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钢渣碳化及其在建筑材料低碳制造中的应用

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摘要:碳捕获、利用与封存(CCUS)技术通过捕获和转化CO_2来减少大气中的CO_2浓度,是实现中国气候目标和可持续发展的关键技术之一.综述了加速碳化钢渣制备低碳建筑材料的研究进度;系统介绍了碳化钢渣建筑制品的性能及其提升机制,以及碳化钢渣辅助性胶凝材料的水化活性及其水化机理;总结了加速碳化技术在建筑材料工业化生产中的应用案例;展望了钢渣碳化未来的发展方向.以期为水泥和混凝土制造业的绿色转型和可持续发展提供参考.

关键词:钢渣;碳捕获;建筑制品;辅助性胶凝材料;微结构;水化机理
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Carbonation of Steel Slag and Its Application in Producing Low-Carbon Construction Materials

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Abstract: Carbon capture, utilization and storage (CCUS) technologies reduces the concentration of CO_2 in the atmosphere by capturing and converting CO_2, is one of the key technologies to achieve China's climate goals and sustainable development. The research progress in the preparation of low-carbon building materials through accelerated carbonation of steel slag was summarized. The performance and enhancement mechanism of carbonized steel slag building products, as well as the hydration reactivity and hydration mechanism of carbonated steel slag supplementary cementitious materials, were systematically introduced. The application cases of accelerated carbonation techndogy in the industrial production of building materials were summarized, and the future development direction was prospected, in order to provide reference for the green transformation and sustainable development of the cement and concrete manufacturing industry.

Key words: steel slag; carbon capture; building product; supplementary cementitious material; microstructure; hydration mechanism

2023年全球与能源相关的CO_2排放量达374亿t,较100年前增加约9倍,加剧了全球变暖.碳捕获、利用与封存(CCUS)技术在应对全球气候变化和实现低碳经济转型中扮演着至关重要的角色,是实现全球气候目标和构建可持续发展未来的关键技术之一.经济和工业的快速发展导致CO_2排放量显著增加,水泥和混凝土制造业的碳排放尤为突出.

中国CO_2排放量约占全球总CO_2排放量的30%.其中,水泥和混凝土制造业所排放的CO_2占比较高[1],对其进行碳减排是实施中国"双碳"战略的重要需求.该背景下,CCUS技术在水泥和混凝土制造业中的应用显得尤为重要.矿物吸收CO_2是一种由富

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含钙或镁离子等碱土金属的矿物与CO_2发生反应,生成碳酸盐产物的过程[2-3].钢渣是钢铁冶炼过程中排放的固体废弃物,近年来其资源化利用问题受到越来越多的关注[4-6].研究发现,与水化反应相比,钢渣中的含钙矿相具有较高的碳化反应活性,在富CO_2环境中可快速发生碳化反应,提升体积安定性和胶凝性能,并且钢渣最高可以封存约20%的CO_2[7].因此,利用CO_2碳化钢渣制备建筑材料成为水泥和混凝土制造业中CCUS技术研究的新热点和新趋势[8-9].

本文综述了加速碳化钢渣制备低碳建筑材料的研究进展;系统介绍了碳化钢渣建筑制品的性能及其提升机制,以及碳化钢渣辅助性胶凝材料的水化活性及其水化机理;总结了加速碳化技术在低碳建筑材料工业化生产中的应用案例;同时也指出了钢渣碳化未来的发展方向.

1 钢渣的组成

由于原矿组成、助熔剂、炼钢工艺和冷却制度等不同,各钢渣的矿物组成差异较大,物化性质各异.从生产工艺上可将钢渣划分为3大类:碱性氧化炉渣(BOFS)、电弧炉渣(EAFS)和钢包炉精炼渣(LFS)[10].

2 钢渣的加速碳化及其在建筑制品中的应用

2.1 低碳钢渣砖

将钢渣微粉、水和细骨料等按需求尺寸加压成型后再碳化,可制备出性能优异的低碳钢渣砖.吴昊泽等[11]研究发现利用窑尾废气碳化14 h后的钢渣砖具备了优异的性能,其碳化增重率可达10.44%,抗折强度和抗压强度分别为5.02、20.06 MPa,经历冻融后抗压强度仍有14.63 MPa.Li等[12]研究了不同碳化压力对低碳钢渣砖性能的影响,发现在压力为0.25 MPa时,其CO_2吸收量(质量分数,文中涉及的吸收量、液固比等除特别注明外均为质量分数或质量比)最高可达16.6%,抗压强度为65.1 MPa.Hou等[13]研究表明,含25%钢渣的碳化钢渣砖CO_2吸收量可达7.5%,7 d抗压强度最高为27.7 MPa.碳化30 min的钢渣砖在3 MPa的压蒸试验条件下表现出良好的体积稳定性和优异的抗冻融性.钢渣砖在碳化过程中的主要反应物为硅酸二钙(C_2S)、硅酸三钙(C_3S)、$Ca(OH)_2$、f-CaO和MgO,碳化产物主要为碳酸盐和硅胶[14].碳化温度和碳化压力影响钢渣砖性能.在碳化初期,适度升高温度和碳化压力有助于增强钢渣砖性能,但温度过高时,水分流失和CO_2溶解度降低导致碳化效率下降,高压力下$CaCO_3$快速沉淀也会堵塞孔隙,阻碍CO_2扩散[15].也有学者利用微生物诱导$CaCO_3$结晶来提升钢渣砖的碳化效率,提高低碳钢渣砖的性能.如Wang等[16]使用能产生碳酸酐酶的细菌对钢渣砖进行碳化.结果表明:碳酸酐酶细菌可以促进CO_2的溶解,并且作为$CaCO_3$沉淀的成核位点能够促进$CaCO_3$的生成;相比未添加碳酸酐酶细菌的钢渣砖,碳化效率提高了48%,强度也大幅提升.有学者也得到类似结论[17].

2.2 碳化钢渣人工骨料

由于骨料体积最大可占混凝土总体积的3/4[18],将钢渣用作混凝土骨料是一种大规模处理钢渣的方法.然而,钢渣骨料中的f-CaO和f-MgO含量较高,可能会导致混凝土体积不稳定[19-20],因此其在混凝土中的应用非常有限,有时甚至被禁用[21].近年来,碳化处理后的钢渣人工骨料因其体积稳定性良好、强度高且能高效矿化CO_2而受到广泛关注.

骨料的CO_2吸收量与钢渣的组成及碳化条件密切相关.通常,较高的CO_2浓度和压力能够增加骨料单位时间内的CO_2吸收量,提高骨料的碳化效率[22-23].但也有研究表明,在碳化过程中提高CO_2压力并非必要条件[24-25].在高碳化压力下,骨料表面迅速形成$CaCO_3$,可能会阻碍CO_2进一步向骨料内部扩散[26-27],对骨料整体的CO_2吸收量有不利影响.此外,环境相对湿度为50%~70%被视为碳化的理想条件[28],相对湿度过高会抑制CO_2的扩散[29].关于温度对钢渣人工骨料碳化过程的影响尚缺乏系统研究,高温有助于Ca^{2+}的溶解,但会降低CO_2的溶解度[30],因此需要进一步明确碳化温度对钢渣人工骨料性能的影响机制.

碳化钢渣人工骨料的CO_2吸收量越高,骨料的单粒颗粒强度越高,其压碎值和吸水率就越低.原因在于,大量$CaCO_3$之间相互胶结使得骨料形成致密的微结构[31-32];骨料碳化过程中永久封存的大量CO_2使得骨料结构更致密,混凝土密度升高[33].由于钢渣中含有f-CaO和f-MgO等易膨胀成分,碳化钢渣人工骨料的体积稳定性问题不可忽视[34].未碳化的人工钢渣骨料往往出现严重的开裂甚至粉化现象;钢渣人工骨料在216 ℃、2 MPa压力条件下蒸压3 h的形态显示,碳化养护消耗了f-CaO和f-MgO,从而改善了碳化钢渣人工骨料的体积稳定性[22].总的来说,CO_2吸收量越高,碳化钢渣人工骨料的体积稳定性越好.

表1总结了碳化钢渣人工骨料的养护制度和性能.提高CO_2吸收量是提升碳化钢渣人工骨料性能的关键.除了改变碳化养护条件,还可以采用增加CO_2

表1 碳化钢渣人工骨料的养护制度及其性能

Table 1 Curing regime of carbonated steel slag artificial aggregates and their properties

Reference No.	$\varphi(CO_2)$/%	CO_2 pressure/MPa	Temperature/℃	Relative humidity/%	Curing time/h	CO_2 uptake (by mass)/%	Compressive strength of individual aggregate/MPa	Bulk density/(kg·m^{-3})	Water absorption(by mass)/%
[35]	99		20	50	96	10.03-15.70	1.8-5.2	1 250-1 280	11.00-12.00
[36]	99	0.1	Room temperature		0.5/1.0/1.5/2.0	12.00-14.00			
[37]	20		20	65	96				8.10
[38]	99	0.2	Room temperature		2	3.44-6.38	2.89-6.10		8.00-11.20
[39]	99	0.3	70						8.12-8.91
[40]	99	0.2	23, 55		3, 9, 72	2.53-11.27			
[41]	20			65	96	4.80-5.57	1.00-3.60	1 166-1 343	8.90-12.10

扩散通道(掺加粉煤灰[22]、稻壳灰[42]等)、添加外加剂(乙二胺四乙酸(EDTA)[43]、聚羧酸减水剂[44]、乙酸[45]、琥珀酸[46]和壳聚糖[47]等)、微生物协同矿化[48-49]、CO_2内养护(掺加生物炭[50]、沸石分子筛[51]和醇胺溶液[52])等多种方法来提升钢渣人工骨料的CO_2吸收量.

3 碳化钢渣辅助性胶凝材料的制备及其性能

3.1 碳化钢渣辅助性胶凝材料的组成和微结构

常钧教授课题组采用半干法碳化技术对钢渣粗粉进行预养护,发现碳化后钢渣粗粉中的f-CaO含量大幅降低,大部分f-CaO转化为$CaCO_3$晶体,硅酸盐矿相的含量也大幅降低[53].由于钢渣中f-CaO的碳化活性最高,当其含量较高时,CO_2主要被f-CaO矿化固定[54].Liu等[55-56]将钢渣微粉与8%水分混合均匀后进行加速碳化,仅30 min即可矿化固定6.14%的CO_2,碳化产物主要由棒状和块状的纳米$CaCO_3$和无定形SiO_2凝胶组成.加速碳化还会导致钢渣微粉颗粒的微结构由致密变为疏松多孔,比表面积大幅增加,并且增大幅度与钢渣碳化程度正相关.文献[57]的研究也得到类似结果.

为提高单位时间内钢渣微粉的碳化转化率,Yang等[58]采用$NaAlO_2$促进钢渣微粉在低浓度CO_2气氛中的碳化效率.Zhang等[59]通过添加微生物显著提升了钢渣微粉的CO_2吸收量,促进了钢渣微粉中f-CaO、f-MgO和C_2S矿相的碳化.Chang等[60]研究发现:超重力、大水灰比条件可大幅提升钢渣微粉的碳化效率,在10 min左右碳化程度(δ_{Ca})即可达80%以上;而在常规的湿法碳化条件下,在碳化60 min时δ_{Ca}仍未达到60%[61].钢渣微粉湿法碳化效率还受到矿物组成、粒径、液固比、CO_2浓度和温度等影响[62-63].总体来看,钢渣微粉的碳化程度越高,其C_2S含量越低,$CaCO_3$含量越高,比表面积越大.

3.2 碳化钢渣辅助性胶凝材料的水化活性和水化机理

碳化钢渣辅助性胶凝材料的组成与微结构较未碳化前有显著差异.利用碳化钢渣辅助性胶凝材料制备的复合水泥力学性能有所提高,尤其是在早期[64-65].Chen等[65-66]研究表明,相比未碳化钢渣制备的复合水泥,碳化钢渣制备的复合水泥3、28 d抗压强度均大幅提高.梁晓杰等[67]发现碳化钢渣3、28 d的活性指数较未碳化钢渣分别提高97%和16%.Liu等[55-56]将碳化钢渣微粉掺入水泥后发现,$CaCO_3$参与水泥水化生成$3CaO\cdot Al_2O_3\cdot CaCO_3\cdot 11H_2O$.无定形$SiO_2$凝胶与水泥水化产物$Ca(OH)_2$反应生成更多的胶凝组分,优化了水泥基材料的孔结构.使用碳化钢渣微粉还能制备超高性能混凝土,其28 d抗压强度可超150 MPa,并且早期强度有一定的提升[57].使用微生物辅助碳化能够进一步提升钢渣的胶凝活性[59,68].原因在于微生物诱导$CaCO_3$成为促进水化的成核位点,并且微生物诱导$CaCO_3$参与水泥水化形成$3CaO\cdot Al_2O_3\cdot CaCO_3\cdot 11H_2O$,提高了试样的密实度.Zhou等[69]和Li等[70]最新研究表明,将含有活性铝相的矿物掺合料(如偏高岭土和高铝矿粉)与碳化钢渣微粉进行复合,掺入水泥基材料可促进体系中$3CaO\cdot Al_2O_3\cdot CaCO_3\cdot 11H_2O$的生成,进一步提升碳化钢渣微粉的水化活性.

碳化钢渣微粉的水化活性还与其碳化程度高度相关[68,71].Liu等[56]发现,相较掺有未碳化钢渣微粉的水泥净浆,掺有高碳化程度钢渣微粉的水泥净浆3 d抗压强度降低17.1%.在28 d龄期时,掺有较高碳化程度钢渣微粉的水泥净浆抗压强度达到110.8 MPa,相比掺有未碳化钢渣微粉的水泥净浆高24.5%.根据核磁共振分析结果可知,较高碳化程度的钢渣微粉与$Ca(OH)_2$混合物中Q^4结构$[SiO_4]^{4-}$的含量由水化

前的39.09%降至水化后的24.78%，表明无定形SiO_2凝胶与$Ca(OH)_2$反应生成了更多的胶凝组分，提高了水泥净浆28 d的抗压强度.

4　加速碳化技术在低碳建筑材料工业化生产中的应用案例

加速碳化技术在建筑材料中的应用主要是将工业产生的CO_2与工业固体废弃物中的碱性成分反应生成稳定的碳酸盐，进而制造出低碳建筑材料，如低碳建筑制品、辅助性胶凝材料和低碳混凝土等.尽管存在技术挑战，但随着政策支持和市场推动，该技术有潜力成为建筑行业绿色转型的重要途径.

图1为低碳建筑材料的工业化生产和应用案例.

2021年12月，在华新武穴工业园基地利用CO_2生产混凝土制品的生产线成功运行.该项目基于湖南大学史才军教授团队提出的“水分调控—气体迁移—碳化反应”的协同作用，成功解决了CO_2难以快速渗透混凝土内部进行有效反应的科学难题.利用水泥窑烟气中CO_2生产的低碳混凝土砖平均抗压强度超过15 MPa.年产的1亿块砖可捕集矿化约2.6万t CO_2，节约的余热蒸汽可发电379万kW·h(图1(a)).

2022年4月，中国煤化工行业万吨级CO_2矿化制备全固废负碳建材项目成功试运行.该项目由同济大学蒋正武教授团队提供核心技术，利用煤化工产生的气化渣、电石渣、粉煤灰等固废，与CO_2在无外热源、无水泥的情况下进行矿化反应，生产出高强度、高固碳率的矿化建材产品(图1(b)).

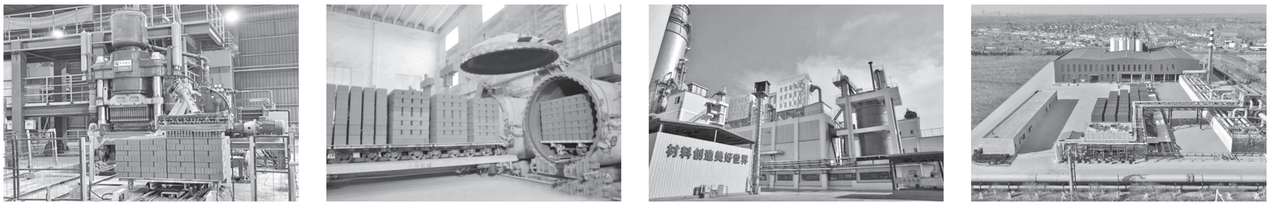
2022年11月，依托南京工业大学莫立武教授团队的核心技术，钢渣捕集水泥窑烟气中的CO_2制备固碳辅助性胶凝材料与低碳水泥生产线在济源中联水泥有限公司竣工投产.该生产线每年不仅能够直接捕集1.6万t水泥窑烟气中的CO_2，还能资源化利用钢渣生产固碳辅助性胶凝材料30万t，综合减碳达25万t(图1(c)).

2023年1月，京博控股集团与武汉理工大学硅酸盐建筑材料国家重点实验室合作，成功建设并投产了万吨级直接利用CO_2的CCUS示范项目.该项目直接利用CO_2含量大于等于10%的工业尾气，并协同处理多种工业固废.相关示范项目已实现年捕集利用与封存6万t CO_2，消纳16万t固废，并产出400万m^2的负碳新材料(图1(d)).

2023年7月，碳化法钢铁渣综合利用一期二阶段10万t示范产业化项目迈入试生产阶段，已产出首批$CaCO_3$产品.与传统技术相比，该项目采用的碳化法省去了焙烧工艺，减少了CO_2的释放，并且能够直接利用CO_2作为原料，具有双重减碳效果(图1(e)).

同样在2023年7月，中国大规模商业化应用固碳预拌混凝土的项目在浙江省湖州安吉开发区的国家电网零碳变电站成功实施.示范基地的生产线年产能达80万m^3固碳预拌混凝土，每年可封存约3 000 t CO_2，与传统工艺相比，能够减少近70 kg/m^3的CO_2排放(图1(f)).

国外也有利用加速碳化技术生产低碳建筑材料的工业化应用案例，如CarbonCure公司(图1(g))和O.C.O Technology公司(图1(h)).



(a) CO_2 cured concrete brick　(b) Production line for producing negative carbon building materials with 10 000 t CO_2 mineralization　(c) Carbon capture supplementary cementitious materials and low-carbon cement production line　(d) CCUS demonstration project for direct utilization of 10 000 t CO_2

(e) Demonstration project for comprehensive utilization of carbonated steel slag　(f) Project of premixed concrete with carbon capture　(g) Application case of premixed concrete with carbon capture by CarbonCure company　(h) Application case of carbonated artificial aggregates by O.C.O Technology company

图1　低碳建筑材料的工业化生产和应用案例

Fig. 1　Industrial production and application cases of low-carbon construction materials

5 结语

随着全球对可持续性和环境友好型材料需求的不断增长,使用冶金固废钢渣捕集矿化CO_2并制备建筑材料,已成为水泥和混凝土制造业中碳捕获、利用与封存(CCUS)技术的研究新热点和新趋势,展现了其独特的价值和应用潜力.

(1)通过特定设备将钢渣、水及其他掺合料按比例混合、成型并进行碳化处理,可制备出适用于不同场景的建筑制品,为大规模消纳钢渣同时捕集矿化CO_2提供了有效途径.建筑制品的CO_2吸收量对其性能起着至关重要的作用,而建筑制品尺寸一般较大,现有研究尚未完全释放其碳矿化潜力.因此,迫切需要有效的方法来提高碳化钢渣建筑制品的CO_2吸收量.此外,评估制备成本及效益对于推广碳化钢渣建筑制品的工业化应用也至关重要.

(2)对于钢渣辅助性胶凝材料,碳化不仅能够有效解决其安定性不良问题,还能提高水化活性.从当前的研究来看,高CO_2含量、高液固比的碳化条件能使钢渣具有较高的CO_2吸收量,但条件要求相对较高,且后续干燥任务重.比较而言,低CO_2含量、低液固比条件下钢渣辅助性胶凝材料的碳化更贴近生产实际.后续的研究应重点关注不同工况条件下钢渣辅助性胶凝材料的加速碳化及其与水泥基材料的相容性等问题.

(3)碳化钢渣制品或辅助性胶凝材料作为一种新兴的低碳建筑材料,在水泥和混凝土制造业中的应用前景十分广阔.通过持续的研究和创新,结合政府提供的政策支持,如财政补贴、税收优惠、制定行业标准、推广示范项目,以及加强国际合作等.有理由相信,碳化钢渣将为推动水泥和混凝土制造业的绿色转型和可持续发展作出重要贡献.

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