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The Use of Dicalcium Silicate (C_2S) Based Composites for Environmentally Friendly Applications

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ABSTRACT

Dicalcium silicate (C_2S, Ca_2SiO_4), commonly known as belite, has attracted significant research attention as a versatile material for environmentally sustainable applications. As the second most abundant mineral phase in ordinary Portland cement clinker, C_2S offers distinct environmental advantages over tricalcium silicate (C_3S), including lower limestone requirements, reduced clinkering temperatures (approximately 200 °C lower), and consequently diminished CO_2 emissions during production. This comprehensive review examines the crystal chemistry and polymorphism of C_2S, covering its five well-established polymorphs (α, α'H, α'L, β, and γ), their interconversion pathways, and the implications of each polymorph for specific applications. The review critically evaluates three principal domains of environmentally friendly application: (i) low-carbon cementitious binders and carbonation-cured construction materials, where C_2S serves both as a low-energy binder and as a CO_2 sink capable of sequestering atmospheric carbon dioxide; (ii) biomedical materials, where the bioactivity, biocompatibility, and osteoconductivity of C_2S-based composites enable applications in bone tissue engineering, dental restorations, and drug delivery; and (iii) industrial waste valorization and environmental remediation, where C_2S phases present in steel slags and other metallurgical by-products are exploited for CO_2 mineralization and heavy metal immobilization. Synthesis strategies—including solid-state reaction, sol–gel processing, hydrothermal synthesis, and combustion methods—are discussed with emphasis on their influence on polymorph selection and reactivity. Finally, current challenges and future research directions are outlined, highlighting the need for enhanced early-age hydration kinetics, scalable activation strategies, and integrated life-cycle assessment of C_2S-based systems.

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INTRODUCTION

The cement industry is one of the largest industrial emitters of carbon dioxide, responsible for approximately 7–8% of total global anthropogenic CO_2 emissions, amounting to over 2.1 billion tonnes annually [1,2]. The majority of these emissions arise from two sources: the calcination of limestone ($CaCO_3$) to produce the clinker precursor calcium oxide (CaO) and the combustion of fossil fuels required to achieve clinkering temperatures exceeding 1450 °C [3]. Against this backdrop of urgent decarbonization imperatives, dicalcium silicate (Ca_2SiO_4, abbreviated C_2S in cement chemistry notation where C = CaO and S = SiO_2), commonly known as belite, has emerged as a material of profound strategic importance for environmentally friendly applications across multiple sectors [4,5]. In ordinary Portland cement (OPC), C_2S constitutes 20–40% of the clinker by weight and is recognized as the mineral phase responsible for long-term strength development [6]. Compared to tricalcium silicate (C_3S, alite)—the dominant phase in OPC—C_2S requires significantly less limestone and lower synthesis temperatures for its formation. The production of C_2S-rich cements (often termed belite cements) requires only approximately 45% CaO compared to 70% CaO for conventional OPC, resulting in a roughly 30% reduction in process-related CO_2 emissions [7]. Furthermore, the clinkering temperature for belite-rich formulations is approximately 1200 °C, some 200 °C lower than conventional OPC, yielding additional reductions in fuel-derived emissions [8].

Beyond its role in cementitious systems, C_2S has demonstrated remarkable versatility. Its bioactivity and ability to release calcium and silicon ions make it an attractive bioceramic for bone tissue engineering and dental applications [9,10]. Its strong carbonation reactivity—particularly in the γ polymorph—enables direct sequestration of CO_2 into stable carbonate forms, transforming a construction material into an active carbon sink [11,12]. Additionally, C_2S is a major mineral phase in steel slags and other metallurgical by-products, and carbonation-based valorization of these wastes presents a dual environmental benefit: waste diversion from landfills and permanent CO_2 storage [13,14]. This review provides a comprehensive, cross-disciplinary examination of C_2S-based composites for environmentally friendly applications. It begins with the fundamental crystal chemistry and polymorphism of C_2S, proceeds through synthesis methods and activation strategies, and then critically evaluates three principal application domains: low-carbon construction materials, biomedical applications, and waste valorization for environmental remediation. The review concludes by identifying key challenges and promising future research directions.

LITERATURE REVIEW

Crystal Chemistry and Polymorphism of C_2S

2.1. Crystal Structure

Dicalcium silicate possesses an island (nesosilicate) crystal structure in which isolated [SiO_4] tetrahedra are linked through [CaO_x] polyhedra to form a three-dimensional framework [15,16]. The chemical formula Ca_2SiO_4 reflects a Ca/Si molar ratio of 2:1, with two distinct calcium coordination environments present in most polymorphs. This structural arrangement governs the material's reactivity with water, its carbonation behavior, and its interactions with biological media [17].

2.2. Polymorphic Transitions

C_2S exhibits five well-established crystallographic polymorphs, designated α, α′H, α′L, β, and γ, which undergo a series of reversible and irreversible transformations as a function of temperature [15,18]. The transition sequence upon cooling from the melt is: α (stable above ~1425 °C) → α′H (~1160 °C) → α′L (~680 °C) → β (~500 °C) → γ (below ~500 °C). The final β → γ transformation is reconstructive and accompanied by a volume increase of approximately 12%, which causes the well-known phenomenon of "dusting" or self-pulverization of clinker [19,20]. The hydraulic reactivity of C_2S polymorphs decreases in the order α > α′H > α′L > β >> γ. The β polymorph is the hydraulically active form present in cement clinker and is stabilized at ambient temperature by foreign ions in solid solution, including Al^{3+}, Fe^{3+}, Mg^{2+}, K^+, and anions such as SO_4^{2-} and PO_4^{3-} [15,21]. On the other hand, γ-C_2S is the thermodynamically stable polymorph at ambient conditions but is essentially non-hydraulic due to its highly ordered crystal structure and the coordination geometry of its calcium ions [22]. Interestingly, this very stability makes γ-C_2S the most reactive polymorph toward carbonation, with a carbonation activity approximately twice that of β-C_2S [23,24].

Table 1.

Summary of C_2S polymorphs and their key properties.

Polymorph	Crystal System	Stability Range	Ca Coord. Number	Hydraulic Activity	Carbonation Reactivity
α	Hexagonal	>1425 °C	10	Highest	Low
α′H	Orthorhombic	1160–1425 °C	8	High	Low
α′L	Orthorhombic	680–1160 °C	8	Moderate	Moderate
β	Monoclinic	500–680 °C	6, 8	Slow	Moderate–High
γ	Orthorhombic	<500 °C	6	Negligible	Highest

METHODOLOGY

Synthesis Methods

The selection of synthesis method profoundly influences the polymorph obtained, particle morphology, specific surface area, and consequently the reactivity of C_2S. Several established and emerging routes exist for preparing C_2S with tailored properties [25–28].

3.1. Solid-State Reaction (Sintering)

The conventional approach involves mixing stoichiometric proportions of $CaCO_3$ (or CaO) and SiO_2, followed by calcination at temperatures of 1200–1500 °C. This method produces well-crystallized phases but suffers from high energy consumption and typically yields β-C_2S (stabilized by impurities upon quenching) or γ-C_2S (upon slow cooling). Repeated grinding and firing cycles are often required to achieve complete reaction and eliminate residual free lime [25,29]. The purity of β-C_2S can reach up to 97% at calcination temperatures of 1500 °C [30].

3.2. Sol–Gel Processing

The sol–gel method offers molecular-level mixing of precursors (typically tetraethyl orthosilicate and calcium nitrate), enabling the formation of C_2S at significantly lower temperatures (800–1000 °C) with higher specific surface area and enhanced reactivity [9,31]. Roy and Oyefesobi pioneered this approach in 1977, demonstrating that sol–gel-derived belite exhibited compressive strength improvements of approximately 20 MPa over sintered counterparts due to higher surface area and more favorable particle characteristics [25]. The method is particularly suited for producing bioactive C_2S powders for biomedical applications.

3.3. Hydrothermal Synthesis

Hydrothermal processing involves the reaction of calcium and silicon precursors in aqueous media under elevated temperature and pressure. This route can produce C_2S with unique morphologies, including thin lath structures that exhibit significantly enhanced carbonation efficiency—up to 3.7 times higher than conventionally calcined β-C_2S [32]. The method is also applicable to the synthesis of C_2S from waste precursors, including rice husk ash and fly ash, adding a dimension of waste valorization to the synthesis process itself [28].

3.4. Combustion Synthesis and Flame Spray Pyrolysis

Self-propagating combustion methods and flame spray pyrolysis (FSP) represent more recent approaches capable of producing nanosized C_2S particles in a continuous process [33,34]. FSP in particular offers the advantage of scalability and rapid quenching, which can stabilize metastable high-temperature polymorphs. However, precise control over polymorph selection and minimization of impurity phases (such as residual $CaCO_3$) remain challenges that require further optimization of process parameters including fuel-to-oxidizer ratio, flame geometry, and quenching rate [34].

RESEARCH RESULT

Low-Carbon Cementitious Binders and Carbonation-Cured Materials

4.1. Belite Cements as Low-CO_2 Alternatives to OPC

Belite-rich cements (BCs) represent one of the most direct pathways to decarbonizing cement production. By increasing the proportion of C_2S relative to C_3S in clinker, BCs reduce both the limestone demand and the required kiln temperature, resulting in estimated CO_2 emission reductions of 20–30% at the point of manufacture [5,7]. High-belite cements have been formulated with C_2S contents exceeding 60%, and specialized variants such as belite–calcium sulfoaluminate (B–CSA) cements combine C_2S with ye′elimite ($C_4A_3\check{S}$) to achieve both environmental benefits and improved early-age strength [35]. The primary limitation of BCs is the inherently slow hydration kinetics of β-C_2S, which results in low early-age compressive strength. Extensive research has been directed toward activating belite hydration through mechanical activation (fine grinding), chemical activation (addition of alkalis, sulfates, or nanoparticles), thermal activation (rapid quenching to preserve reactive high-temperature polymorphs), and the stabilization of reactive α' polymorphs through doping with elements such as Na, K, B, and P [1,36,37]. Recent studies have demonstrated that incorporating 1% nano-SiO_2 in combination with low-alkalinity chemical activators ($NaHCO_3$, NaOH, K_2CO_3, KOH) substantially enhances the hydration activity of γ-C_2S by providing nucleation sites for C–S–H gel growth and promoting polymerization of the hydration products [2].

4.2. Carbonation-Cured C_2S Systems

Perhaps the most transformative application of C_2S for environmental sustainability is its use in carbonation-cured construction materials. Unlike conventional hydration-based curing, carbonation curing involves exposing C_2S-based materials to a CO_2-rich atmosphere, where the mineral reacts exothermically with CO_2 to form calcium carbonate ($CaCO_3$) and amorphous silica gel ($SiO_2 \cdot nH_2O$) [8,11]. The key reaction can be expressed as:

$$Ca_2SiO_4 + 2CO_2 + nH_2O \rightarrow 2CaCO_3 + SiO_2 \cdot nH_2O$$

This reaction is particularly significant because it transforms CO_2—a greenhouse gas—into a thermodynamically stable mineral carbonate permanently locked within the material matrix. Comparative studies have demonstrated that among all calcium silicate phases, γ-C_2S exhibits the highest carbonation reactivity, achieving a degree of carbonation (DOC) of approximately 50% within 24 hours under 100% CO_2 at 0.4 MPa pressure [11]. The carbonation products predominantly consist of calcite as the primary $CaCO_3$ polymorph, with aragonite as a secondary phase, encapsulating unreacted grains coated by calcium-modified silica gel [11].

The mechanical performance of carbonation-cured C_2S compacts is impressive. Carbonated β-C_2S achieves compressive strengths exceeding 80 MPa within 24 hours of CO_2 curing, while γ-C_2S compacts reach strengths of approximately 60–70 MPa [11,38]. Self-pulverized low-calcium clinkers with γ-C_2S as the primary mineral have demonstrated compressive strengths of 69.3 MPa after just 4 hours of CO_2 curing and 82.2 MPa at 24 hours, while simultaneously sequestering approximately 277 kg of CO_2 per tonne of clinker compared to the hydration of OPC [38]. These values compare favorably with conventional OPC concrete and demonstrate that C_2S-based materials can deliver both superior performance and substantial environmental benefits. Commercial implementation of carbonation-cured calcium silicate technology has progressed significantly. Solidia Technologies has developed and patented a CO_2-cured cement system whose primary mineral phases include C_3S_2, CS, and C_2S, achieving CO_2 emission reductions of up to 70% compared to conventional OPC production when considering both reduced production emissions and CO_2 uptake during curing [7,39]. The technology has been successfully demonstrated in the production of concrete pavers, blocks, and precast elements.

4.3. Enhancing Carbonation Performance

Several strategies have been explored to optimize the carbonation performance of C_2S systems. Key parameters include CO_2 concentration (higher concentrations accelerate the reaction), relative humidity (optimal at approximately 50–70%), carbonation temperature (elevated temperatures improve kinetics but may alter product morphology), and water-to-cement ratio [7,40]. Recent innovative approaches include the use of biomimetic molecules: L-aspartic acid, L-glutamic acid, and polyacrylic acid at low dosages (2.5–5%) have been shown to enhance the compressive strength of carbonated γ-C_2S pastes by modifying $CaCO_3$ morphology and promoting matrix densification [40]. A novel carbon capture coating based on CO_2-activated C_2S has also been developed, achieving a carbonation degree of 23.3% after just 30 minutes while reducing capillary water absorption of concrete substrates by 20.5% [32].

DISCUSSION

Biomedical Applications of C_2S-Based Composites

5.1. Bioactivity and Biocompatibility

Calcium silicate-based ceramics, including C_2S, belong to a class of bioactive materials that form hydroxyapatite (HA) upon immersion in simulated body fluid (SBF), demonstrating their ability to bond directly with living bone tissue [9,10]. The bioactivity of C_2S stems from its capacity to release calcium (Ca^{2+}) and silicon (SiO_3^{2-}/H_4SiO_4) ions upon hydration or dissolution, which play pivotal roles in stimulating osteogenesis and angiogenesis [41,42]. Calcium ions mediate essential metabolic responses in bone regeneration, while silicon ions promote early-stage calcification, enhance cell attachment, and stimulate the formation of new blood vessels critical for nutrient supply to regenerating tissue [42,43]. C_2S cement exhibits high apatite-forming activity and significantly lower degradation rates in acidic environments compared to many alternative bioceramics. Cytotoxicity assessments have demonstrated that C_2S cement is substantially less cytotoxic than mineral trioxide aggregate (MTA), the conventional standard for endodontic applications [10,44]. These favorable biological properties have positioned C_2S as a candidate material for root-end filling, pulp capping, and as a model system for controlled drug release [9,10].

5.2. Bone Tissue Engineering Scaffolds

The application of C_2S in bone tissue engineering has expanded significantly with advances in three-dimensional (3D) printing technologies. Porous β-C_2S scaffolds fabricated by sintering $CaCO_3$ and SiO_2 mixtures have demonstrated appropriate porosity for cell infiltration while maintaining compressive strengths in the range of trabecular bone [45,46]. The addition of C_2S to calcium sulfate hydrate (CSH) bone cements has been shown to improve both the physicochemical properties and osteogenic activity of the composite, addressing the limitations of rapid resorption and weak bioactivity inherent to CSH alone [47]. Composite scaffold strategies have proven particularly effective. 3D-printed calcium silicate/polycaprolactone (CS/PCL) scaffolds coated with decellularized extracellular matrix (dECM) have demonstrated excellent biocompatibility, enhanced cellular adhesion, proliferation, and osteogenic differentiation, as well as anti-inflammatory characteristics characterized by decreased expression of TNF-α and IL-1 [48]. The incorporation of bioactive ions into C_2S scaffolds represents another promising strategy: zinc-doped and copper-doped C_2S cements have exhibited combined osteogenic and antibacterial properties, while strontium-doped calcium silicate scaffolds promote osteoblastic differentiation while reducing osteoclastogenesis [42,49].

5.3. Dental Applications

In dentistry, C_2S is a principal constituent of calcium silicate-based cements used for endodontic applications, including direct and indirect pulp capping, root-end filling, and perforation repair [10,44]. The integration of zinc into C_2S to form hardystonite ($Ca_2ZnSi_2O_7$) has generated particular interest, as this composite phase demonstrates improved setting characteristics, enhanced antibacterial properties, and superior bioactivity in artificial saliva media [50]. Recent research has focused on improving the injectability, anti-washout performance, and radiopacity of C_2S-based dental cements through the incorporation of various additives, including nanoparticles of bismuth oxide and zirconium oxide for contrast enhancement under radiographic examination [10,44].

Industrial Waste Valorization and Environmental Remediation

6.1. Steel Slag Valorization

Steel production generates vast quantities of slag containing C_2S as a major mineral phase. In basic oxygen furnace (BOF) slag, electric arc furnace (EAF) slag, ladle furnace slag (LFS), and argon oxygen decarburization (AOD) slag, C_2S is present predominantly in the γ polymorph—a consequence of slow cooling during slag processing that allows the β → γ transformation to proceed [13,23,51]. While γ-C_2S's negligible hydraulic activity has historically limited slag utilization as cementitious material, its high carbonation reactivity has opened a transformative pathway for waste valorization [23,24]. Accelerated carbonation of steel slag converts the reactive calcium silicate and oxide phases into stable carbonates while simultaneously improving the physical and chemical properties of the slag. Research has demonstrated CO_2 uptakes of up to 300 g CO_2/kg for AOD slag and 180 g CO_2/kg for milled EAF slag under optimized conditions (50 °C, 3 bar, 100% CO_2) [14]. The carbonated slag products exhibit improved volume stability (through consumption of expansive free CaO and free MgO), enhanced mechanical strength (exceeding 30 MPa in carbonated compacts), reduced alkalinity, and diminished heavy metal leachability [14,52]. Carbonation processes applied to steel slag can potentially sequester over 10% of the CO_2 emissions from a typical EAF steel manufacturing plant, equivalent to approximately 0.4 Mt CO_2 per year in industrialized countries [14].

6.2. CO_2 Mineralization and Carbon Capture

Mineral carbonation of C_2S-bearing industrial wastes offers a promising pathway for permanent CO_2 sequestration. The process involves the reaction of CO_2 with the calcium-bearing mineral phases to form geochemically stable carbonates, thereby converting waste materials into useful construction products while permanently storing CO_2 [13,53]. The integration of carbonation technology with waste recycling has been demonstrated for recycled concrete fines, where full carbonation of hydrated cement paste fines sequesters all CO_2 originally released during limestone calcination, while simultaneously generating amorphous silica and alumina gels with pozzolanic reactivity suitable for use as supplementary cementitious materials [53].

Ground granulated blast-furnace slag (GGBFS) modified with 5% γ-C_2S has demonstrated enhanced compressive strength under accelerated carbonation curing, with the γ-C_2S facilitating the formation of additional calcium carbonate phases that densify the microstructure [54]. Cemented paste backfill (CPB) technology, which combines tailings, binders, and water for mine backfilling, has also been identified as a potential vehicle for CO_2 uptake, with each tonne of CPB capable of absorbing approximately 78 kg of CO_2 from mine ventilation air [55]. These applications illustrate the broader potential of C_2S-based carbonation to contribute to circular economy principles within the construction and mining industries.

6.3. Heavy Metal Immobilization

The carbonation of C_2S-bearing slags provides an additional environmental benefit through the immobilization of heavy metals. Carbonation-hydration curing of stainless steel slag has been shown to reduce the cumulative leaching fractions of chromium and nickel by 65.8% and 43.2%, respectively, at a carbonation degree of 36.2% [52]. The formation of dense carbonate and silica gel matrices physically encapsulates heavy metal species, while the reduction in pH and changes in pore solution chemistry alter the speciation and solubility of contaminant ions [52,56]. The inclusion of C_2S in calcium sulfoaluminate-based systems has also been explored for the immobilization of heavy metals in contaminated soils and industrial waste streams, leveraging the formation of ettringite and C–S–H phases as effective sorbents [57].

Current Challenges and Future Research Directions

Despite the remarkable progress outlined in this review, several challenges must be addressed to realize the full potential of C_2S-based composites for environmentally friendly applications. The following areas represent the most critical research priorities:

Hydration kinetics enhancement: The inherently slow hydration of β-C_2S remains the most significant barrier to widespread adoption of belite-rich cements in structural applications. While chemical and physical activation strategies have shown promise at the laboratory scale, translating these approaches to industrial production at reasonable cost and complexity remains challenging. The development of synergistic activation systems—combining nano-seeding, chemical activators, and optimized particle size distribution—represents a particularly promising avenue [2,36].

Scalable carbonation curing infrastructure: Carbonation-cured C_2S products require concentrated CO_2 sources, specialized curing chambers, and process optimization for industrial throughput. The establishment of supply chains that link CO_2 emitters (e.g., power plants, cement kilns) with C_2S-based product manufacturers is essential for economic viability. Integration with carbon capture systems currently under development for industrial decarbonization could provide the required CO_2 streams at reduced cost [7,8].

Polymorph control and stabilization: Precise control over C_2S polymorph formation and stabilization is critical because each application demands specific polymorphic properties. The stabilization of reactive α′ polymorphs for enhanced hydraulic activity, or the deliberate promotion of γ-C_2S for maximal carbonation reactivity, requires deeper understanding of the thermodynamic and kinetic factors governing phase transitions in doped systems [1,18,22].

Biomedical long-term performance: While in vitro studies of C_2S-based bioceramics are encouraging, comprehensive long-term in vivo studies and clinical trials remain limited. Understanding the degradation behavior, ion release kinetics, and immunological response over extended implantation periods is essential for regulatory approval and clinical translation [42,48].

Life-cycle assessment and techno-economic analysis: Comprehensive, cradle-to-grave life-cycle assessments of C_2S-based systems—encompassing raw material extraction, synthesis energy, carbonation or hydration curing, service life, and end-of-life scenarios—are needed to quantify the true environmental benefits relative to conventional alternatives [7,53].

CONCLUSIONS AND RECOMMENDATIONS

Dicalcium silicate stands at the intersection of multiple sustainability challenges facing the materials engineering community. As this review has demonstrated, C_2S-based composites offer a portfolio of environmentally friendly applications that spans low-carbon construction materials, bioactive medical ceramics, and industrial waste valorization strategies. The unique polymorphism of C_2S—far from being a liability—provides a toolkit of distinct properties: β-C_2S for hydraulic cementitious binders, γ-C_2S for carbonation-cured CO_2-sequestering materials, and bioactive C_2S ceramics for tissue engineering and dental repair. The carbonation curing of C_2S-based materials is perhaps the most environmentally compelling application, offering the dual benefit of reduced production emissions and active CO_2 sequestration within the cured product.

The achievement of compressive strengths exceeding 80 MPa through 24-hour CO_2 curing, coupled with CO_2 savings of approximately 277 kg per tonne of clinker, demonstrates the viability of this approach for commercial construction products. Similarly, the valorization of C_2S-rich steel slags through accelerated carbonation addresses waste management challenges while permanently storing CO_2 and producing useful construction materials. Looking forward, the convergence of advances in nanotechnology, computational materials science, additive manufacturing, and carbon capture infrastructure is expected to accelerate the development and deployment of C_2S-based solutions. The continued evolution of belite-rich cement formulations with enhanced early-age performance, the integration of C_2S carbonation with industrial CO_2 capture systems, and the clinical translation of C_2S bioceramics collectively represent a multifaceted pathway toward a more sustainable materials economy.

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