矿山环境保护

关闭/废弃煤矿甲烷排放研究现状及减排对策

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摘　要: 全球正向低碳能源结构转型，加之煤矿资源枯竭、瓦斯灾害等问题，关闭/废弃煤矿数量快速增加。煤矿关闭退出后，采空区残存甲烷持续向地面逸散，成为温室气体重要排放源。针对关闭/废弃煤矿甲烷残存量、排放速率、减排措施等系列问题，通过大量文献调研、梳理，明确了国内外关闭/废弃煤矿数量及高瓦斯矿井分布，归纳总结了采空区残存甲烷来源及残存量的估算方法，并借鉴天然气成藏研究领域中甲烷地质渗漏理论与研究方法，分析了残存甲烷排放机制及监测手段，最后提出了残存甲烷减排对策及面临挑战。研究发现:我国山西、贵州、重庆、湖南、江西等地区存在大量关闭/废弃煤矿，且残余煤主要是具有强甲烷吸附力的无烟煤，导致矿井残存甲烷量大，成为重要甲烷排放源；关闭/废弃煤矿甲烷从残余煤中解吸释放至采空区，然后经由井口、采动裂隙等通道排放至大气；通过卫星遥感、通量室法、地球化学探针法、微气象技术等手段，可实现对煤矿甲烷排放的有效监测。基于甲烷排放预测模型预测，到 2050 年关闭/废弃煤矿排放甲烷在煤炭开采释放甲烷总量中占比可能超过 20%，解决关闭/废弃矿井甲烷排放问题刻不容缓。为此，提出了抽采利用、原位爆燃发电、微生物降解甲烷、注水淹没、甲烷排放通道封堵等减排对策，综合考虑减排成本、甲烷排放持续时间、地下水污染等限制性因素，认为采用矿化修复方法封堵覆岩采动微裂隙，可低成本实现关闭/废弃煤矿甲烷减排目标。

关键词: 甲烷减排；关闭/废弃煤矿；采动裂隙；甲烷地质渗漏；矿化修复；温室效应

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Research status and reduction strategies of methane emissions from closed/abandoned coal mines

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Abstract: With the global transition to a low-carbon energy structure, coupled with the depletion of coal mine resources, gas disasters, and other problems, the number of closed/abandoned coal mines is increasing rapidly. After the coal mine closes, the residual methane in the goaf escapes to the ground continuously, becoming an essential source of greenhouse gas emissions. In view of a series of issues such as methane residual stock, emission rate and emission reduction measures in closed/abandoned coal mines, the number of closed/abandoned coal mines and the distribution of high-gas mines at

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home and abroad were clarified through a large number of literature review, and the sources and estimation methods of methane residual stock were summarized. At the same time, the theory and research method of methane geological leakage in the field of natural gas accumulation was used for reference, and the emission mechanism and monitoring means of residual methane were analyzed. Finally, the countermeasures and challenges of residual methane emission reduction were put forward. The study found that there are many closed/abandoned coal mines in Shanxi, Guizhou, Chongqing, Hunan, Jiangxi, etc., and the residual coal is mainly anthracite with strong methane adsorption capacity, resulting in a large amount of residual methane in the mine, which will become a critical methane emission source. Methane from closed/abandoned coal mines is desorbed and released to the goaf, and then discharged to the atmosphere through channels such as wellhead and mining-induced fractures. Methane monitoring in coal mines can be realized by means of satellite remote sensing, flux chamber method, geochemical probe method, micro-meteorological technology, etc. Based on the methane emission prediction model of closed/abandoned coal mines, the methane emissions from closed/abandoned coal mines may account for more than 20% of total methane emissions from coal mining operations by 2050, so it is urgent to solve the problem of methane emissions from closed/abandoned mines. Therefore, the countermeasures of emission reduction, such as extraction and utilization, in-situ deflagration power generation, microbial degradation of methane, water flooding, and methane emission channel closure are put forward. Considering the limitations of cost, treatment time, groundwater contamination, and other limitation factors, it is concluded the mineralized remediation method can be used to seal large-scale mining-induced fractures in overlying rocks, which can achieve the methane emission reduction goal of closed/abandoned coal mines at a low cost.

Key words: methane emission reduction; closed/abandoned coal mines; mining-induced fracture; methane geological leakage; mineralized remediation; the greenhouse effect

在碳中和目标与国家新能源战略背景下,我国能源结构性改革不断推进,加之煤矿自身服务年限期满,关闭/废弃煤矿数量快速增加。关闭/废弃煤矿是指由于煤炭资源枯竭、不符合安全开采条件或政策等原因而被关闭和废弃的煤矿[1-3],我国关闭/废弃煤矿数量不断增加,且多为高瓦斯和突出矿井,受到开采技术、地质构造情况复杂性等因素制约,井下遗留大量含瓦斯煤炭[4-5],在温度差、压力差和地下水等因素影响下[6-10],关闭/废弃煤矿采空区游离态甲烷极易通过井口、覆岩采动裂隙等通道逸散至地表大气。

甲烷是一种对气候影响仅次于二氧化碳的第二大长寿命温室气体,政府间气候变化专门委员会 (IPCC) 指出,100 a 时间范围内,甲烷升温潜势是二氧化碳的 28~36 倍,约占全球温室气体排放的 20%,对全球环境及气候变化有重大影响[11-14]。化石燃料开采是甲烷排放的重要来源,其中,煤矿甲烷排放量约占全球人为甲烷排放的 11%[15-16]。联合国气候变化框架公约数据显示,2010 年煤矿关闭/废弃后甲烷排放量占总甲烷排放量的 17%。SHENG 等[17]通过甲烷排放卫星观测数据及区域贝叶斯逆向分析评估了中国甲烷排放趋势,认为关闭/废弃煤矿甲烷的持续无组织排放将导致中国甲烷排放量不断上升。KHALIL 等[18]发现中国人为甲烷排放量不断增加,且煤炭及天然气排放甲烷占比逐年增加。

关闭/废弃煤矿井中甲烷的持续无组织逸散,对气候具有长期、潜在的影响。KHOLOD 等[19]基于关闭/废弃煤矿甲烷排放预测模型估算发现,2050 年关闭/废弃煤矿甲烷在煤炭开采甲烷排放中占比可能增加到 23%,并于 2100 年增加至 27%。美国于 2002—2018 年间共关闭地下煤矿 747 座,与此同时,其关闭煤矿甲烷排放量在煤矿甲烷总排放量中的占比增加了 5.5%。截至 2021 年底,中国累计关闭或废弃煤矿约 5 700 处,重庆地区所有煤矿已全部关闭,四川、贵州等地区大部分煤矿也逐渐关闭。由此可见,随着废弃煤矿数量持续增加,关闭/废弃煤矿将成为重要的甲烷排放源,为实现煤炭行业低碳、绿色、持续发展,解决关闭/废弃煤矿甲烷排放问题刻不容缓。

准确获取关闭/废弃煤矿甲烷资源量数据、正确把握井下甲烷排放机制及监测方法、革新技术手段利用井下甲烷或封堵其排放路径是关闭/废弃煤矿甲烷减排的重要理论基础。基于此,笔者围绕上述 3 个方面内容研究进展开展综述,为煤炭行业甲烷减排提供理论依据和方向。

1 全球煤矿关闭/废弃现状

由于煤炭资源的枯竭及全球"碳中和"计划不断推进,国内外煤矿都在相继减产或关闭,关闭/废弃煤矿数量正在迅速增加。而煤矿甲烷量对甲烷排放量

有重要影响。由图 1 可知, 2019 年中国甲烷排放量的大部分来自化石燃料、畜牧业、农业。其中, 采煤排放甲烷约占 38.3%, 是中国最大的人为甲烷排放源。随着中国地下矿的逐渐关闭, 废弃矿井甲烷排放占比由 2010 年 2% 增至 2019 年 15%, 达 4.7 Mt/a。正确把握全球煤矿关闭/废弃现状并对关闭趋势合理预测对关闭/废弃煤矿甲烷减排有深远意义。
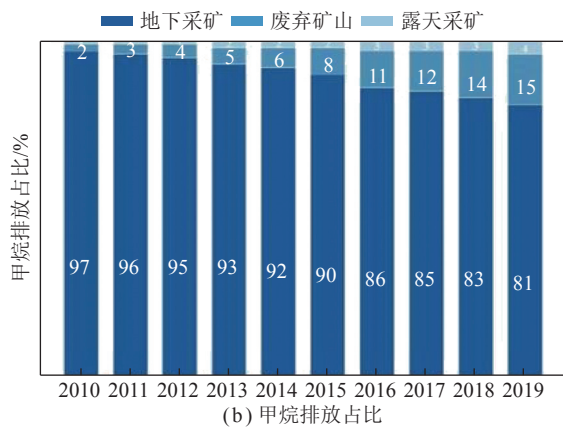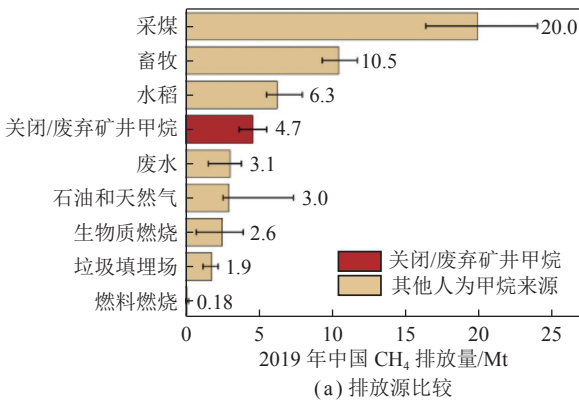

图 1　2019 年中国人为甲烷排放源的比较[20] 和中国露天矿、地下矿与关闭/废弃矿井甲烷排放占比[21]

Fig.1　Comparison of anthropogenic methane emission sources in China in 2019[20] and Proportion of methane emission from open-pit mines, underground mines, and closed/abandoned mines in China[21]

数据显示, 美国煤矿数量从 2008 年的 1 435 座减少至 2020 年的 552 座, 其中地下煤矿占比 60%, 废弃煤矿共计 22 000 余处[22-24]。加拿大煤炭行业发展历史悠久, 估计有 10 000 余处废弃煤矿[25]。德国已于 2018 年 12 月关闭了所有硬煤矿, 1981 年英国煤炭委员会控制的 211 座煤矿, 目前也已全部关闭。

随着我国经济发展方式的转变和"碳达峰、碳中和"目标的持续推进, 我国关闭煤矿数量快速增加[26]。王家臣等[23] 调查显示, 中国煤矿数量由 2000 年的 30 000 余处减至 2019 年的 5 300 处。截至 2021 年底,

我国煤矿数量减少到 4 500 处以下[27]。根据中国工程院重点咨询项目"我国煤炭资源高效回收及节能战略研究"预测, 2030 年我国关闭/废弃煤矿数量将达到 15 000 处[1, 28]。我国煤矿关闭是大势所趋, 未来关闭/废弃煤矿数量将更多。

中国大陆煤炭资源分布较广且储量丰富, 但分布格局不平衡, 高瓦斯矿井数量多且相对集中分布在我国中南和西南地区。根据 2012 年瓦斯等级鉴定结果, 在全国 12 281 处煤矿井中, 高瓦斯和突出矿井达 3 284 处, 约占 26.7%, 其中贵州、四川、湖南、山西、云南、江西、重庆、河南高瓦斯和突出矿井数量较多, 占全国高瓦斯和突出矿井总数的 87.2%, 然而目前这些矿井已全部关闭 (如重庆) 或面临大规模关闭 (如四川、贵州), 且这些地区井下残余煤主要是吸附能力强的无烟煤[21], 随着时间推移煤中解吸出的甲烷量不容小觑。

2　关闭/废弃煤矿甲烷来源与残存量

2.1　残存甲烷来源

我国煤炭开采大多为地下开采, 煤炭采出率较低, 因此关闭/废弃煤矿中遗留大量煤炭[29], 且残存甲烷量巨大, 而煤矿关闭后, 甲烷会持续向地表逸散, 带来环境、安全问题[30-31]。采动作用影响下, 煤岩体中产生采动裂隙场, 应力释放并不断降低, 当煤层压力处于煤层甲烷的临界解吸压力以下时, 煤中甲烷将从吸附态转化为游离态。吸附态的甲烷主要分布在未开采煤层和煤柱中, 游离态甲烷主要赋存于关闭/废弃矿井采空区垮落带及断裂带中, 还有一部分甲烷以溶解态存在于矿井水中[32]。关闭/废弃煤矿采空区中的甲烷主要来源于以下 3 个位置: 采空区遗煤、煤柱、采空区"三带"采动影响范围内邻近未采煤层和围岩, 如图 2 所示。
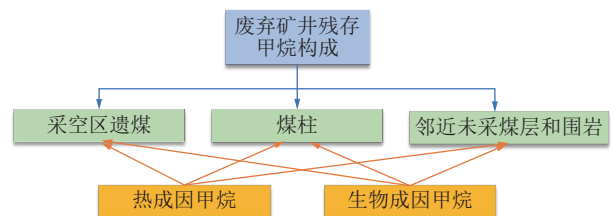

图 2　关闭/废弃矿井残存甲烷构成

Fig.2　Composition of residual methane in closed/abandoned mines

由于热成因、生物成因作用, 煤体会不断产生甲烷并通过裂隙通道释放至大气[33-36], 且废弃矿井环境

更有利于生物成因甲烷生成。BECKMANN 等[37-38]研究发现,煤矿关闭数年后井下甲烷主要是生物成因作用产生,其原因主要在于: 关闭/废弃时间较长的煤矿的低氧气浓度条件,为产甲烷菌等厌氧微生物提供了适宜的生存环境,产甲烷菌的存在和活动致使甲烷不断释放出来。

2.2　甲烷残存量估算

目前国内外对于关闭/废弃煤矿甲烷残存量的评价主要采取月下降曲线法、物质平衡法和物质构成法 3 种方法[39],其主要计算公式及优缺点见表 1。

表 1　关闭/废弃矿井甲烷残存资源量主要估算方法及评价

Table 1　Main estimation methods and evaluation of methane residual resources in closed/abandoned mines

估算方法	计算公式	优点	缺点
月下降曲线法[40]	$q = q_i s (1 + b D_i t)^{(-1/b)}$	不需要大量关闭/废弃矿井资料,计算简便	获取参数需对矿井整体逸散量进行监测
物质平衡法[41]	$Q = Q_z - Q_1 - Q_2$	原理简单,使用经验数据准确性更高	参数难收集,需要收集的资料和数据多,计算工作量大
物质构成法[42]	$Q = Q_y + Q_x$	人为干扰少,准确性高	参数多,需要大量煤矿基础数据资料

注:q 为 t 时的气体流量,m^3/d;q_i 为 t_0 时刻的初始气体流量,m^3/d;s 为密封矿井占比,%;t 为从 t_0 开始经过的时间,a;D_i 为初始下降率,a^{-1};Q 为现在矿井甲烷残存总量,m^3;Q_z 为原始条件下的甲烷残存总量,m^3;Q_1 为矿井抽排的甲烷量,m^3;Q_2 为矿井废弃后逸散的甲烷量,m^3;Q_y 为采空区内的游离甲烷残存量,m^3;Q_x 为废弃煤炭中的吸附甲烷量,m^3。

月下降曲线基于对大量关闭/废弃矿井的甲烷逸散数据监测,构建甲烷逸散经验曲线模型,对该曲线按时间积分到无穷大 (此时甲烷排放量已趋近于 0),得到的总排放量数据即关闭/废弃矿井的全部甲烷残存量[43]。物质平衡法基于物质守恒定律,在煤矿开采前评估的煤层气总量基础上,减去由于煤矿开采等原因损失的甲烷量,得到关闭/废弃矿井内的残存甲烷量。物质构成法考虑到孔隙度、孔隙体积等差异,根据甲烷赋存方式和赋存位置的不同,对采空区、遗煤、邻近层等位置的游离态及吸附态甲烷分别建立数学模型,计算出甲烷残存量。

KUNZ 等[44]通过分析关闭/废弃煤矿甲烷排放数据,发现煤矿关闭后的前 2 个月中甲烷处于快速排放阶段,然后保持平稳排放。DUDA 等[45]基于甲烷排放模型[46],估算 2017 年波兰 Anna 煤矿 713/1-2 煤层甲烷排放量达 4.78×10^6 m^3,且该煤层甲烷排放将于 2023 年停止。KARACAN 等[47]以美国宾夕法尼亚州关闭煤矿为研究对象,采用物质平衡法评价关闭/废弃煤矿甲烷残存量,依据煤矿内 278 个甲烷勘探钻孔实测数据,推导得出该煤矿关闭 700 d 将排放甲烷 3.68×10^6 m^3。PALCHIK[48]采用递减曲线分析法,通过垂直钻孔监测甲烷排放,研究了乌克兰 3 个关闭/废弃煤矿甲烷渗漏特征,优化了月下降曲线法中的参数,估算得到 Centralnaya 2 号矿井在 60 d 内甲烷排放超过 1.4×10^3 m^3。

国内学者通过不同评估方法构建关闭/废弃煤矿甲烷资源模型,估算关闭/废弃煤矿内甲烷残存量。李日富[29]通过采动稳定区孔隙体积、遗煤总量、围岩煤炭量等参数建立采动区甲烷残存量评估模型,以重庆石壕煤矿为研究对象评估其甲烷残存量约 7.86×10^6 m^3。韩保山等[40]通过煤心解吸测试数据的数学分析,得到解吸速度–时间曲线变化模型,近似代替甲烷涌出速度下降曲线,再对曲线积分计算得到关闭/废弃矿井甲烷残存量。李袭明等[49]、孟召平等[50]采用物质构成法,根据不同的赋存方式和赋存位置分别建立数学模型,构建了关闭/废弃矿井甲烷赋存量预测模型,将关闭/废弃煤矿甲烷分为游离态、吸附态,分别估算其甲烷残存量,累加得到残存总量。张江华等[51]建立了甲烷残存量及相关参数估算模型,估算山西晋城 4 座煤矿 9 号及 15 号煤层采空区下伏甲烷残存量,研究发现残存甲烷量大且游离气总量占比超过五成,抽采利用经济效益显著。

3　关闭/废弃煤矿甲烷排放机制及监测方法

21 世纪以来,甲烷地质渗漏理论在油气领域已取得较为显著的进展,相对于油气井,关闭/废弃煤矿甲烷渗漏量被认为较少且渗漏数据缺失[52-57],未受到重视。但据国内学者统计,中国现有关闭/废弃的矿井中残煤量高,残存甲烷总量大[1, 58]。因此,正确把握关闭/废弃煤矿甲烷排放速率等机制,采取有效监测手段提高地表甲烷渗漏检测效果,有利于后续减排技术的开展,为关闭/废弃煤矿甲烷减排奠定理论基础。

3.1　甲烷排放运移机制

甲烷在浓度差、压力差作用下,通过采矿竖井井口、采动裂隙等通道由采空区扩散至地表。EICKER[59]研究发现,将关闭/废弃煤矿连通采空区与大气的井口进行封堵能显著降低甲烷排放量。PALCHIK[48]揭示了甲烷从采空区到地表的排放路径主要有贯穿到地

表的垂直裂隙和钻孔。THIELEMANN 等[60]研究发现 2 处位于断层带顶部的甲烷监测点甲烷排放量远高于其他监测点, 揭示了覆岩裂隙对气体运移的作用。ETIOPE[61]证实了甲烷的地质排放是重要的温室气体来源, 大量的甲烷通过断层和覆岩裂隙从地壳自然释放到大气中。PALCHIK[62]通过原位研究地下裂隙的甲烷释放, 发现裂隙是甲烷由采空区上覆岩层释放到大气的重要通道。SECHMAN[10, 63-64]在波兰西里西亚盆地的关闭/废弃矿区采集土壤气体样本进行分析, 研究结果表明, 异常的甲烷浓度主要与被大量断层和裂隙切割的煤层及其露头有关, 断层和裂隙提供了甲烷流出的主要通道。

　　绝大多数地下煤矿都会释放甲烷, 尤其是长壁开采煤矿, 采动会对煤层上下区域产生很大程度的影响[65]。煤矿关闭后, 采动作用导致矿井内甲烷分压降低、解吸并由煤岩层沿采动裂隙释放至采空区, 并在气体密度的影响下浮升并聚积在采空区顶部。甲烷在压力差、浓度差的驱使下不断向上运移, 最终在裂隙带顶部形成高浓度富气区 (图 3), 而采动作用会导致上覆岩体发育产生竖向破断裂隙及离层裂隙[66-69]。随着工作面推进, 采动裂隙不断向上发展, 最终引起地表的移动变形, 产生地表裂隙, 形成连通采空区与大气的裂隙网络, 关闭/废弃煤矿采空区甲烷通过裂隙网络运移、流动, 渗入地表及大气[70-73]。
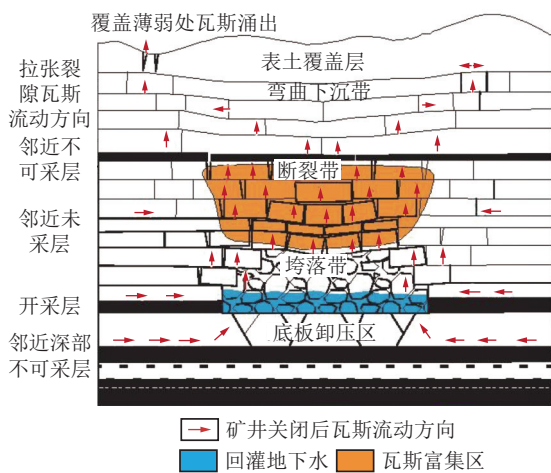

图 3　废弃/关闭煤矿覆岩采动裂隙及残存甲烷排放路径[69]

Fig.3　Abandoned/closed coal mine mining-induced fractures and residual methane emission path[69]

3.2　甲烷排放速率及影响因素

3.2.1　甲烷排放速率

　　根据月下降速度−时间方程[41], KHOLOD 等[19]提出了一种计算关闭/废弃煤矿甲烷排放速率的新方法, 利用煤炭废弃率、煤矿目前状态 (是否被水淹), 将

煤矿分为干煤矿及水淹煤矿, 分别计算其排放速率, 建立煤矿甲烷计算模型。通过以下公式可以估算任意年份的关闭/废弃煤矿甲烷排放率:

$$q = q_i s(1 + bD_i t)^{-1/b}（干煤矿）\tag{1}$$

$$q = q_i e^{-tD_i}（水淹煤矿）\tag{2}$$

　　系数 b 和 D_i 可用现场监测排放数据拟合得到, 煤矿废弃时甲烷的初始流量和废弃时间可通过资料收集获得。在关闭/废弃煤矿现场对甲烷排放速率进行监测, 然后根据测得的数据代入下降方程 (1)、(2) 中, 拟合出关闭/废弃煤矿甲烷排放下降速率曲线。

3.2.2　甲烷排放速率主控因素分析

　　随着采矿活动的停止, 关闭/废弃煤矿中甲烷总排放量减少, 但由于残余煤炭仍会解吸甲烷, 关闭/废弃的煤矿可以在很长一段时间内以接近稳定的速度释放甲烷。影响关闭/废弃煤矿甲烷排放速率主要有以下因素:

　　(1) 煤矿废弃时间。甲烷排放速率在煤矿关闭/废弃后显著下降并随着时间的推移趋于平稳, 煤矿废弃时间是影响甲烷排放速率最重要的因素。WILLIAMS 等[52]通过静态通量室以及温室气体分析仪在关闭/废弃煤矿、油气井布置采样点进行连续监测, 研究结果表明由于储层逐渐枯竭或压力差平衡等原因, 甲烷排放量在关闭/废弃后的最初几年达到峰值, 而后随着时间的推移而减少。关闭/废弃煤矿甲烷释放速率符合图 4 所示下降规律, 但在干矿中, 甲烷排放量会在前 5 a 迅速下降而后持续数十年, 而水淹煤矿甲烷释放会随着水淹程度迅速下降。美国国家环境保护局 (U.S. Environmental Protection Agency, EPA) 等调查显示, 容易发生洪水的煤矿甲烷排放量将在 3 a 内减少到其原始产能的 20%, 而后 8 a 内完全淹没几乎不再释放甲烷[74-75]。
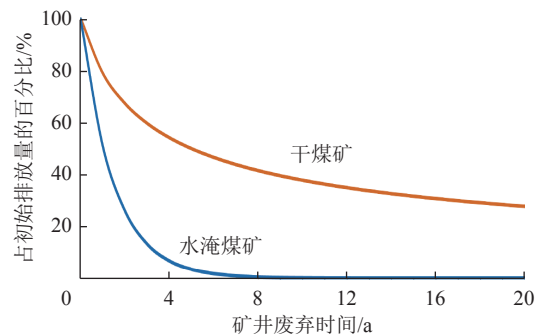

图 4　水淹煤矿和干煤矿甲烷排放量随时间变化情况[19]

Fig.4　Variation of methane emission from flooded coal mine and dry mine over time[19]

　　(2) 煤中残余甲烷含量。关闭/废弃煤矿采空区及

邻近煤层残余甲烷含量会对甲烷排放速率造成影响[76],煤阶、煤层深度等因素会导致煤中残余甲烷含量出现差异[19]。不同煤阶煤的吸附解吸性能不同,进而影响煤矿采空区游离甲烷含量[77]。美国长壁开采煤矿数据显示,煤层压力会导致煤层甲烷量随着开采深度的增加而增加。2018 年,我国煤矿平均开采深度为 510 m,最深已达 1 450 m[78],我国埋深超过 1 000 m 的煤炭资源丰富[79],随着浅部煤炭资源的枯竭,中国煤炭开采正向深部快速推进[80],未来关闭/废弃煤矿会面临更严重的渗漏隐患。

(3) 煤矿状态。采空区甲烷排放速率会受到煤矿状态的影响,如通风、密封、水淹等情况。SECHMAN 等[10, 81] 研究发现,2004 年关闭的 Maja 矿区甲烷体积分数明显升高,主要原因在于通风系统的关闭导致甲烷聚集在采空区,加剧井下甲烷通过断层和裂隙系统向地表的迁移。许多煤矿在关闭/废弃后会对其通风口、井口进行堵塞密封,防止甲烷气体的逸出,但随着时间的推移,甲烷可能会通过密封塞或联通地表的裂隙渗出[82]。除此之外,洪水通过稳定煤层静水压力,阻塞甲烷排放通道,大幅减少或有效抑制煤矿中甲烷流动,也限制了关闭/废弃煤矿的甲烷排放。

(4) 覆岩性质。地下煤矿开采改变了岩体中的应力状态,若开采强度超过岩石单元的抗剪强度,将引起岩石结构的变形或破坏。THIELEMANN 等[60] 使用通量室法监测到煤矿覆岩断层带顶部的甲烷排放,阐明了破碎岩石对气体运移的集中作用,并监测了 3 个覆岩厚度不同的监测点的甲烷排放量,研究发现随着覆岩厚度的增加,气体运移受阻,甲烷排放量将减少。

3.3　地表渗漏甲烷监测方法

国内外对于特定领域的甲烷排放监测主要针对油气井领域,煤矿区的甲烷排放未受到重视,监测手段较少。准确监测甲烷排放,可以为开展甲烷利用和减排提供支撑。因此,本节借鉴油气等领域中甲烷地质渗漏监测手段,为煤矿区甲烷渗漏监测手段提供参考。

3.3.1　卫星遥感监测

通过卫星追踪甲烷排放、收集甲烷排放信息是甲烷监测最常用的方法之一,利用卫星上的光谱仪捕捉图像,而后通过算法自动识别,实现石油和天然气设施释放的甲烷羽流检测。日本于 2009 年发射了“温室气体观测卫星”(GOSAT)、NASA 的 Aqua 卫星、哥白尼哨兵-5P 卫星上的对流层监测仪器等卫星都具有甲烷观测能力[83]。美国环境保护协会预计于 2023 年初发射新的甲烷遥测量化卫星,搭载基于光谱仪的高性能高精度的甲烷遥感系统,以帮助追踪并最终减少

全球油气开采过程排放的甲烷。

MAASAKKERS 等[84] 使用 GOSAT 卫星获得 2010—2015 年北美上空的大气甲烷柱数据,并据此反演出北美不同部门的甲烷排放量。LAUVAUX 等[85] 利用哥白尼哨兵-5P 卫星遥感监测数据,得到了全球含油气盆地甲烷排放情况,发现北美、东欧−中亚、中东等甲烷排放量远大于其他地区 (图 5)。SCHNEISING 等[86] 通过 ENVISAT 卫星探测器数据,推测近年来油气井盆地甲烷排放量的增加是由于石油和天然气产量而引起的。然而,卫星遥感监测的精确度还需要技术突破,气候条件对监测有较大影响,且容易受到与甲烷波长相近的气体 (如二氧化碳) 的干扰。

图 5　基于卫星遥感监测的全球含油气盆地甲烷排放速率[85]

Fig.5　Methane emission rate of global oil-gas basins based on satellite remote sensing monitoring[85]

3.3.2　地表 (含近地表) 监测

地表监测主要是对煤矿采空区上部土壤或井口、通风口等位置,通过通量室法、地球化学探针法等方法进行监测。目前广泛采用通量室法或静态箱法测定点源甲烷通量,该方法通过室中体积分数随时间的变化来追踪甲烷的排放速率[87],静态室测量特定时间段内甲烷水平的波动,而动态室能够通过比较入口和出口体积分数之间的差异记录瞬时或连续甲烷排放体积分数[88]。唐俊红等[89] 使用便携式快速温室气体分析仪与静态通量箱技术结合,监测新疆大宛齐油田甲烷渗漏情况。LEBEL 等[90] 以美国加利福尼亚州的 121 口关闭/废弃油气井为研究对象,使用快速移动羽流积分法和静态通量室对其甲烷排放量进行检测,该方法操作简单,成本低,且可实现在线监测。HENDEL 等[91] 通过在地表安装 43 个地球化学探针,监测了波兰 Murcki-Staszic 关闭/废弃煤矿地表区域甲烷碳同位素变化,发现部分区域地表土壤中甲烷体积分数明显上升,证实关闭/废弃煤矿残留甲烷能够通过采空区上覆岩层采动裂隙与未完好密封的关闭/废弃煤矿竖井渗漏至地表。THIELEMANN 等[60] 对鲁尔

盆地 7 个位置进行了为期 28 个月的通量室测量, 检测到在活跃和关闭/废弃的矿区都存在甲烷排放, 且与活跃矿区相比, 关闭/废弃矿区的排放强度局部缓慢下降, 证实了甲烷排放下降规律。

3.3.3　其他监测方法

随着甲烷对温室效应的主控作用愈来强烈, 国内外对甲烷排放的关注也随之增强, 对其监测方法也一直在改进与革新。对于煤矿等无法封闭测量的排放源可以利用微气象技术监测, 通过在特定地点、高度建设发射塔或测量站, 并在塔上配备甲烷浓度传感器和风速/风向传感器, 结合大气传输模型估算甲烷排放情况, 该方法能够连续获取数据且能获得较大区域范围内甲烷排放量, 但成本高、技术复杂、环境要求较高[92-93]。配备甲烷红外传感器无人机具有移动性强、灵活度高的优势, 结合遥感技术可以实现微小渗漏的监测, 获得精度较高的甲烷排放数据[94-95]。顺风测量甲烷体积分数再测量背景环境中甲烷体积分数和风速、风向及空气湍流, 再将测量数据与扩散模型结合也是估算甲烷排放速率的一种方法[96-97]。甲烷渗漏在一定程度上会改造地表物理、化学及生物特征, 因此, 可以通过监测甲烷氧化菌数量、地表植物生长发育情况、矿物特征等变化实现间接甲烷监测[98-100], 但间接监测受环境条件干扰大。

4　关闭/废弃煤矿甲烷减排对策与技术挑战

煤矿废弃关闭后, 采空区不断积聚大量游离态甲烷并由井口、裂隙等通道排放至地表。袁亮等[1, 101-102]提出, 要推动废弃煤矿残余甲烷全浓度利用, 减少能源浪费。尽管甲烷对温室效应的影响极大, 但作为一种清洁高效的能源, 有必要结合关闭/废弃煤矿甲烷特点, 采取相应技术手段将其回收利用, 或通过微生物、水淹、封堵排放通道等手段遏制甲烷向大气排放, 实现关闭/废弃煤矿甲烷的有效减排。

4.1　地面抽采利用

随着煤矿废弃时间的推移, 遗煤、邻近煤岩层等位置中甲烷会逐渐释放到采空区, 形成甲烷储层, 特别是对于未密封且没有被水淹没的关闭/废弃煤矿[8]。除抑制甲烷排放、减缓温室效应外, 抽采利用井下甲烷还有利于提高矿山安全性, 同时带来经济效益[103], 但我国煤矿甲烷的抽采利用目前主要集中于煤矿开采前预抽采和开采中卸压抽采, 对于关闭/废弃后煤矿内甲烷的抽采技术还有待探索。由于能源结构转型等原因, 英国自 1947 年开始相继关闭 900 多座煤矿, 截至 2010 年, 英国关闭/废弃煤矿瓦斯涌出量约为 6.8×10^7 m³, 回收利用量约为 4.6×10^7 m³。赵向东[104]

考虑到井下甲烷体积分数差异大, 提出了废弃矿井采空区分级抽采利用技术体系, 并已在山西晋城、西山等矿区成功应用。HU 等[105]在关闭的山西沁水永安矿使用井底位置不同的地面井提取采空区甲烷, 现场研究表明, 采空区井底甲烷体积流量是裂隙带井底的 2.5 倍, 井底靠近采空区底部更利于抽采。

对于关闭/废弃煤矿甲烷的抽采利用, 国内外许多专家学者认为主要存在 3 方面局限性[31-32, 106]: ① 部分井下甲烷体积分数处于甲烷的爆炸极限范围, 低浓度甲烷的安全运输成为问题; ② 采空区地质条件复杂, 且采空区钻孔等关键数据缺失, 甲烷直接抽采存在较大盲目性; ③ 采动裂隙不断发育形成的裂隙网络连通地表大气, 抽采时空气的渗入将导致甲烷浓度及甲烷抽采效率降低。

4.2　井下甲烷原位爆燃发电

甲烷是一种易燃易爆气体, 理论上甲烷爆炸极限为 5%～16%, 但由于井下温度及煤尘等影响因素, 其爆炸极限远宽于此[107]。我国煤层地质条件复杂, 透气性差、渗透率低, 抽采钻孔密封性较差, 导致体积分数在 8%～30% 的低体积分数甲烷占比较大[30], 借鉴聂百胜等[108-109]提出的深部流态化开采中原位煤粉爆轰发电技术, 可采用井下甲烷原位爆燃发电技术, 避免甲烷长时间管道运输可能带来的安全问题, 原位建立发电系统, 实现井下发电。借鉴连续爆轰发动机原理[110-111], 诱导爆轰管中甲烷点燃爆轰, 产生爆轰波, 推动涡轮机转动从而带动发动机, 实现机械能转变为电能, 完成甲烷爆轰做功发电的整个过程。

关闭/废弃煤矿井下爆燃发电技术尚未成熟, 还存在以下问题有待解决。甲烷爆轰会产生巨大的能量造成爆轰管道内的温度快速升高, 需要创新技术快速降温以维持管道温度恒定。另外, 井下环境大多是甲烷/煤尘混合体系, 该体系爆炸危险性大[112-113], 一旦爆炸会产生巨大爆炸能量会导致煤矿坍塌, 因此如何选择合适的点火、作业方式保证操作人员的生命安全亟待解决。

4.3　微生物降解甲烷

甲烷氧化菌是一类能够以甲烷为唯一碳源和能源的微生物[114], 并且在常温常压下便可利用自身含有的酶将甲烷氧化为甲醇, 最终产生 CO_2 和水。微生物降解甲烷技术构想最早是在 1939 年由著名煤化学家 A.З.尤洛夫斯基提出, 此后各国都展开了类似的研究。陈东科等[115]在煤样中添加甲烷氧化菌, 并对甲烷浓度变化进行监测, 证实了甲烷氧化菌对煤矿甲烷的降解作用。CHEN 等[116]探究了 pH 值、培养温度和菌种比对菌体生长的影响, 研究发现 pH 为 6.87、培养

温度 28.83 ℃、细菌浓度 0.84 CFU/mL、降解时间 7.16 h 时，甲烷氧化菌降解甲烷效果最好。崔学峰等[117]自主设计了一套模拟煤矿内低浓度甲烷的风流系统，研究发现在风流中甲烷体积分数 (10%～30%) 内，风速越小、甲烷体积分数越高，甲烷氧化菌的氧化效果越明显。王振江[118]通过甲烷氧化菌降解低体积分数甲烷实验，发现在甲烷氧化过程中会存在有机化学原料甲醇的生成，带来经济效益。借鉴微生物降解甲烷机理，向煤矿注入微生物实现煤矿瓦斯治理受到学者广泛关注，并已通过工程实践证实[119-120]。

利用微生物减少关闭/废弃煤矿甲烷排放技术还存在一定局限性，如关闭/废弃煤矿井下环境复杂，而甲烷氧化菌培养过程中所处环境条件对其活性有较大影响[121-124]；另外，微生物降解甲烷过程中会产生高毒性产物甲醛[125-126]，危害人体生命健康。

4.4　注水淹没关闭/废弃煤矿

研究发现，许多关闭/废弃煤矿会由于地下水或地表水的侵入被淹没，水的静水压力大大减少了煤中甲烷的解吸[46]，与此同时还会阻断甲烷在采空区中的流动，从而从根源上抑制甲烷排放[31, 46]。EPA 数据显示，一旦关闭/废弃地下矿山被淹没，甲烷排放量减少到接近零。近年来，持续性强降雨等极端天气频发，2021年仅山西就由于洪灾关停 27 座煤矿。欧洲、乌克兰、俄罗斯大部分容易发生洪水的关闭/废弃煤矿会在8 a 左右被完全淹没，甲烷排放随着淹没程度迅速下降，直至彻底淹没后完全不排放甲烷[127-128]。基于此，提出注水淹没关闭/废弃煤矿，防止采空区内甲烷排放的构想。

但该构想存在以下潜在问题：① 注水淹没关闭/废弃煤矿可能会带来矿井水污染问题。WRIGHT等[129]对澳大利亚关闭/废弃 Berrima 煤矿研究发现，洪水涌入矿井后，引发了重金属严重超标的酸性矿井水泄漏。英国冲突与环境观察站报道，由于洪水等原因，乌克兰 Red October 煤矿酸性矿井水伴随重金属甚至放射性核素迁移到当地地下水和河流环境，给人类健康及环境带来极大危害；② 地下水位的恢复可能导致"活塞效应"[10, 81, 130]。随着关闭/废弃煤矿井中注水工程的推进，地下水位不断上升，使水面上方开放空间中的自由气体压力增大，产生压力梯度，届时甲烷会通过上覆岩层被推向地层表面。因此，水能否实现关闭/废弃煤矿的甲烷减排还存在争议；③ 关闭/废弃煤矿的水层深度、涌水量等水文信息仅有通过位于煤矿中的水文监测井来获取，而目前已经关闭/废弃的煤矿中大部分没有水文测井且废矿条件难以实现水文测井的建设，因此该工程实现较为困难[131]。

4.5　甲烷排放通道封堵

煤矿关闭/废弃后，甲烷排放通道主要包括井口、通风口、采动裂隙带[101, 132-133]。国内外学者指出，密封状态良好的关闭/废弃煤矿井可以成为气藏，防止甲烷渗漏以减缓温室效应，同时减少抽采时漏风，提高抽采效率[4, 31, 134]。王双明等[135] 提出通过功能性充填技术，使采空区盖层、功能性充填体和底板形成封闭空间，实现采煤工作面空间封闭。通过对甲烷排放通道的全方位封堵，防止采空区与地表连通，能够实现甲烷减排，同时有利于提高甲烷抽采率，因此，亟需对甲烷排放通道进行封堵。

对于关闭/废弃煤矿，主要采用密实材料或混凝土对井口、通风管道及钻孔填堵以防止甲烷泄漏。但即使煤矿已被封闭，甲烷可能通过堵塞材料与堵塞位置间的缝隙、材料本身产生的裂隙以及煤矿上覆岩层的裂隙带渗出地表，仍会造成强温室气体甲烷的排放[136]。井口、通风口等数量少、封堵容易，而覆岩采动裂隙分布广、开度小，封堵难度最大、成本最高。

岩体裂隙封堵可通过自修复或人工修复实现，基本原理包括黏土矿物水化膨胀、盐岩再结晶、钙/铁质矿物化学沉淀、注水泥浆等[137-142]。对于黏土岩和泥岩地层，可通过水化膨胀实现自修复，再结晶主要用于盐岩地层裂隙自修复。煤矿上覆岩层的岩性主要为泥岩、砂岩、碳酸盐岩，在地下水及 CO_2 气体影响下，覆岩中矿物元素受溶解和溶蚀作用生成次级矿物及新的结晶沉淀物[143-145]，可实现裂隙修复。JU等[146-147] 基于典型覆岩裂隙自愈案例的阐述和实验验证分析，揭示了地下水与覆岩裂隙之间的长期相互作用发生的自修复机制，证实了裂隙中发生矿化反应导致 $Fe(OH)_3$、$CaCO_3$ 等沉淀物的形成。鞠金峰等[142, 148]通过水−CO_2−岩相互作用实验验证了覆岩导水裂隙在地下水作用下的自修复现象，并提出通过注入含 Ca^{2+} 和 Fe^{3+} 的反应试剂，使其与地下水发生反应，产生矿物胶结沉淀，能够有效实现覆岩导水裂隙的修复。李全生等[149]根据神东矿区煤层开采 10 a 后钻孔原位探测数据，揭示了垮落带、导水裂隙带均存在明显的自修复效果。如图 6 所示，在自修复作用下，地表采动裂隙开度显著降低。同时，理论研究与工程实践证实[149-152]，废弃煤矿垮落带和离层裂隙带也可通过人工注浆修复。

裂隙通过自修复达到完全封堵所需时间普遍介于数月、数年，为此工程上常利用微生物加速诱导碳酸钙沉淀封堵裂隙[154-159]。钱春香等[160] 试验发现，在含 Ca^{2+} 的裂缝环境中，细菌能加速诱导 $CaCO_3$ 沉积，实现微生物对水泥基裂缝的快速修复。微生物诱导
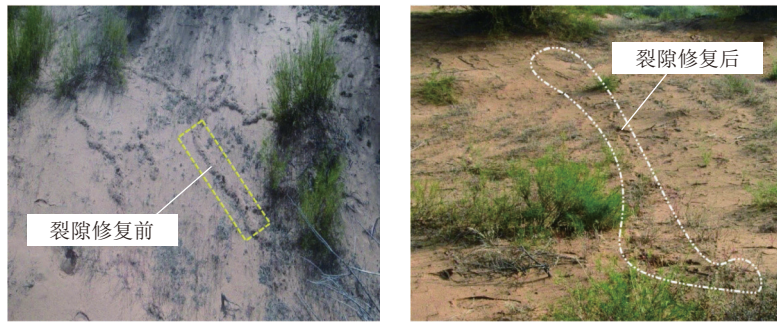
图 6　煤矿地表采动裂隙自修复前后对比[153]

Fig.6　Self-healing of surface fracture in closed/abandoned coal mine[153]

碳酸钙沉淀方法虽具有高可注性、强密封的优势，然而其用于废弃煤矿覆岩巨大数量裂隙的修复封堵时，面临着大规模细菌培养带来的经济、时间成本等问题。

地下水在长期水岩反应作用下封堵导水裂隙、微生物诱导碳酸钙沉淀封堵水泥裂隙等修复机制表明，水−CO_2−Ca^{2+}反应沉淀对采动裂隙通道具有显著封堵作用，说明利用矿化反应方法进行采动裂隙封堵是可行的。因此，借鉴微生物诱导碳酸钙沉淀封堵裂隙的基本原理，笔者提出采动微裂隙矿化修复方法，其原理是直接采用 CO_2 气体作为碳酸根离子来源，替代微生物催化分解产生碳酸根的缓慢化学反应过程，同时

以高碱性 $Ca(OH)_2$ 溶液作为钙离子供应，通过阴阳离子在相向方向对流、扩散，最终在裂隙内快速实现碳酸钙沉淀胶结，封堵采动裂隙网络。该方法的技术要点如图 7 所示，首先向上覆岩层附近钻水平孔，通过钻孔向裂隙带注入高碱性 $Ca(OH)_2$ 溶液，然后向采空区注入低压 CO_2 气体，向下渗流的 $Ca(OH)_2$ 溶液与向上扩散的 CO_2 气体接触后，在裂隙带内形成碳酸钙沉淀。该方法采用 $Ca(OH)_2$ 溶液代替传统注粉煤灰浆，具有安全环保、成本低、黏度低、密封强、可以实现覆岩大规模微裂隙封堵等优势，但目前封堵甲烷排放通道技术仍不完善，需要不断研究创新，例如钻孔位置、方式的优化以及如何控制沉淀分布等问题仍有待解决。
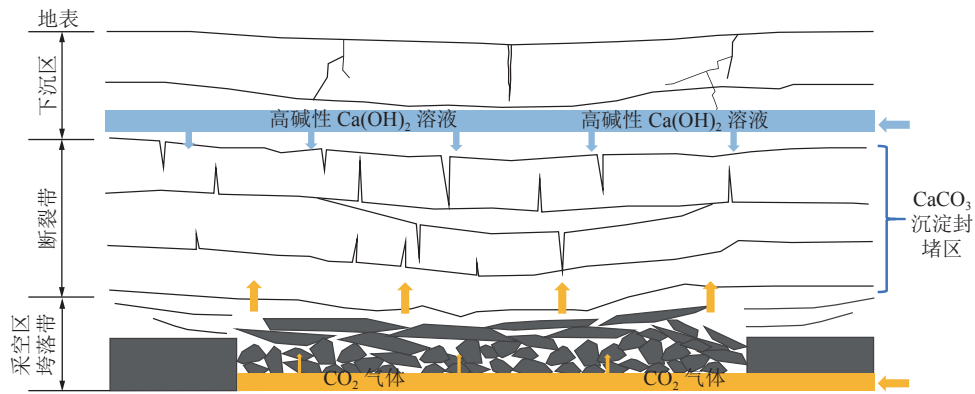
图 7　关闭/废弃煤矿覆岩采动裂隙矿化修复示意

Fig.7　Schematic diagram of mining-induced fracture mineralization restoration in closed/abandoned coal mine overlying rock

5　结　　论

(1) 全球关闭/废弃煤矿数量逐年增加，到 2050 年关闭/废弃煤矿甲烷排放量在煤炭开采释放甲烷总量的占比可能超 20%，解决关闭/废弃矿井甲烷排放问题刻不容缓。基于月下降曲线法、物质平衡法及物质构成法估算结果，合理推断出我国现有关闭/废弃煤矿甲烷残存量巨大，将成为重要的碳排放源。

(2) 关闭/废弃煤矿上覆岩层存在大量采动裂隙，

加之存在未封闭井口，煤中残留甲烷解吸后将排放至大气，造成甲烷长时间持续排放。采用下降速率曲线拟合得到关闭/废弃煤矿甲烷排放速率与煤矿废弃时间关系，揭示了煤矿废弃时间对甲烷排放速率的重要影响，概述了利用卫星遥感、地表直接测定甲烷排放速率等方法可以实现煤矿甲烷排放监测。

(3) 根据关闭/废弃煤矿甲烷排放特征，提出抽采利用、原位爆燃发电、微生物降解甲烷、注水淹没关闭/废弃煤矿、甲烷排放通道封堵的减排对策。考虑

到安全、成本、治理时间及地下水污染等局限性，认为矿化修复封堵采动裂隙强化甲烷封存具备反应速率快、成本低等优势而具有较高可行性，未来工作可以进一步结合关闭/废弃煤矿采动裂隙发育特点进行研究，推动矿化沉淀修复采动裂隙强化甲烷封存成为关闭/废弃煤矿碳封存的重要发展方向。

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