

Research Progress of Low-Carbon Cementitious Materials Based on Synergistic Industrial Wastes

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Abstract: Cementitious material based on synergistic industrial wastes can be used as a new product for low-carbon transformation. It can aid in resource recycling and suitable consumption and utilisation of various industrial wastes. The proposed material can reduce a large amount of CO₂ emitted during calcination in cement production and the decomposition of raw limestone. In addition, the material exhibits high durability and high resistance to corrosion in the marine environment that can further reduce CO₂ emissions over the lifecycle of the carbon footprint of the building. Currently, many similar chemical kinetic processes and mineralogical reaction processes of particle migration and rebinding exist in the hydration and hardening reactions, service processes and durability evolution of different industrial waste cementitious systems for low-carbon production. The theoretical basis of preparing various low-carbon cementitious materials (LCCMs) with industrial waste systems is discussed herein, including the two theories of ‘complex salt effect’ and ‘isomorphic effect from tetrahedral coordination of silicon-oxygen’. Further research on LCCM is based on the theoretical foundation of ‘passive hydration kinetics’. Furthermore, this study presents the CO₂ reduction potential of LCCM prepared using industrial wastes and provides future research directions in this regard.



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1. Introduction

For nearly half a century, CO₂ emissions have garnered much global attention and have become the common challenge of the world. China has made a clear commitment to strive to peak CO₂ emissions before 2030 and achieve carbon neutrality before 2060. China’s goals to reduce CO₂ emissions are an important part of its efforts to achieve internal sustainable development and build a global community of shared future.

The traditional building materials industry, especially the cement and concrete industry, is a major carbon emitter and faces a big challenge of CO₂ emission reduction, which has become an opportunity to promote transformation and upgrading of the industry and achieve green development. A series of complex interrelated issues such as basic theories, technical foundations, application technologies and protocols, and socio-cultural and political-economic constraints must be clearly addressed to alleviate CO₂ emission in the building materials industry. The cement manufacturing process is a highly energy-consuming and emission intensive process. Globally, cement production accounts for 5% to 7% of global anthropogenic CO₂ emissions. Being the world’s largest cement producer [1–3], China produces and consumes more than half of the world’s total cement production [4]. In 2020, China produced 2.39 billion tons of cement [5] and consumed more than 1.8 billion tons of cement clinker, emitting nearly 1.6 billion tons of CO₂ and a large

amount of other air pollutants such as sulphide and nitrogen oxides. Thus, it is necessary for China to significantly decrease CO₂ emissions of the cement industry to be in line with its commitment to combat climate change.

The rapid development of industrialisation in China has increased the production of industrial wastes in the country as well. According to statistics from the Ministry of Ecology and Environment, China generated 3.68 billion tons of general industrial wastes in 2020 (waste rock not counted), with a comprehensive utilisation rate of less than 60% [6]. Massive stockpiles of industrial waste take up land resources, pose environmental threats and cause social problems. Thus, comprehensive utilisation of industrial waste is of great significance to ecological civilisation and sustainable socio-economic development. Scientists around the world have developed a keen interest in producing low carbon cementitious material (LCCM) with industrial wastes. Various typical bulk industrial wastes, including granulated blast-furnace slag (GBFS), steel slag (SS), fly ash (FA), self-combusted coal gangue, coal-to-liquid slag, oil shale residue, desulphurisation ash, flue gas desulphurised gypsum (FGDG) and various kinds of industrial by-product gypsum, have been used to produce LCCM through their synergistic reaction. The preparation of LCCM, which can replace cement, using industrial wastes does not require calcination of limestone and has low energy consumption, and, thus, has a high potential for carbon reduction. In this regard, many research and application cases have reported on industrial waste-based LCCM without cement clinker [7–15].

The present paper summarises and analyses the current research progress of LCCM with industrial wastes in China and globally. LCCMs of interest include SS-GBFS-FGDG cementitious material system, FA-FGDG-less clinker cementitious material system, super-sulphated cement (SSC) and complex cementitious material used for underground cemented backfill mining. The strength performance, durability, heavy metal curing ability and hydration and hardening mechanism of each cementitious system are explored. This paper aims to summarise the core theoretical support, technical basis, and future R&D direction of LCCM through extensive summary and in-depth analysis of data from research, production and application sectors, in combination with the past 20 years of experience of the author's research team. An outlook on the CO₂ reduction potential of industrial waste-based LCCM in the cement and concrete industry is also provided.

2. Current Status of Research

2.1. Steel Slag-Granulated Blast Furnace Slag-Flue Gas Desulphurised Gypsum (SS-GBFS-FGDG) Cementitious Material

In China, SS emissions account for about 8% to 15% of crude steel production [16,17]. The annual SS emissions in China exceed 100 million tons, but its utilisation rate is only about 30% [18]. In recent years, China has increased its R&D efforts on the resource utilisation of SS [19]. In this regard, many scholars have made SS into cementitious materials to consume SS on a large scale and realise its high value-added utilisation. This has also broadened the direction for the development of industrial waste-based LCCM.

Qingjun [20] studied the mechanical properties, wear resistance and hydration products of cementitious materials prepared using SS and GBFS as the main raw materials and a small amount of cement clinker as a chemical activator. Hong et al. [21] prepared SS concrete using finely ground SS powder as a substitution cement. They found that the resistance of concrete decreased with the increase in SS powder substitution, e.g., when 40% of cement mass was replaced with SS powder, the resistance of the concrete decreased up to 46.2%. Zhang [7] addressed the problem of slow hydration and hardening rate of SS-GBFS-barium slag composite cementitious materials by adding crystal seeds to accelerate the hydration and hardening process. They prepared the composite crystalline species through rapid pre-hydration of the cementitious material. Elahi Manjur and Ramyar [22,23] showed that, in the preparation of cementitious materials, when some cement is replaced with industrial wastes, such as low-calcium FA, GBFS powder and SS powder, the pore structure of the prepared hardened concrete is improved. This improvement in pore structure is due

to better particle filling and volcanic ash reaction, which leads to a denser microstructure and reduced susceptibility to external sulphate penetration, i.e., improved sulphate resistance. The above-mentioned studies prove that cementitious materials can be prepared using small amounts of cement clinker with industrial wastes, such as GBFS, barium slag (BS) and gypsum. The prepared concrete shows a positive correlation in the direction of resistance to sulphate attack and electrical conductivity and can be used as a substitute for ordinary Portland cement (OPC) in terms of strength properties.

Many in-depth studies have been conducted on all-industrial waste cementitious materials (ASWCMs) to further reduce the amount of cement clinker and improve the utilisation of industrial wastes. Several studies have adopted the technical route of industrial waste synergistic hydration reaction and have proved that the raw materials in the ASWCM system can synergistically promote the hydration process.

Junzhong et al. [8] used SS powder, GBFS powder, FGDG powder and mine tailing powder to prepare ASWCM. They used ASWCM and aggregates made of mine tailings and waste rocks to develop ready-mixed pumped high-performance concrete of C35, C40, C45, C50, C55 and C60 strengths for various construction projects. Xiaowei [9] used steel slag, slag and desulphurised gypsum to prepare cementitious materials. They used aggregates made of iron tailings and waste rock to prepare high-strength and high-performance concrete with good flowability and excellent durability. Ligu [24] formulated clinker-free SS cement with SS and FGDG as the main raw materials, mixed with an appropriate amount of GBFS and a small amount of activator. They reported that the prepared clinker-free slag cement reached the technical requirements of PC42.5. Xuan [10] et al. reported that the contribution of SS powder to the compressive strength growth of concrete at early stage is less than that of GBFS in SS-GBFS-FGDG cementitious material. They also reported that the compressive strength of concrete gradually increases with the increase in specific surface area of SS. Xiaosheng [10] et al. used SS, GBFS and FGDG as raw materials to prepare the solid foam concrete by physical foaming method. Dong et al. [12] added alkali slag (AS) to GBFS-SS-FGDG ternary cementitious material to develop a new quaternary cementitious material. They proved that the addition of AS could promote the dissociation of glass phase in GBFS, which can solve the problems of slow setting time and low early compressive strength of GBFS-SS-FGDG ternary cementitious material.

Chengwen et al. [13] prepared all-industrial wastes marine concrete with AS, SS, GBFS and FGDG as cementing materials and iron ore tailings and waste rock as aggregates. They showed that the chloride ions introduced by AS produced Friedel salts in the hydration process, and the all-industrial waste marine concrete exhibited excellent mechanical properties and resistance to chloride ion attack. Wen et al. [14] prepared ASWCM using electric furnace reduction slag with high alkalinity instead of converter SS. The soundness of the cementitious material was in accordance with national standards. The synergistic hydration of the cementitious material resulted in very low-solubility Aft-like complex salt and amorphous calcium silicate gels (C-S-H). Huihui et al. [15] prepared ASWCM using high titanium GBFS, SS and FGDG. They reported that the 28-day compressive strength of the prepared concrete specimens reached more than 40 MPa.

LCCM is made of basic components, such as GBFS, SS, industrial by-product gypsum and supplementary materials, such as FA and iron tailings. LCCM with hydration hardening properties is prepared via the synergistic hydration reaction between divalent metal oxides and hydroxides in SS, FGDG, GBFS, FA and tailings powder in the aqueous solution. Mixing the above-mentioned cementitious materials with aggregates processed from iron tailings and waste rocks can form full industrial waste high-performance concrete, which has good constructability, volume stability and durability, and is fully suitable for engineering construction.

In 2019, the Housing and Urban-Rural Development Department of Hebei Province in China issued the specifications for acceptance of constructional quality of low CO₂ cementitious material high-performance concrete structures. [25] In 2020, the China Engineering Construction Standardisation Association formulated the Technical Regulations for the

Application of Solid Waste-based Cementitious Materials [26]. In 2021, the Housing and Urban-Rural Development Department of Hebei Province in China issued the Technical Standards for the Application of All Solid Waste High-Performance Concrete [27]. Under the support of these series of standard systems, China had poured more than 5 million m³ of ASWCM concrete by the end of 2021 and successfully applied it to a small number of housing structure projects. The country also successfully produced a large number of housing facade envelope materials, pipe piles and large municipal concrete pipes.

The above studies used industrial wastes as the main raw material to prepare cementitious materials, proving that cementitious materials prepared without cement clinker can be used to replace OPC in most applications.

2.2. Fly Ash-Flue Gas Desulphurised Gypsum-Less Clinker Material System (Low Calcium to Silicon Ratio Cementitious Material System)

In the 1930s, a group of scholars began to study the properties and performance of FA due to the rapid increase in its production worldwide. In 1935, the American scholar R. E. Davis (hereafter referred to as Davis) first used FA to replace part of the Portland cement to create a new type of concrete: FA concrete [28]. During the period of 1948–1953, the FA was first applied to dam projects in the United States on a large scale. Qibiao [29] produced a new wall material using gypsum, cement and FA, and this new material exhibited good water resistance, high strength and low drying shrinkage. The wall material showed good strength performance and softening coefficient when the material included 20% cement. Yong et al. [30] prepared cementitious materials using FA, FGDG and OPC. They reported that the strength value of cementitious material can reach the maximum value at any maintenance age when FGDG is 20% and the ratio of FA to cement is 1:1.

The study of cementitious material systems containing large amounts of FA or other volcanic ash-like substances, high sulphate and cement clinker-less cementitious material systems and the application of these materials in concrete began in the early 21st century. Wen et al. [31] prepared cementitious materials using finely ground GBFS, cement clinker, FGDG and FA. The cementitious material was mixed with fine aggregates and coarse aggregates to prepare high-strength precast concrete parts that showed a compressive strength of 89.8 MPa. After 28 days of curing at room temperature. Zhong et al. [32] used 20% FGDG, 20% GGBFS and 60% FA to prepare cementitious material, and the mortar made from this cementitious material exhibited compressive and flexural strengths of 22.6 MPa and 4.3 MPa, respectively, at 28 days of curing. Fusheng et al. [33] made cementitious material from red brick powder, cement and gypsum. Red brick powder is ground from waste red bricks used in urban construction with similar main composition to FA. The maximum strength of concrete prepared with 50% of brick powder can reach the requirement of C50 concrete when cured for 28 days.

Guoping et al. [34] prepared a ready-mixed pumped concrete with excellent performance by mixing cementitious material (FA, cement and FGDG) and aggregates. They reported that the concrete at 28-day curing exhibited a strength of 35–61 MPa. Huaqiang et al. [35] concluded that highly reactive circulating fluidised bed (CFB) ash with high alumina and suitable SO₃ content can be introduced to replace GBFS in the preparation of SSC. The percentage of CFB combustion ash can be increased to 70%, with corresponding clinker and added hard gypsum contents of 20% and 10%, respectively. They also demonstrated that negligible destructive swelling was caused by calcium aluminate formation when hyper-sulphate cements were prepared using highly reactive CFB combustion ash. The amount of FA blended in the crushed concrete used in the Three Gorges water conservancy project dam in China can reach up to 50% of the cementitious material [36]. The hydrated C–S–H in cement concrete can be categorised into two types according to the calcium-silica ratio. C–S–H gels (I) have a calcium-silica ratio below 1.5 and C–S–H (II) have a calcium-silica ratio above 1.5 [37]. Nonat and Lecoq further categorised the first category into two types. C–S–H (a) indicates $C/S < 1.0$ and C–S–H (b) indicates $1 < C/S < 1.5$ [38]. When the C–S–H gel has $C/S > 1.5$, it is in a stable state in a saturated solution

of calcium hydroxide. When the C/S of C–S–H gel is <1.5 , its stable existence does not need to be in calcium hydroxide saturated solution [39]. Moreover, the lower the calcium to silicon ratio in the above C–S–H gel phase, the greater the contribution of the same mass of hydration products to the strength of cement concrete [40].

The above discussion shows that, in the FA-FGDG-less clinker cementitious material system, the industrial wastes, such as FA and FGDG, from the coal power industry are used as the main raw materials. Here, the passive hydration kinetics is based on the complex salt effect, which provides the driving force for the main reaction of hydration and hardening of the system. The use of FA-FGDG-less clinker cementitious material, which uses more than 50% less cement clinker than conventional cement, can reduce CO₂ emissions by more than 50% compared to conventional cement concrete systems.

2.3. Granulated Blast Furnace Slag-Flue Gas Desulphurised Gypsum-Less Clinker Material System (SSC)

As early as 1908, Hans Kühl in Germany discovered that sulphate could be used as an activator to excite GBFS, inspiring research on the use of sulphate to activate GBFS hydration [41–45]. The GBFS-gypsum-cement system, also known as SSC, is clinker-less cement with GBFS as the main raw material, gypsum as the sulphate activator and clinker or lime as the alkaline activator. In 1920, SSC was first used in France and Belgium. Between 1940 and 1945, it was produced and used on a large scale, especially in Germany, due to resource constraints caused by the war. Later, SSC was reported to have excellent durability and resistance to chemical attack. After extensive research, SSC was officially licensed by Austria in 2002 for sale in the local construction materials market. In recent years, the UK, France, EU, Germany and other European countries have started to revise or develop corresponding standards to create conditions for the application of SSCs. In China, research on SSC started as early as 1956, and SSC was, thus, produced and applied on a large scale. Later, SSC was found to have low early strength and poor carbonation resistance, which led to surface ashing, poor weathering resistance and poor frost resistance. In the 1990s, Mingkai et al. developed a special cement for pavement base materials using GBFS, industrial by-product gypsum and a small amount of alkaline activator. However, the development of SSCs is still in the research stage [46].

In general, SSC is composed of 75–85% GBFS, 10–20% sulphate (dihydrate gypsum, gypsum, phosphogypsum, etc.) and 1–5% alkali activator (cement clinker, calcium hydroxide, lime, etc.) [47–55]. SSC exhibits good sulphate attack resistance and excellent durability due to its main hydration products of ettringite and C–S–H gel [56,57]. Thus, SSC can be applied to mass concrete works, road concrete works and concrete works in complex sulphate attack environment [58]. Guo et al. [59] produced SSC with 28-day strength higher than that of the P.O42.5 benchmark cement by mixing 7.5% cement clinker with 15% gypsum powder and 77.5% commercial GGBFS. They reported that the SSC exhibited lower water requirement for normal consistency, longer setting time and better soundness. Its 28-day strength was even higher than that of OPC. Zhang et al. [58] prepared a type of FGDG-based SSC using GBFS, FGDG and SS as the main components, adding a small amount of FA, cement and compound exciter. They reported that the 28-day compressive strength of the prepared SSC was more than 60 MPa. Zhao [60] developed a type of super-sulphur hydraulic cementing material with FGDG as the main raw material (up to 45%), GGBFS as active ingredient, and clinker and SS as alkaline activator. When activated by 8% SS, the 28-day compressive strength of the cementitious material can reach 50.7 MPa.

In a study of the existing SSC buildings, the Finger-Institute for Building Materials Engineering of Bauhaus University in Weimar, Germany, found that SSC has excellent durability and resistance to chemical attack [61]. Although GBFS-based SSC is considered a reliable alternative to Portland cement, the production of GBFS is reportedly insufficient to meet the needs of the cement industry [62].

2.4. All-Industrial Waste Cemented Filling Material for Underground Mining and Industrial Waste-Based Cementing Material for Filling

In addition to the preparation of cementitious materials, such as SS, GBFS and FA, from industrial wastes with potential cementitious activity possibly meeting the requirements of mine filling materials, they can also be partially or completely substituted for cement to prepare cemented filling materials and filling cementitious materials.

Hu et al. [63] prepared cementitious material for cementitious filling with fine tailing sludge of SS after wet grinding and wet separation of iron micronised, mixed with 12% desulphurisation gypsum and 25% slag. They reported that the fluidity of slurry meets the fluidity requirement of self-flow cemented backfill, and the compressive strength meets the mine filling strength requirement. Zhang et al. [64] prepared the low-carbon and clinker-free cementing material for backfill of underground cemented infill mining using SS, BGFS and FGDG, which is of great significance for infill mining and consumption of tailings. Liu et al. [65] prepared filling slurry with 40% SS, 6% lime, 12% FGDG and 42% GBFS as cementing materials, with 28-day strength of 4.03 MPa. Ren et al. [66] prepared cementing material with 55% SS tailings, 30% GBFS and 15% FGDG. They reported that the 28-day compressive strength of the prepared filling material can reach 4.78 MPa, meeting the requirements of mine filling material performance. They also found that the amount of hydration products in the system increases obviously with the increase in hydration age.

Yan et al. [67] took lead-zinc mine tailings as the research object and prepared paste filling material with all-industrial waste cementing material. They reported that the fluidity of filling slurry is 300 mm when GBFS, SS, FGDG and $\text{Ca}(\text{OH})_2$ were used; the fluidity meets the requirements of paste filling gravity flow transportation. They also found that the compressive strength of the solidified body maintained at 40 °C for 28 days is 20.19 MPa, which meets the requirements of cemented fill mining for compressive strength of the filling body. In their study, the leaching concentrations of arsenic and all other heavy metals in the tailings reach the limit value stipulated by the drinking water of the People's Republic of China. Guo et al. [68] used SS, GBFS and FGDG to prepare cementing material to solidify heavy metal tailings. They reported that the mass of cementing material is 9% of the mass of the treated heavy metal tailings when the composition 60% SS, 20% GBFS and 20% FGDG is used in the cementing material. The 3-day strength reaches 1.2 MPa; the fluidity is large, which is suitable for gravity filling; and the product strength is high. The leaching results of heavy metals are as follows: lead ≤ 0.01 mg/L and zinc ≤ 1 mg/L, both of which can meet the pollutant discharge standard (GB18918-2002). Yan et al. [69] used SS, GBFS and FGDG to prepare paste filling cementing material for lead-containing tailings. The strength of this material met the requirements of general filling body, and the fluidity reached the pumpability index. Moreover, the all-industrial waste filling material exhibited higher ability to solidify heavy metal lead than that of filling material prepared from cement and tailings.

Zhang et al. [70] used CFB fly ashes, GBFS and FGDG to prepare all-industrial waste cementitious material and then used this material and metal-containing tailings to prepare underground mining cemented fill. In addition, they also reported that the compressive strength of the filling materials after 3-day curing exceeds 1 MPa, and the leaching levels of target heavy metal elements (Pb, Zn, Cd, Hg, Cr and As) when the content of FGDG is 12% and the content of CFB fly ashes is 20%, 40% or 60%. Soluble salts (Cl^- and SO_4^{2-}) and dioxins are lower than the limit value of underground III water quality standard and are harmless to the environment. Li et al. [71] prepared clinker-free binder with refining slag, SS, GBFS and FGDG to meet the filling requirements of mines in alpine and high-altitude areas. Moreover, after 28 days of curing at 5 °C, the prepared binder meets the performance requirement of the filling material and greatly reduces the filling cost. At present, the development trend in cemented filling technology is to prepare binders with better working performance and lower production cost. The preparation of filling materials with industrial wastes not only meets the above conditions, but also has important

significance for CO₂ emission reduction and collaborative solidification of heavy metals. Thus, this is the most effective way to realise waste-free mining and create green mines.

3. Analysis and Discussion

3.1. Steel Slag-Granulated Blast Furnace Slag-Flue Gas Desulphurised Gypsum (SS-GBFS-FGDG) Cementitious Material

GBFS is mainly composed of SiO₂ and CaO, as well as a small part Al₂O₃ and MgO. It has good pozzolanic activity because more than 80% of the glass phase is generated by quenching during discharge [72,73]. However, it comprises almost no active chemical reaction potential energy. Therefore, it is necessary to drive the reaction kinetics under certain conditions to achieve further hydration [74]. The hydration potential of GBFS cannot be fully developed by chemical activation methods, such as alkali activation (lime, cement clinker) and sulphate activation (gypsum). In addition, the most common activation methods for GBFS are mechanical activation (increasing the surface area of materials through mechanical grinding) and thermal activation (increasing the hydration temperature of GBFS) [74–76]. The process of preparing cementitious materials with industrial wastes, such as SS, FA and FGDG, also includes synergetic hydration reactions and passive hydration kinetics based on the theory of complex salt effect.

SS is the second-largest industrial waste of the iron and steel industry in China. Hence, there is an urgency to determine effective methods for large-scale consumption of SS. Nevertheless, there are challenges with respect to its utilisation. This is because f-CaO and MgO in SS expand in volume after hydration, resulting in serious stability problems [77–79]. The existing SS-GBFS-FGDG system can use a large amount of SS because the poor stability of SS can be greatly improved through grinding. Sun explains that, after the SS is ground to a certain fineness, the free CaO and MgO are activated and react with water to generate Ca(OH)₂ and Mg(OH)₂ at the early stage of cement hydration. Although the volume of the reaction product is larger than that of the reactant, the material is still in the plastic state at this time and will, thus, not cause any structural damage [80]. Li believes that the dispersion degree of free CaO and free MgO was greatly improved after the SS underwent superfine grinding, resulting in the effective control of the stability problem. Meanwhile, promoting the formation of more ettringite and increasing the hydration reaction rate of GBFS before concrete hardening can also inhibit the poor volume stability of SS [81].

No free Ca(OH)₂ or Mg(OH)₂ is found in the hardened body of cementitious material hydration for several years (the longest observation time of the sample has been nearly 10 years) in the case of low proportion of SS in the system ($\leq 30\%$ for example) and high reactivity of GBFS. The pH values of hundreds of short-term and long-term hardened powders were also monitored by the author. The results also showed that the pH values of saturated solutions of the samples were all below 12.5, while most of them were below 11.8, and the saturated pH value of Ca(OH)₂ was about 12.8 (with slight fluctuation with temperature change). A recent study also proves that magnesium ions in the solution enter the hydrated silicate products preferentially compared to calcium ions during the hydration and hardening process of the cementitious materials of the system. The research in the last ten years shows that there is no chemical reaction process in the industrial waste-based LCCM system without cement clinker and that free CaO and free MgO are converted into solid Ca(OH)₂ and Mg(OH)₂, respectively, when the proportion of SS is less than 30%. Therefore, the problem of stability of an ordinary cement concrete system caused by SS can be completely avoided in the above industrial waste synergistic clinker-free binder system.

In the past ten years, the monitoring results of the research team of the author on the volume change law of concrete show that the concrete prepared by the magnetite-quartzite sand and gravel and SS-GBFS-FGDG system cementitious material is always in a micro-shrinkage state, when the content of SS in the cementitious material is less than 35%. When the content of SS in the cementitious material reaches 60%, the concrete sample prepared two years ago does not expand, and the strength continues to increase. Several research results show that the specific surface area of pre-grinding SS increases, the hydration rate is

accelerated, the activity index is also improved, and the bending strength and compression strength of cementitious materials containing SS also improve in varying degrees [82–89].

Solid waste-based cementitious materials have been aimed at preparing high-performance concrete since their inception. In high-performance concrete, the proportion of harmful pores in the hardened body and the boundary between cement and aggregate approach zero. However, this does not mean that all the porosities in concrete approach zero. The cementing matrix part of the high-performance concrete hardened body prepared by the SS-GBFS-FGDG cementitious material contain 15–20 vol% porosity. This is based on the difference between the true density of various minerals and the apparent density of the concrete, and the difference between the total water consumption in the concrete preparation process and the total amount of newly added chemical bonding water of the formed concrete cementing matrix hardened body. In addition to millimetre-level bubbles left in the concrete pouring and vibrating processes, the pores are basically submicron and nanometre pores. The air bubbles and the micro-nanometre pores are uniformly distributed in the cementing matrix. If a small amount of micron-sized SS remains in that hardened body, the expansion amount of the micron-sized SS can be totally absorbed by nearby micron-sized harmless pores and millimetre-sized bubbles in the concrete gel material. Thus, this indicates that the structural performance or the material mechanical performance of the concrete cannot be damaged.

The ‘complex salt effect’ and ‘isomorphism effect from tetrahedral coordination of silicon-oxygen’ are important theoretical bases for the hydration process of LCCM. Zhang et al. [90] proposed the preliminary concept of the complex salt effect in 2011. The solubility product constant of ettringite is $10^{-111.6}$ [14], which makes it easy to crystallise ettringite, continuously consuming even a small amount of soluble aluminium ($\text{Al}(\text{OH})_2^+$, $\text{Al}(\text{OH})_2^+$, AlO_3^{3-} , HAlO_3^{2-} , H_2AlO_3^- , AlO_2^-) under saturated $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, Ca^{2+} concentration and a pH value that is close to SS. Consequently, the original balance between the aluminium-oxygen tetrahedron and the solution is broken, and the aluminium-oxygen tetrahedron is continuously and rapidly transferred to the interior of the ettringite crystal, taking the solution as the medium for the ferrosilicon-manganese alloy powder (the same applies to the GBFS [91,92]). In silicon-manganese-iron alloy slag (or GBFS) micropowder, almost 100% of aluminium exists in the form of aluminium-oxygen tetrahedron, which is connected with silicon-oxygen tetrahedron. When a large amount of aluminium-oxygen tetrahedron is separated from the chain or frame-shaped silicon-oxygen tetrahedron in the waste residue, the residual silicon-oxygen tetrahedron becomes a silicate anion group that is mutually connected by a 2–3 silicon-oxygen tetrahedron. These anionic groups will quickly combine with Ca^{2+} , OH^- and water in the solution to form a hydrated calcium silicate gel. These silicate anion groups can reconnect with some of the Al–O tetrahedra when the concentration of Ca^{2+} and OH^- is low and the concentration of soluble Al is high. For this, the silicate anion groups bridge oxygen to form a ‘zeolite-like phase’ containing hydroxyl and pore water. The process of reconnecting the Si–O tetrahedron with bridging oxygen and bringing soluble aluminium into the Si–O tetrahedron in the form of four oxygen coordination is called ‘isomorphism effect from tetrahedral coordination of silicon-oxygen’.

The clinker-free all-industrial waste cementing material is established based on the research on the synergistic effect of multiple industrial wastes. It is mainly composed of the SS-GBFS-FGDG cementing material system, wherein the FGDG can stimulate the activity of the SS and the GBFS. In addition, the SS and the GBFS show a synergistic promotion effect during hydration and have a superposition effect of ‘1 + 1 > 2’ [93]. Many studies have shown that the main mineral phases of SS in China include the RO phase (solid solution of FeO, MgO, MnO and CaO), C_2S , C_3S , C_4AF (calcium ferroaluminate), Fe_2O_3 and a small amount of $\text{Ca}(\text{OH})_2$, f-CaO (free calcium oxide) and f-MgO (free magnesium oxide) [94–98]. In typical SS, the total content of divalent metal oxides CaO, MgO and FeO reaches 72.3%. Divalent metal cations in SS are easy to hydrate, making the system alkaline [94]. These metal cations activate GBFS and tailings to form C–S–H gel and capture Al_2O_3 and Fe_2O_3

in GBFS and tailings to form iron-containing ettringite complex salt minerals with the participation of gypsum.

FGDG provides a large amount of Ca^{2+} and SO_4^{2-} for the reaction of the SS-GBFS-FGDG cementing material system. The SS hydrates slowly to form C–S–H gel, Ca^{2+} and OH^- at the early stage, rapidly increasing the alkalinity of the slurry. In addition, a large amount of FGDG and alkaline environment promote the dissociation of Si (Al) O tetrahedron in GBFS, and a part of soluble Si (Al) reacts with Ca^{2+} in the system to form C–S–H gel and AFt and other hydration products. SS continues to hydrate to form a small amount of C–S–H gel and maintain the alkalinity of the paste with the increase in hydration age. The continuous hydration of GBFS absorbs Ca^{2+} , SO_4^{2-} and OH^- in the system, promoting the hydration of SS and the dissolution of OH^- and FGDG [9,91,99]. The needle-like crystals of hydration product AFt penetrate into C–S–H gel due to the continuous reaction of SS and the increasing hydration degree. This results in a more compact structure of the hardened paste and increases the compressive strength [100]. Under the synergistic effect of the SS micro powder, the FGDG micro powder and the GBFS micro powder, a large amount of needle-like complex salt crystals are formed, tightly wrapped by a large amount of formed near-amorphous C–S–H gel and zeolite-like phases. This leads to improved stability of the whole system.

3.2. Fly Ash-Flue Gas Desulphurised Gypsum-Less Clinker Material System (Low Calcium to Silicon Ratio Cementitious Material System)

With respect to the FA-FGDG-less clinker system, most of the researchers working on FA Portland cement and volcanic ash Portland cement have focused on improving the activity of FA or volcanic ash admixture, as well as optimising the performance of cement. In addition, people regard cement clinker powder as the material basis of ‘active hydration kinetics’ when they use non-cement clinker powder materials. This is because of the serious disconnection between cement standard and concrete standard (such as the optimised use of water reducer and the chemical reaction between cementing material and sand aggregate).

When calcium sulphate dihydrate substance (FGDG, natural gypsum, phosphogypsum, waste ceramic membrane gypsum, etc.) or anhydrous calcium sulphate substances (natural anhydrite, or anhydrous gypsum calcined at high temperature from industrial by-product) are added to a mixture of more than 60% of FA and a small amount of cement clinker and these are ground together, the main reaction process of the system undergoes essential changes. The finer the grinding fineness of the clinker is, the more intense the active hydration reaction is in the early stage (within tens of minutes) when the system meets water, although the ‘active hydration reaction’ of the ground cement clinker powder still exists. Nevertheless, the total amount of FA participating in the early passive hydration reaction can be greatly increased after ultrafine grinding. In addition to the optimisation and adjustment of the concrete admixture, the superposition effect of the active chemical reaction and the early passive hydration reaction enables the concrete considering the FA-FGDG low-clinker system as the cementing material to overcome the defect of slow growth of the early strength of the traditional ‘SSC’ concrete to a certain extent. This also ensures that the concrete of the system facilitates the reaction of a large amount of FA with the formed high-calcium C–S–H gel to continuously and rapidly convert into low-calcium C–S–H gel after hardening. Moreover, this passive hydration kinetic reaction continues for a long time. Therefore, the concrete prepared by the cementing material has a later-stage strength growth margin far exceeding that of the concrete of an OPC system. The one-year strength growth margin can reach 5–10 times that of the ordinary cement concrete. According to the analysis results of the microstructure and micro-area composition of the concrete prepared by the cementing material, free calcium hydroxide has disappeared completely in the hydration hardening body of the system in one year, and the ratio of Ca/Si in C–S–H gel continues to decrease. The results of multi-sample and multi-site micro-analysis show that the average ratio of Ca/Si in C–S–H gels decreased to less than 1.2. The short chain C–S–H

gels are transformed into long chain C–S–H gels, and the proportion of calcium-containing zeolite-like phase increases with time.

According to Kunther [40], hydration products with low Ca/Si ratio, such as long-chain C–S–H gel and zeolite-like gel, exhibited a strength of more than 3 times higher than that of high Ca/Si products, such as short-chain C–S–H gel to concrete strength. In addition, the new hydration products also increased with the extension of curing time. The long-term strength margin growth rate of hardened concrete of this cementitious material system is 5–10 times higher than that of OPC concrete, mainly due to the superposition effect of the transformation of C–S–H gel and the increase of the total amount of newly formed hydration products. The extensive application of the system concrete in construction projects can effectively reduce CO₂ emissions. Moreover, it can greatly improve the durability of concrete due to the substantial improvement of the compactness of concrete and the long-term spontaneous optimisation of the micro-structure at the atomic group level.

3.3. Granulated Blast Furnace Slag-Flue Gas Desulphurised Gypsum-Less Clinker Material System (SSC)

For over a century, many researchers have conducted exploratory research and have contributed to the subsequent large number of engineering applications of SSC, with the research work of Kühl et al. in Germany being one among the early studies [41]. There was a lack between theory and practice of high-performance concrete before the middle of the 20th century, especially before adding water reducing agent in concrete manufacturing. This led to failures in the application of SCC in practical concrete projects, although its laboratory research was relatively successful. The early SSC concrete has a high proportion of harmful holes due to its large water consumption per cubic metre. In addition, it involves the extensive curing method for traditional Portland cement concrete, which generally causes exfoliation of ash and sand on the concrete surface [101]. This is because CO₂ can easily migrate inward along a large number of harmful capillary pores; react with ettringite within a certain depth of the surface layer; generate calcium carbonate, gypsum dihydrate and aluminium hydroxide gel; and lose a lot of crystal water. This further results in volume reduction, formation of crack generation and pulverisation of the hardened layer on the concrete surface. Exfoliation of ash and sand can be mitigated to a certain extent by increasing the proportion of cement clinker and gypsum dihydrate appropriately and reducing the proportion of GBFS micro-powder. However, this method almost returns to the technical system of traditional GBFS cement.

From the end of the 1990s to the beginning of the 21st century, there was an upsurge in the research on SSC, and its applications reached a peak. The application of ultra-fine mineral admixtures in concrete also garnered much attention with the further development of the production and application of concrete water reducer technology. Thus, ‘high-performance concrete’ technology began to be widely used in EU member countries, the United States, Great Britain and their ‘protected countries’ or colonial countries around the world [102–108]. The transplantation of the ‘production and application technology of water reducing agent’ and ‘ultra-fine admixture technology’ in high-performance concrete technology to the research, production and massive application of SSC overcame the problem of exfoliation of ash and sand and also greatly optimised and improved the other properties of SSC concrete.

In the SSC system without cement clinker or with only a small amount of cement clinker (less than 5%), the calcium to silica ratio of the C–S–H gel formed is mostly below 1.2, or even lower than 1.0 because the mole ratio of (CaO + MgO)/(Al₂O₃ + SiO₂) of water quenched GBFS is about 1.0 [109]. Under the condition that the crystallised ettringite consumes a large amount of divalent ions (such as Ca²⁺ and Mg²⁺), long chain hydrated silicate products with a calcium to silica ratio less than 1.2 will be formed inevitably if the GBFS is fine enough and the hydration is sufficient. In addition, even zeolite-like phase with a good network structure will also be formed. These phases with low calcium to silica

ratio are conducive to the substantial improvement of the strength of hardened body and various durability indexes of SSC concrete [110].

After solving the original problem of USC that is largely used in structural concrete projects, the EU member states, the United States and other countries seriously worked towards CO₂ reduction at the end of the 20th century and the beginning of the 21st century. They established relatively perfect standards and specifications and completed numerous important structural projects [43].

However, most of the blast furnace ironmaking processes were eliminated because the Western developed countries entered the historical period of short process steelmaking with electric furnace. In addition, products such as S105 slag powder play an increasingly important role in the new concrete technology due to the matured processing technology of superfine GBFS. S105 slag powder applied in high-performance concrete relies on the application of high-efficiency water reducing agent technology and other related technologies. When S105 slag powder replaces about 50% of OPC in high-performance concrete, the strength of the concrete improves by more than 160%, and other durability indicators are also greatly improved and optimised. Therefore, the price of S105 slag powder is much higher than that of OPC in Western developed countries.

Since the beginning of the 21st century, the research on SSC has basically lost its economic and social significance. With this situation becoming serious, the application of SSC aimed at consuming a large amount of GBFS has been gradually withdrawn from the main market.

The activation of GBFS, secondary pozzolanic activity reaction and the damages of concrete due to secondary ettringite formation mainly reflect that the research idea of SSC is influenced and dominated by the traditional Portland cement theory system. Although it has been proved in practice that the assumed secondary ettringite in the SSC concrete causes little damage to structural concrete, its study outside the framework of the existing standard system has not received the attention it deserves. This is because of the lack of in-depth research on the basic theory of SSC and its application in concrete, the micro and ultra-micro hydration and hardening process, as well as the changes in micro composition, structure and corresponding performance during long-term service. The latest research progress and large-scale engineering applications of the 'passive hydration kinetics' with the 'complex salt effect' as the core involve the aforementioned 'SS-GBFS-FGDG' and 'FA-gypsum-less clinker' cementitious materials. Therefore, cementitious materials are regarded as the theoretical innovation and the pioneer of the practical application technology of the traditional 'SSC' system.

3.4. All-Industrial Waste Cemented Filling Material for Underground Mining and Industrial Waste-Based Cementing Material for Filling

The total amount of ore (including raw coal) mined underground in China exceeds 6,000,000,000 t/y [111]. The total volume of filling body will exceed 3,000,000,000 m³ if all goafs are back and filled using the cemented filling method, which theoretically requires over 300,000,000 t of cementitious materials. The application of LCCM with multiple industrial wastes coordination that do not contain or contain a small amount of cement clinker in cemented filling mines is gradually increasing. At present, the LCCM accounts for up to 50% of the cementitious agents (including OPC) that are used in all cementitious filling mines in China. However, over 80% of the underground goafs are still not treated via the cemented filling method due to the high procurement cost of cementitious agent (mainly OPC system and the GBFS system). Therefore, in the past 10 years, the research of cementitious materials used for cemented filling in underground mines in China has generally aimed at reducing costs and improving the performance of the filling body. Most of these studies involved the passive hydration kinetics theory and basic control technology, which is supported by the two core theories of 'complex salt effect' and 'isomorphic effect from tetrahedral coordination of silicon-oxygen'. The concepts of two basic theories, 'complex salt effect' and 'isomorphic effect from tetrahedral coordination of silicon-oxygen'

were compiled into the Mining and Metallurgical Engineering Volume of the Encyclopaedia of China (Third Edition) in 2014 and were officially published online in 2021 and printed in 2022 [112]. Zhu et al. [113] used red mud as the main raw material to prepare low-cost and high-performance cementitious filling materials for underground mining. They found that both ‘complex salt effect’ and ‘isomorphic effect from tetrahedral coordination of silicon-oxygen’ occurred simultaneously in this system, which mutually promoted the hydration, hardening and strength growth of cementitious materials. Fine silicate tailings can deeply participate in the passive hydration kinetic reaction with the core of ‘complex salt effect’ and ‘isomorphic effect from tetrahedral coordination of silicon-oxygen’, driven by reasonably designed industrial waste-based cementitious materials [114]. This can lead to the use of cheap raw materials (such as SS and desulphurisation ash) in the preparation of high-performance cementitious filling body. They could also act as the main raw materials to drive ultrafine tailing particles to participate in hydration and hardening reaction. Compared with OPC and GBFS cementitious materials, these pioneering and innovative research results led to 90% and 80% decrease, respectively, in the manufacturing costs of cementitious materials for the cemented filling mining method.

Using waste incineration fly ash (WIFA) as one of the main components in cementitious materials for the cemented filling method in underground mines has been a pioneering research direction in recent years [70,115]. The study of cementitious materials has three characteristics of general leading-edge research: hot spots, highlights and difficulties. Different studies have shown that heavy metals and dioxins in WIFA can be effectively solidified by the optimised design of industrial waste-based cementitious materials. The leaching concentrations of all toxic and harmful substances (including dioxin) in the solidified filling body containing less than 20% WIFA in the cementitious material meet the requirement in the drinking water standard of China [70,115]. The results of extreme condition tests show that the cementitious material contained a maximum of 60% WIFA when tailings collaborated and were optimised in the system for cemented filling in underground mine. According to the existing solidification mechanism research, the ‘complex salt effect’, ‘isomorphic effect from tetrahedral coordination of silicon-oxygen’ and their synergistic effect on the cemented filling body ensure the solidification/stabilisation of various toxic and harmful substances in the hardened cemented filling body and eliminated the pollution in the groundwater environment.

4. Outlook

4.1. CO₂ Emission Reduction Potential of Cement-Concrete Industry Chain in China

In China, the cement industry is the second largest CO₂ emission industry, following the power industry [116–118]. The cement production process includes high-temperature calcination, two stages of grinding and high-temperature decomposition of limestone, which emits a large amount of CO₂ [119–122]. The production of each ton of standard cement clinker emits about 0.78 tons of CO₂ into the atmosphere. In 2020, China produced 2.39 billion tons of OPC, accounting for 55% of global cement products [123,124].

It is known that the greater the amount of CO₂ emissions, the greater the potential for CO₂ reduction in this field. China produces about 300 million tons of GBFS every year. [125] The mole ratio of (SiO₂ + Al₂O₃)/(CaO + MgO) in GBFS and cement clinker are about 1.0 [126] and 0.3, respectively. Therefore, the potential contribution of GBFS to the silicon (aluminium) oxygen tetrahedron of concrete is about three times that of cement clinker. From among all kinds of industrial wastes produced worldwide, GBFS has the most potential for hydration and hardening. This can be fully exploited through the preparation and application of cementitious materials and the systematic optimisation of the whole industrial chain, based on the theoretical support of ‘complex salt effect’ and ‘isomorphic effect from tetrahedral coordination of silicon-oxygen’, and on the technical basis of passive hydration kinetics regulation of multiple industrial wastes. The contribution potential of various hydration products to the strength of concrete prepared using GBFS can reach over three times that using cement clinker. Therefore, if all the GBFS generated in China is

optimised and applied, each ton of GBFS could contribute nearly 3 tons of cement clinker to the strength of concrete, and 772 million tons of CO₂ emissions could be reduced.

Studies have either directly or indirectly discussed the problem of the formation of long-chain C–S–H gel and zeolite-like phase with low-calcium silica ratio in concrete or cemented filling body in underground mine. Kunther et al. [40] confirmed that the strength of C–S–H products with low calcium-to-silicon ratio is nearly three times higher than that of C–S–H products with high calcium-to-silicon ratio. C–S–H gel and zeolite-like phase in the hardened body with lower calcium-to-silica ratio than that produced by OPC can be observed in the ultra-sulphate cement system (widely used in structural engineering in the EU and the United States at the end of the 20th century). Multiple industrial wastes were used to coordinate the low-carbon cementitious material concrete system (rapidly developed in China in the past 10 years) and the all-industrial waste cementitious material system for cemented filling in underground mines (fast developing now). In addition to GBFS, other industrial wastes (such as FA, self-ignited coal gangue, oil shale ash, waste brick and tile powder, ultrafine silicate tailings, desulphurised ash and various industrial by-product gypsum) can participate in the hydration and hardening reaction of the corresponding system at a practical reaction rate under passive hydration kinetics. Therefore, the demand for cement clinker can be greatly reduced in the future cement–concrete industry chain and related applications through optimisation design of processing and application. This will also result in the formation of C–S–H gel with low calcium-to-silica ratio in concrete and filling materials and improve the application performance of hydration hardening body, with passive hydration dynamics as the main driving force. If the calcium-to-silicon ratio in the cementitious hardening body is reduced from the current 1.5–2.0 (average 1.75) to 1.5–0.8 (average 1.15), the demand for cement clinker can be reduced by 30–35%. In 2020, China consumed about 1.8 billion tons of cement clinker. By applying 30% as the cement clinker reduction ratio, 540 million tons of cement clinker can be reduced, or an equivalent of 420 million tons of CO₂ emission can be reduced. This value can be regarded as the minimum value of CO₂ emission reduction for cement-concrete industry chain transformation based on passive hydration kinetics technology. In other words, the total amount of expected CO₂ emission reduction in this field is 1.193 billion tons, taking into account the maximum expected CO₂ emission reduction value of non-clinker cementitious materials produced from the industrial wastes in iron and steel metallurgical plants (772 million tons CO₂), and the minimum CO₂ emission reduction value of the traditional cement concrete industry chain modified by passive hydration kinetics technology, which applies little clinker to collaborative utilisation of other industrial wastes (420 million tons CO₂).

4.2. Theory and Basic Preparation Technology in Cementitious Material

The ‘complex salt effect’ and the ‘isomorphic effect from tetrahedral coordination of silicon-oxygen’ are two important theoretical supports for understanding the migration of particles, the movement state of extranuclear electrons, the change laws of phase and microstructure, and the change laws of various properties during the preparation and application of the above-mentioned four types of LCCM with multiple industrial wastes. Future research should accumulate further test data and establish reaction mechanism models under different environments. The thermodynamic and kinetic parameters of the hydration and hardening reaction of LCCM coordinated by multiple industrial wastes should be analysed using multi-dimensional correlation analysis with the process parameters of the preparation process and the product detection index parameters. This analysis can further contribute to the theoretical understanding of the technical foundation.

There is still a need to accumulate the solubility product constants of different subclasses of chalcocite minerals under standard conditions so as to improve the ‘complex salt effect’, which is dominated by the crystallisation process of chalcocite minerals. In addition, the variation of each subclass of complex salt minerals with homogeneous substitution, temperature change, pH change and solution ion effect also needs to be explored.

In terms of ‘isomorphic effect from tetrahedral coordination of silicon-oxygen’, the focus of future theoretical research is to strengthen the boundary relationship between the residual core of raw material particles that are relatively rich in silicon/aluminium and the newly generated hydration products. It is essential to conduct micro-area observation and statistical analysis of micro-area composition, such as field emission scanning electron microscope, transmission electron microscope and atomic force microscope, to perform an in-depth study of the hydration hardening body of a large number of LCCMs. The obtained micro- and macro-analysis data can then be processed in multiple dimensions using the powerful data mining and statistical analysis functions of modern digital technology. Thus, the driving mechanism and the quantitative description and prediction method of space-time change laws of the ‘isomorphic effect from tetrahedral coordination of silicon-oxygen’ in LCCM could be gradually formed.

4.3. Basic Research on the Application Technology of 1

Research in the past 20 years has showed that the hydration and hardening bodies of the above four types of LCCMs with multiple industrial waste collaboration have good service durability and strong solidification ability of heavy metals. Many researchers have also examined the high-performance concrete prepared with cementitious materials of ‘SS-GBFS-FGDG’ and have found that its service durability in ordinary building structures is about 5 times that of OPC concrete of the same strength level. In a marine environment, its service durability is about 10 times that of OPC concrete with the same strength level. However, the concrete structure design departments of various industries in China are still unable to optimise the structural design of LCCM with multiple industrial wastes in various building structures due to the lack of concrete structure parameters under different structures and service conditions. In the field of ocean engineering, LCCMs with multiple industrial wastes have particularly excellent erosion resistance to marine environment. However, it cannot be widely used due to the lack of structural performance parameters for optimisation design. Therefore, the future main task of the basic research on the application of LCCM with multiple industrial wastes is to further examine the characteristics of structural parameters of LCCM with multiple industrial wastes in various structural concretes, and determine their microscopic changes in service under various extreme environments. In this way, their wide application could be promoted through adaptation of the optimised design schemes.

In summary, with respect to ‘carbon peaking and carbon neutralisation’, industrial waste recycling and green development of the cement-concrete industry chain have become two hot issues that need to be solved urgently [127]. As an important part of China’s green and low-carbon circular economy system, the comprehensive utilisation of bulk industrial waste is a long-term strategic policy to ensure the sustainable development of China’s industry [128,129]. The comprehensive utilisation efficiency of industrial resources can be effectively improved via research and application of LCCM with multiple industrial wastes. In addition, this research can also promote the overall green transformation of economic and social development and help achieve the carbon peaking and carbon neutrality goals as scheduled.

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