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煤基固废高值化利用研究进展

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摘　要

煤炭在开采、利用过程中产生了大量煤基固废，资源化利用是煤炭行业绿色转型的关键。基于煤基固废的理化性质对其在高值化利用方面的研究进展进行了归纳总结：(1) 构建了多级孔吸附材料用于环境修复；(2) 开发出硅铝基土壤调理剂；(3) 制备了光催化复合材料。同时，阐述了从煤基固废中回收有价金属如锗、氧化铝的技术及其优化策略。最后，指出煤基固废当前资源化过程中存在的二次污染与能效问题，提出了“组分梯级利用－过程协同调控－产业链延伸”的优化框架，为构建煤基固废闭环利用体系提供理论与工程指导。

关键词

煤基固废，高值化利用，有价元素，复合材料

Research Progress on High-Value Utilization of Coal-Based Solid Waste

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Abstract

Coal mining and processing generate substantial solid waste, and its resource utilization is crucial

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for the green transformation of the industry. The research progress in the high-value utilization of coal-based solid waste is summarized based on its physical and chemical properties: (1) constructing hierarchical porous adsorbents for environmental remediation; (2) developing silica-aluminum-based soil conditioners; (3) designing photocatalytic composites for pollutant degradation and hydrogen production. It systematically elaborates on the technologies and optimization strategies for recovering strategic metals (e.g., germanium, aluminum oxide) from the solid waste. Finally, it identifies the secondary pollution and energy efficiency issues existing in the current resource utilization process, and proposes an optimization framework of "graded component utilization - synergistic process regulation - industrial chain extension", providing theoretical and engineering guidance for constructing a closed-loop utilization system of coal-based solid waste.

Keywords

Coal-Based Solid Waste, High Value Utilization, Valuable Element, Composite Material

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1. 引言

中国是全球最大的煤炭生产与消费国。在这一背景下，煤炭全生命周期产业链将持续产出大量煤基固废(包括煤矸石、粉煤灰、脱硫石膏和气化渣等)，其产生的次生环境问题已成为制约煤炭行业发展的瓶颈问题[1]。其中，我国煤矸石的堆存量高达约 50 亿吨，且每年新增产量超过五亿吨[2]。大规模的露天堆放不仅侵占大量土地资源，其产生的扬尘与淋溶污染对区域大气、水体和土壤构成复合型环境威胁，如废弃物浮尘影响空气质量，有害物质随降水入渗污染地下水与地表水等[3]。

当前“双碳”目标大背景下，煤基固废高值化资源利用，是实现煤炭行业清洁高效绿色转型的必由之路。近年来，国内外学者开展了大量煤基固废再利用方面的研究工作，如煤矸石用于发电(基于流化床燃烧技术的发电效率可达 38.7%)、地下矿山充填(膏体充填材料抗压强度突破~2.4 MPa)以及土木材料(骨料替代率大于 30%的混凝土耐久性提升 17.2%) [4]-[7]。此外，煤基固废中含有大量的 SiO_2、Fe_2O_3 和 Al_2O_3 等，可作为高附加值的硅源、铝源[8]。粉煤灰可作为光催化剂的载体[9]、水泥浆原料[10]和回收有价金属等[11]。煤气化渣规模化利用的重要途径是作为建材[12]和生态修复原料等。

2. 粉煤灰的化学组成及其应用

2.1. 粉煤灰的化学组成

由表 1 可知，粉煤灰的物相结构主要由无定形基质和晶相组分构成，且含有大量的 SiO_2 和 Al_2O_3，其理化性质与原料煤的类型密切相关[13]。根据钙、硅、铝、铁的比例可分为 F 类和 C 类，C 类粉煤灰主要由褐煤和次烟煤燃烧产生，CaO 的含量超过 20%；同时，F 类粉煤灰来自高等级煤的燃烧，且 CaO 含量少于 10% [14]。

如图 1 可知，粉煤灰为粒径大小不一、较为规则的球形结构，其表面光滑致密，基本无孔隙，覆盖了大量的玻璃微珠[15]。粉煤灰的形态效应会影响混凝土的流变性能以及硬化混凝土的初始结构。粉煤灰颗粒形貌影响其在混凝土中的分散性和比表面积，从而其水化反应速率和活性。

Table 1. Chemical compositions of fly ash (mass fraction/%)
表 1. 粉煤灰的化学成分(质量分数/%)

组成	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	其它
占比	1.3~65.76	1.5~40.12	1.5~6.22	1.4~16.80	1.2~3.72	1.0~6.00	1.1~4.23	1.0~2.14	1.63~29.97
平均值	33.5	20.8	3.9	9.1	2.5	3.5	2.5	1.6	15.8

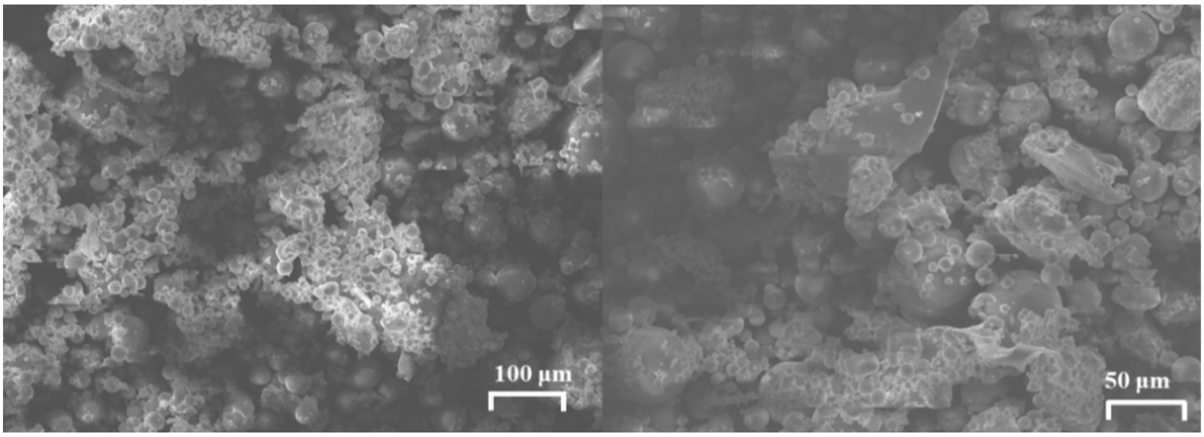

Figure 1. SEM image of fly ash
图 1. 粉煤灰的 SEM 图

2.2. 粉煤灰的高值化利用

2.2.1. 粉煤灰基分子筛

近年来，因原料来源广泛和合成技术的不断成熟，粉煤灰的高值化应用引起人们的关注[16]。Zhang 等[17]采用单步热液水热法，合成了结晶良好的 Na-P1 型沸石，对 8000 mg/L 高浓度工业含磷废水吸附效果显著；He 等[18]以粉煤灰、氢氧化钠和水为原料，经 700℃焙烧除碳、3%盐酸酸化、碱熔活化后，在 100℃下水热晶化 24 h 合成出 A 型沸石分子筛，对水中 Ni^{2+}去除率达 94%；Panek 等[19]以粉煤灰为原料制备出比表面积 728 m^2/g 的 Na-X 型沸石，在 pH = 5、100 r/min 振荡 3 h 条件下开展吸附实验，可高效吸附水中的 Pb^{2+}、Zn^{2+}重金属离子。

Adamczyk 等[20]通过引入超声空化预处理，采用 3 mol/L NaOH 碱性热液法于 80℃反应 8 h 合成晶相纯度为 92%的沸石分子筛；Chen 等[21]采用超声和微波协同法制备出粉煤灰基 A 型分子筛，晶化时间较单一水热法缩短 70%；Marhoon 等[22]利用水热合成法合成高纯沸石，能有效去除水溶液中的 Ni、V 重金属离子；Teng 等[23]采用新型热堵塞法，将粉煤灰与硫化钠混合后于 140℃反应 4 h 制备出具有多孔结构的 CAN 型分子筛，对水溶液中的阴离子具有良好的吸附性能；El-Naggar 等[24]采用熔融法提取法制备出 NaA-X 分子筛，能有效去除水溶液中的铯离子。

2.2.2. 粉煤灰基光催化材料

近年来，粉煤灰基制备光催化材料制备工艺的研究不断发展。Liu 等[25]采用两步法(溶剂热 + 原位水解)合成了 Ag@AgCl 掺杂 TiO_2/粉煤灰复合光催化材料，对罗丹明 B 的降解率接近 100%，分别是 AgCl-TiO_2/粉煤灰和 Ag@AgCl 的 1.56 倍，1.33 倍；Zhou 等[26]通过溶胶–凝胶法在粉煤灰表面负载纳米 TiO_2，其光催化活性提升至 42%；Yang 等[27]以活化沸石粉煤灰漂珠为载体负载 TiO_2，其活性位点暴露率增加 37%。与此同时，ZnO/粉煤灰、Bi_2WO_6/粉煤灰、ZnS 包裹的胺功能化粉煤灰纳米复合材料(ZnS/A-FA)均

实现光催化性能的显著提升[28]。除水热合成法、溶胶–胶凝法外，静电纺丝工艺、超声辐照耦合法等新兴技术在粉煤灰基光催化材料制备领域逐步兴起[29]。

2.2.3. 有价元素的提取

粉煤灰中富含锂、铝、镓、镍、稀土及锗等有价元素，提取利用是其高值化利用的重要途径之一[30]。其中，锗氧化物具有高折射率特性，可用于光纤玻璃组件。Hernández-Expósito 等[31]采用离子浮选法，以邻苯二酚为络合剂、控制 pH 为 4~7，从粉煤灰渗滤液中选择性回收锗，回收率超 92%。Dardona 等[32]对粉煤灰进行酸浸预处理后，利用有机硅吸附材料实现稀土元素高倍富集，分离产物中稀土含量达 10 wt%。Xing 等[33]采用碱溶脱硅协同 HBTA-TOPO 萃取工艺，实现从高铝粉煤灰中铝组分的连续提取，效率高达 99.68%；Cao 等[34]采用一步水热–碳分工艺，在硫酸浸出体系中加入氟化钠，将氧化铝浸出率从 76%提升至 91%。针对传统酸浸法存在的重金属浸出、硅渣生成等环境风险；Zhou 等[35]利用粉煤灰污泥中残留 Al^{3+}，在常温下通过非均相酯化反应改性吸附剂表面，实现 Gd^{3+}的高效分离；Xue 等[36]采用电化学氧化还原技术，包括浆料电解、膜电解、熔盐电解等，实现有价元素的绿色提取。

3. 煤矸石的高值化利用

3.1. 煤矸石的化学组成

煤矸石是原煤开采与清洗过程中产生的伴生固体废弃物，长期堆存会对环境造成严重的污染[37]。由表 2 可知，煤矸石的主要化学成分为 SiO_2 和 Al_2O_3。目前，煤矸石主要用于发电[38]、填埋[39]、道路建设[40]、建材[41]。煤矸石中还含有镓、锗、铀等微量有价元素。镓、锗等稀有金属元素是通信、航空航天、可再生能源等行业所需的新型材料[42]，铝和铁等金属元素也有利用价值[43]。

Table 2. Main chemical compositions of coal gangue
表 2. 煤矸石的主要化学成分

煤矸石种类	SiO_2/%	Al_2O_3/%	Fe_2O_3/%	CaO/%	MgO/%
粘土岩类	24~56	14~34	1~7	0.5~9	0.5~6
砂岩组类	53~88	0.4~20	0.4~4	0.3~1	0.2~2
钙盐类	10~40	3~10	1~10	40~80	1~4
铝盐类	40~55	35~45	0.2~4	0.1~0.7	0.1~1

3.2. 煤矸石基材料高值化利用

由图 2 可知，煤矸石表面粗糙松散，大小不一，层状形态不规则。与煤矸石中的主要矿物成分–高岭石密切相关。煤矸石的化学成分与分子筛相似，因此煤矸石也常被用于制备分子筛[44]。煤矸石火山灰活性偏低，需自燃或锻烧才可获得与石灰化合成新的水化物的能力。

3.2.1. 煤矸石基分子筛

煤矸石的主要矿物相为高岭石，具有 1:1 型层状硅酸盐结构，为分子筛制备提供了天然原料优势。Jin 等[45]以煤矸石为原料，采用水热碱活化法，制备出 NaA 型沸石；Zheng 等[46]以煤矸石为硅铝源，采用中温氯化焙烧与压力酸浸工艺调控煤矸石的硅铝比，并通过碱熔水热法制备出 ZSM-5 分子筛，为其低成本制备开辟了路径；Zhao 等[47]采用“矿物基因重构”法，在不添加有害试剂的条件下将煤矸石转化为高性能多孔多金属硅酸盐吸附剂。

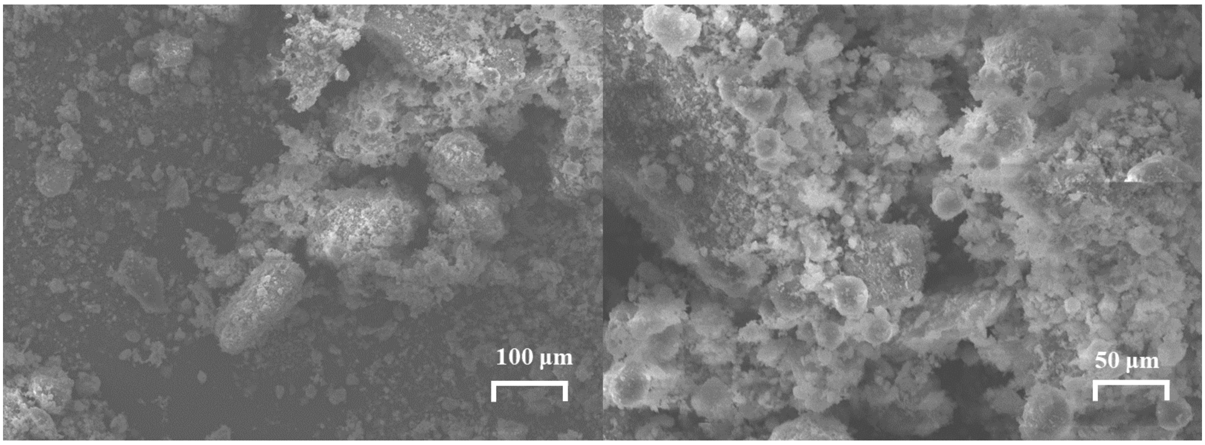

Figure 2. SEM image of coal gangue
图 2. 煤矸石的 SEM 图

3.2.2. 煤矸石基土壤改良剂

煤矸石在农业领域展现出土壤修复与肥效协同的多重价值。研究表明：可将煤矸石开发为兼具土壤修复与肥效缓释功能的智能材料体系。Lv 等[48]将煤矸石、$CaCO_3$与玉米秸秆混合并煅烧时，在 $CaCO_3$与玉米秆粉的协同效应下，制备出速效硅含量 22.97%的硅基肥料；Wang 等[49]通过共热解法制备煤矸石改性生物炭复合材料，兼具污染物吸附与养分缓释功能，可促进种子萌发与生长；Zhang 等[50]采用碱激发矿渣与多硫化钙协同固化体系实现 Cr(Ⅵ)原位固定，为重金属污染土壤修复提供了新方案。

3.2.3. 煤矸石基光催化材料

研究表明，煤矸石的硅铝基骨架结构为构建高效光催化体系提供了便利条件。Guo 等[51]通过水热法合成煤矸石基纳米管光催化复合材料，对亚甲基蓝的降解率达 90%；Bian 等[52]采用原位沉积和水热生长法制备 Ag_3PO_4/TiO_2@分子筛(MS)三元复合光催化材料，可见光下对 100 ppm 甲醛和 50 mg/L 异丁基黄原酸钠的降解半衰期分别缩短至 15 min 和 22 min。

3.2.4. 有价元素提取

有价元素提取是煤矸石高值化利用的重要途径之一。其中，高铝煤矸石的资源化主要依赖酸性浸出、碱性浸出两大路径：酸浸可使氧化铝转化为 Al^{3+}进入溶液，硅富集于浸出渣，实现硅铝分离；碱性浸出通过调控苛性比($\alpha k = 1.4\sim1.6$)与溶出温度(240℃~260℃)，可使铝硅分离系数超 300。Wang 等[53]采用碱浸法处理高铝煤矸石，赤铁矿经还原焙烧后，高岭石转化为铁铝尖晶石与方石英固溶体，实现 96%以上二氧化硅和 97%以上氧化铝的提取；Luo 等[54]采用焙烧活化－拜耳法提取－硫酸浸出组合工艺，在最佳条件下，获得 69.5%的 Al_2O_3；胡等[55]以鄂尔多斯煤系高岭土为原料，与浓硫酸直接反应(无需煅烧)制备硫酸铝与聚合硫酸铝，展现出良好絮凝效果。

4. 煤气化渣的高值化应用

4.1. 煤气化渣的化学组成

煤气化渣是煤炭在清洁转化过程中的典型副产物。煤中有机组分经部分氧化生成合成气，灰分物质则形成粗渣、细渣两类残渣，二者均具备火山灰活性，细渣为含水矿渣，随气流排出后冲刷沉淀而成，含碳量可达 30%以上[56]-[58]，由表 3 可知，煤气化渣中的主要成分以硅、铝、铁为主，此外还有少量的金属氧化物。因粗、细渣在水分、含碳量、粒度等性质上差异显著，由图 3 可知，煤气化渣微观形貌呈

现黏结球形、多孔不规则、孤立球形等多种形态。丰富的孔隙结构赋予其吸附性能，在废水处理、气体吸附等领域具有应用价值。

Table 3. Main chemical compositions of coal gasification slag
表 3. 煤气化渣的主要化学成分

成分	SiO_2	Al_2O_3	Fe_2O_3	CaO	K_2O	SO_3	TiO_2	Na_2O	MgO	P_2O_5	SrO	其它
含量	53.673%	25.929%	7.185%	5.219%	2.331%	1.582%	1.422%	1.136%	1.074%	0.137%	0.09%	0.234%

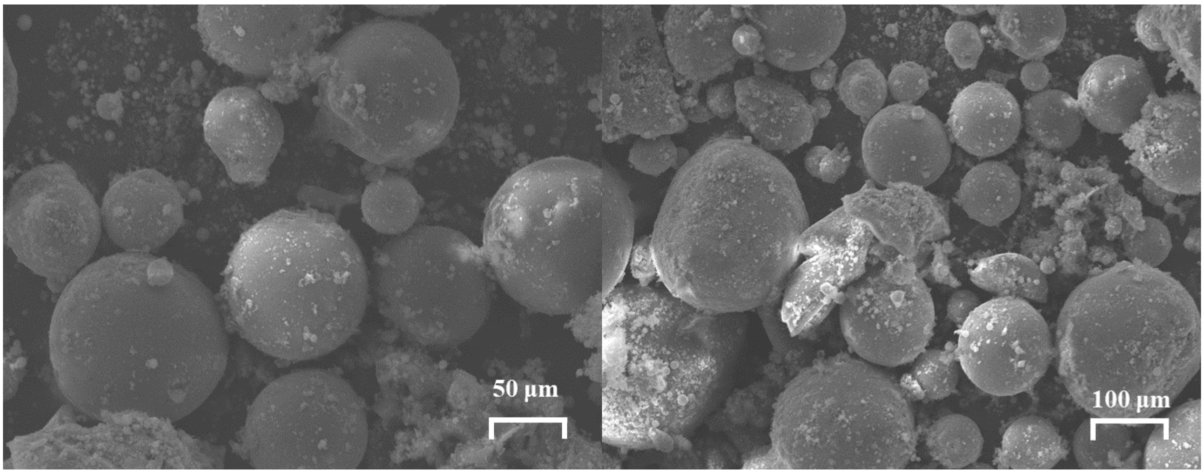

Figure 3. SEM image of coal gasification slag
图 3. 煤气化渣的 SEM 图

4.2. 煤气化渣基材料高值化利用

4.2.1. 煤气化渣基分子筛

煤气化渣硅铝含量高、铁钙含量低，适合制备介孔材料[59]。Duan 等[60]以煤气化渣为吸附剂，实现低浓度汞的高效去除，吸附平衡仅需 10~40 min，在 pH = 4 时去除效果最佳，材料对汞离子具备良好选择性；Liu 等[61]以煤气化粗渣、碳酸钠为原料，采用无溶剂固相法制备出单相 ZSM-5 分子筛；Ma 等[62]以细渣为原料，制备出碳/沸石复合材料，经硫酸亚铁改性后，对 5 mg/L NH_4^+ 和 10 mg/L PO_4^{3-} 的去除率分别达 88%和 99%；Chai 等[63]以细渣为原料，采用梯度合成法，借助 1,3,5-三甲基苯与 CO_2 预活化调控孔结构，并进一步进行氨基改性，制备出结构规则、比表面积大和吸附性能优异的 SBA-15。

4.2.2. 煤气化渣基土壤改良剂

研究表明，气化渣中丰富的硅、钙元素可有效调节土壤 pH 值并固化重金属[64]。Xiang 等[65]研究发现，煤气化渣中的硅铝基骨架与活性组分协同作用，可同步调控土壤理化参数与养分平衡；Zhu 等[66]通过实验发现，施用 5 wt%煤气化细渣可显著促进水稻生长；Xiang 等[67]采用污泥衍生氨基酸改性磁性煤气化渣，不仅可大幅降低除 Fe 外其他重金属的生物利用性，同时提升三种土壤酶活性。

4.2.3. 煤气化渣基光催化材料

近年来，煤气化渣基光催化材料的开放主要聚焦载体与活性组分协同作用。Zhu 等[68]以煤气化细渣中的介孔石英玻璃微球为载体，负载 TiO_2 制备吸附－光催化复合材料，可协同去除罗丹明 B，催化性能优于纯 TiO_2；焦等[69]以改性煤气化渣为载体，水热法制备 ZnO/CGS 复合材料，可高效降解亚甲基蓝，

最优条件下降解率可达 98%；Wang 等[70]利用煤气化渣中的内源铁为原料，成功制备出 Fe_2P 光催化析氢助催化剂。

4.2.4. 碳回收

研究表明，煤气化渣残碳高效回收技术的核心在于界面调控与分离工艺的协同优化；Zhang 等[71]利用煤粉封堵孔隙提高残碳浮选回收率，碳回收率比常规浮选提高了 14.11%；Lyu 等[72]采用充气倾斜液固流化床富集细渣残碳，精矿的点燃损失达到 66.38%，富集度达 82%且能耗较传统浮选降低 37%，表明液固流化床有效地富集了细炉渣中的残余碳。捕收剂体系创新方面，Shi 等[73]采用 7:3 比例的煤油–油酸复配得到捕收剂 PS-1，相较传统煤油捕收剂可减少约 50%药剂用量。

5. 煤基固废的组分差异与协同利用

由表 4 可知，粉煤灰呈玻璃质或微晶质物相且含量高、活性好，适合制备分子筛与光催化材料；煤矸石的主要矿物相为高岭石，结构疏松、分布及来源广泛，在土壤改良、生态修复和化工产品的制备方面具有明显优势[74]；煤气化渣晶相为石英，残碳含量高、孔隙多，可用于硅铝基复合材料合成与碳组分回收[75]。

三种材料侧重点虽有不同，可根据组分之间的差异，从而实现不同固废优势互补，提升煤基固废高值化利用的效率。

Table 4. Comparison of characteristics differences of coal-based solid wastes
表 4. 煤基固废特征差异性对比

固废类型	物相特征	SEM 微观形貌	局限性	高值化研究方向
粉煤灰	玻璃质或微晶质物相且含量高、活性好	粒径大小不一、较为规则的球形结构	钙含量波动大，易影响产物纯度，反应活性受物相条件制约	分子筛制备、光催化材料、建材与充填材料等
煤矸石	主要矿物相为高岭石，结构疏松、分布及来源广泛	表面粗糙松散，大小不一，层状形态不规则	原生矿物活性低，需活化预处理，加工能耗相对较高	土壤改良修复、胶凝材料制备、有价元素提取等
煤气化渣	晶相为石英，残碳含量高、孔隙多	呈现黏结球形、多孔不规则、孤立球形等多种形态	组分波动大，残碳干扰晶化反应，存在重金属淋溶隐患	多孔功能材料、硅铝基复合材料、碳组分回收等

粉煤灰、煤矸石与煤气化渣组分特征存在明显差异，制备分子筛路径与方法有所不同。粉煤灰由无定形基质和晶相组分构成，适合制备高纯度、高比表面积的沸石材料；煤矸石以高岭石为主要矿物相，制备高结晶度沸石成本较低；煤气化渣残碳含量较高，适合制备介孔材料。不同固废间虽存在明显差异，也为制备分子筛提供多种路径及理论依据。

不同煤基固废的晶型组成、表面官能团等存在差异，使得光催化材料的转化路径有所不同[76]。粉煤灰富含活性硅铝组分，适合作为载体负载组分。煤矸石、气化渣残炭更容易构建成光催化复合材料。需不断完善煤基固废转化路径，提高固废实用性。

煤矸石、煤气化渣含有腐植酸等有机物和 N、P、K、B、Zn、Cu 等植物生长的微量元素，可用于制备土壤改良剂、透气保水材料、农肥等用于生态修复的材料，还能促进生态循环[77]。

煤炭在燃烧或转化过程中，微量元素在变性煤基固废中会得到富集，如粉煤灰等变性煤基固废中富

集铝、锗、镓、锂、稀土等战略性矿产资源，采用焙烧、浮选、浸出、萃取、沉淀等工艺进行提取实现资源化利用。煤矸石中高岭石组分的反应活性较弱，主要采用热活化、机械活化以及多种方法协同作用的复合活化等技术手段提取。从煤基固废中提取有价元素是资源循环利用和绿色低碳发展的重要方向。

粉煤灰、煤矸石与煤气化渣应用具有局限性，粉煤灰原料钙含量波动易影响产物纯度，活化工艺易产生碱性废液；煤矸石原生矿物活性低，焙烧预处理能耗较高，部分原料存在重金属淋溶隐患；煤气化渣粗细分化明显，残碳会干扰晶化反应。各种高值化应用仍存在不足，焙烧、酸碱活化等过程存在能耗偏高、废液处置方式仍需改进，高值化产品生产技术需进一步优化，规模化应用开展难度大。

6. 结论

煤炭在中国的能源结构中仍然占主导地位，伴随其开采加工产生的煤基固废产量持续增长，高值化利用路径也日益多样。本文归纳总结了三种主要的煤基固废的高值化利用途径。

(1) 粉煤灰呈现出光催化材料开发、有价金属提取、硅铝基分子筛精准制备的多元发展态势；

(2) 煤矸石需依据其矿物学特性制备高性能分子筛。煤矸石功能化改性材料在土壤修复中有望实现重金属钝化与酶活性提升的环境－农业协同效益；

(3) 煤气化渣的高效利用，需考虑残碳分级回收、硅铝基分子筛制备及土壤改良剂开发均具有显著价值。

当前，煤基固废高值化利用大多停留在实验研究阶段，尚未实现规模化推广。因此，开发步骤简单、适应性强、处理效果优异的综合应用技术，是当前的核心需求。未来还需进一步强化煤基固废的生态效益、经济效益，拓展新领域与新方向，为其高值化利用提供创新思路。

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