云驾岭煤矿9号煤上部侵入岩蚀变产生的Ca、Fe离子为方解石和黄铁矿的形成提供了物质来源。煤中高岭石与镁铁质热液作用脱硅后形成自生脉状石英;流体中携带着Fe离子与环境中的H₂S反应形成了黄铁矿。环带状结构发育的含铜黄铁矿也反映热液成因。

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