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Application Progress in the Preparation of Eco-Friendly Composite Ceramics from Solid Waste

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Abstract. Against increasingly prominent resource and environmental issues, the resource utilization of solid waste has become a research hotspot. The preparation of environmentally friendly composite ceramics using solid waste as the primary raw material not only facilitates the resource utilization of solid waste and ecological improvement, but also enables low-cost green ceramic manufacturing. This paper focuses on the application of coal-based solid waste, smelting slag, desert sand, and mining tailings in eco-friendly composite ceramics, and points out some challenges encountered in the practical application and development of converting solid waste into composite ceramic raw materials.

Keywords: Solid Waste; Environmental Protection; Composite Ceramics; Green Manufacturing

1. Introduction

With the acceleration of industrialization, the global annual production of industrial solid waste such as fly ash, steel slag and tailings has exceeded 10 billion tons, while desert sand, as a naturally abundant raw material, constitutes as much as one-third of the Earth's total land area. The stockpiling of these wastes not only occupies land resources but also leads to severe environmental issues including heavy metal leakage, soil acidification and dust pollution.

Meanwhile, the traditional ceramics industry has long relied on natural mineral resources like clay and quartz, where excessive mining has exacerbated resource depletion and ecological damage. The eco-friendly composite ceramic products produced from solid waste can not only reduce mineral consumption in industrial ceramic production but also improve the ecological environment and lower production costs, achieving low-cost green manufacturing for the ceramics industry and pioneering a sustainable development path.

2. Coal-Based Solid Waste

Coal-based solid wastes are industrial byproducts generated during coal mining, processing, and utilization that are difficult to repurpose, with coal gangue and fly ash constituting the largest proportion. Fly ash, a fine powder collected from flue gases after pulverized coal combustion, is the primary solid waste from coal-fired power plants. Coal gangue, a multi-component solid waste produced during coal

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mining and washing, accompanies approximately 150-250 kg/ton of raw coal. Long-term stockpiling of coal gangue and fly ash can lead to dust emissions, heavy metal leakage, and soil acidification. Utilizing coal gangue or fly ash as raw materials for composite ceramics addresses environmental pollution and achieves high-value utilization of coal-based solid waste.

2.1. Fly Ash

Fly ash is rich in SiO_2 and Al_2O_3, with fine particles and compositions similar to traditional ceramic raw materials (e.g., kaolin, feldspar). Compared to conventional ceramics, fly ash contains mullite crystal nuclei or microcrystals that reduce the free energy required for nucleation during sintering, lowering phase formation temperatures.

Li et al. [1] successfully synthesized needle-like mullite whiskers in situ within ceramics using fly ash as the primary raw material, with $Al(OH)_3$ and Al_2O_3 added as aluminum sources and AlF_3 as a sintering aid. The addition of AlF_3 significantly increased the aspect ratio of the whiskers and enhanced the mechanical strength of mullite, achieving a bending strength of approximately 100 MPa at 1550°C (as shown in Figure 1). However, this fluoride-assisted in situ synthesis method for mullite whiskers poses environmental pollution risks. Mineral decomposition presents a novel approach for preparing mullite whiskers using fly ash as the base material. Peng et al. [2] utilized high-alumina fly ash as the main raw material and employed mineral decomposition to produce porous ceramics with an average pore size of 0.5 mm, an apparent porosity of 77%, and a bending strength of 5 MPa, achieving whiskers with an aspect ratio of 8.8. This method enables the fluoride-free synthesis of mullite whiskers from fly ash, and the resulting material shows promising potential for future applications in thermal insulation, adsorption, and filtration fields.

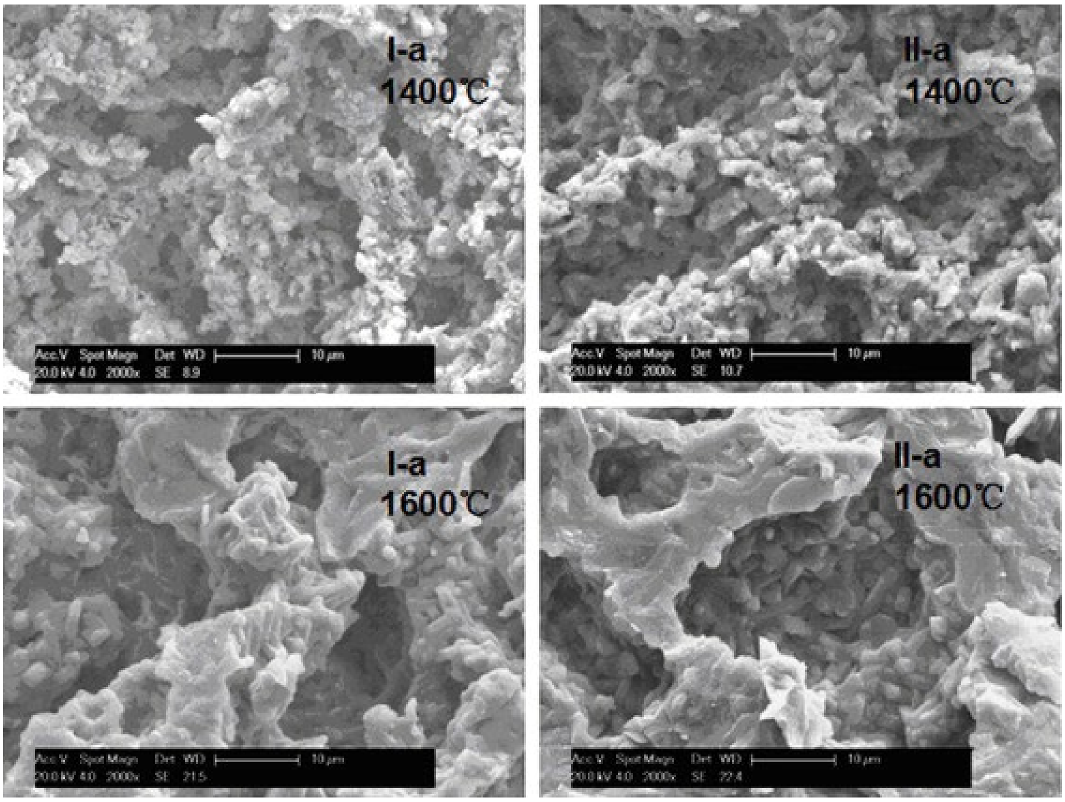

Figure 1. Microstructure of sample fracture surface.

Ma et al. [3] synthesized a ZrO_2-mullite-corundum composite ceramic with excellent properties, including high strength, toughness, wear resistance, and thermal shock resistance, using fly ash, ZrO_2, and Al_2O_3 as raw materials at 1500–1600°C. In subsequent research, they further produced porous

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mullite ceramics with high closed porosity and strength by incorporating SiC. When the SiC content was increased from 0 to 15 wt%, prismatic mullite crystals grown from the saturated glass phase enhanced particle bonding (as indicated by the red arrows in Figure 2), resulting in a 25.24% improvement in cold compressive strength [4].

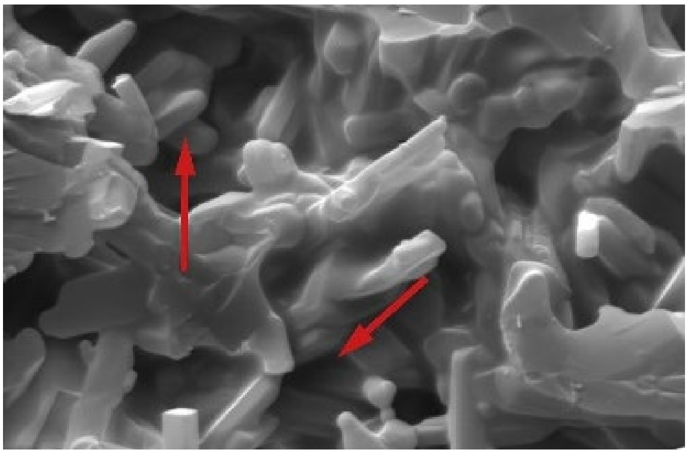

Figure 2. SEM image of fracture surface of SiC sample added

At 1500°C, with the addition of a fluxing agent, the mullite in fly ash completely decomposes into SiO_2 and Al_2O_3, which subsequently react with MgO to form spinel [5]. Therefore, using forsterite, fly ash, and Al_2O_3 as raw materials enables the synthesis of forsterite-spinel ceramics with excellent thermal shock resistance and refractory properties [6].

2.2. Coal Gangue

The chemical composition of coal gangue is similar to that of fly ash, as both are solid waste materials rich in aluminum and silicon. However, coal gangue particles are relatively coarser than fly ash, and its primary mineral constituents are clay and quartz sand. Through proper formulation and sintering processes, it can be used to prepare ceramic phases such as mullite and cordierite.

β-SiAlON composite ceramics were successfully synthesized via carbothermal reduction using coal gangue under nitrogen atmosphere [7], as shown in Figure 3. The main phase was β-SiAlON, accompanied by minor amounts of SiC and Al_2O_3. The optimal sample, obtained after holding at 1500°C for 4 hours, exhibited flexural and compressive strengths of 10.1 MPa and 24.7 MPa, respectively, with linear shrinkage and porosity reaching 13.1% and 22.3%. This study provides an efficient and environmentally friendly solution for coal gangue recycling, achieving dual benefits of optimized material performance and reduced environmental risks.

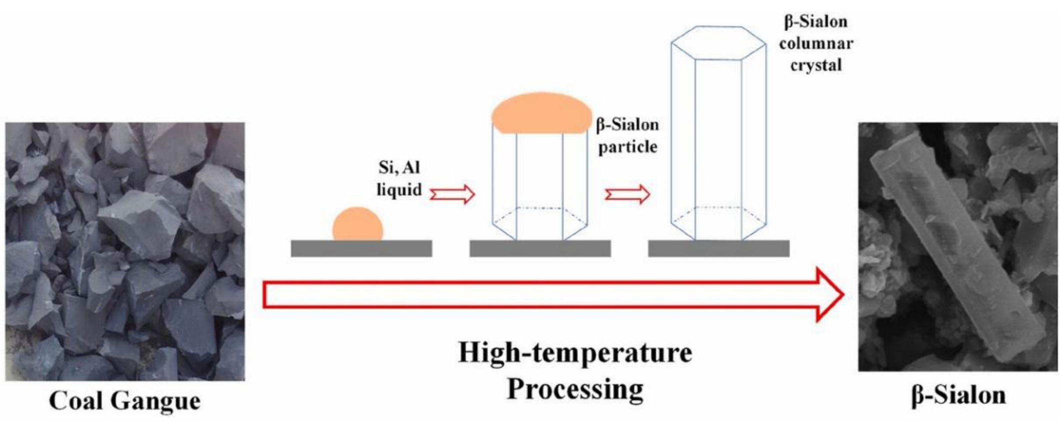

Figure 3. The reaction process of transforming coal gangue into β-SiAlON phase.

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Zirconia-mullite ceramics were successfully prepared using $ZrOCl_2 \cdot 8H_2O$ and coal gangue [8]. With 12% $ZrOCl_2 \cdot 8H_2O$ addition, the sample density reached 2.83 g/cm³. Research indicates that the mullite crystalline matrix effectively restrained the phase transformation of zirconia from the high-temperature tetragonal phase to the monoclinic phase. The uniformly distributed tetragonal zirconia particles contributed to toughening effects, significantly enhancing the flexural strength and fracture toughness from 162.40 MPa and 2.38 $MPa \cdot m^{1/2}$ (for pure mullite ceramics) to 285.04 MPa and 3.55 $MPa \cdot m^{1/2}$, respectively.

Using natural bauxite and coal gangue as raw materials, a cost-effective and energy-efficient corundum-mullite based ceramic support was fabricated through $CaCO_3$ addition [9]. During sintering, $CaCO_3$ decomposed into CaO, facilitating liquid-phase sintering and thereby reducing the required sintering temperature. With increasing $CaCO_3$ content, mullite grains became progressively refined and developed an interlocking rod-like microstructure, leading to improved toughness and crush resistance of the material.

3. Smelting Slag

Smelting slag, a solid waste generated during metal or mineral smelting processes, features complex composition and numerous harmful impurities. Traditional disposal methods such as stockpiling or landfilling can lead to environmental hazards including soil salinization and surface corrosion. For smelting slags with different chemical compositions, selecting appropriate raw material formulations and process flows can effectively transform them into low-cost, environmentally friendly ceramic products.

3.1. Metallic Slag

Metallic slags not only contain their respective metal oxides (such as copper oxide, nickel oxide, zinc oxide, etc.) but also possess high concentrations of calcium oxide, magnesium oxide, or iron oxide. Compared to silica- and alumina-rich solid wastes, metallurgical slags are typically incorporated in smaller quantities when preparing quartz-based ceramic materials. However, they can be utilized in larger proportions in ceramic systems where anorthite or pyroxene serve as the primary phases. Composite ceramic materials within the $CaO\text{-}MgO\text{-}SiO_2$ system (diopside/augite-based) can be synthesized using over 30 wt% steel slag combined with clay as the main raw materials, achieving a bending strength of up to 99.84 MPa. The glass phase present in steel slag facilitates liquid-phase formation at lower temperatures, reducing the firing temperature to 1220–1230°C [10]. This demonstrates that steel slag-derived pyroxene ceramics not only enable waste valorization and energy conservation but also exhibit excellent mechanical properties.

Furthermore, metallurgical slag exhibits excellent high-temperature thermal stability and chemical stability, making it a viable candidate for high-temperature thermal energy storage applications. Wu et al. [11] successfully fabricated a low-cost $Al_2O_3\text{-}CaAl_{12}O_{19}$ ceramic composite using ferrotitanium slag and alumina as raw materials. The TiO_2 and MgO present in the ferrotitanium slag dissolved into the Al_2O_3 and $CaAl_{12}O_{19}$ crystal lattices, forming solid solutions that significantly enhanced sintering densification. When sintered at 1500°C with a 60 wt.% ferrotitanium slag addition, the resulting composite achieved a relative density of 94.07%, a flexural strength of 206.53 MPa, and exhibited a specific heat capacity and thermal conductivity of 1.0801 $J \cdot g^{-1} \cdot K^{-1}$ and 5.278 $W \cdot m^{-1} \cdot K^{-1}$, respectively—markedly superior to those of pure alumina and pure ferrotitanium slag specimens.

In a separate study [12], researchers developed a composite thermal energy storage material by fabricating a cordierite-mullite porous ceramic scaffold from secondary aluminum slag and ferronickel slag, followed by impregnation with $NaNO_3$ phase change material (PCM) via melt infiltration. The composite demonstrated a thermal conductivity of 1.84–1.98 $W \cdot m^{-1} \cdot K^{-1}$ at 25°C, representing a >270% improvement over pure $NaNO_3$, while maintaining a thermal conductivity exceeding 1.4 $W \cdot m^{-1} \cdot K^{-1}$ even at elevated temperatures. After 100 thermal cycles, the composite exhibited minimal latent heat loss (2.9–5.6%) and retained a compressive strength above 100 MPa, confirming its structural integrity. This

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innovative material combines cost-effectiveness with environmental sustainability, positioning it as a promising solution for thermal energy storage.

3.2. Red Mud

Red mud, an industrial solid waste generated during alumina extraction from bauxite, primarily consists of Al_2O_3, SiO_2, CaO, and trace amounts of other metal oxides. By utilizing red mud as the primary raw material and incorporating SiC powder to reinforce the mechanical strength of the matrix, along with $Al(OH)_3$ and catalysts (V_2O_5 and AlF_3) to promote mullite phase nucleation and enhance the ceramic's physical-mechanical properties, researchers successfully fabricated SiC/mullite composite porous ceramics [13]. The addition of graphite as a pore-forming agent yielded a high-performance eco-friendly porous ceramic exhibiting both high compressive strength (49.4 MPa) and moderate porosity (31.4%).

3.3. Boron Mud

Borate ores generate boron-rich sludge (boron mud) during mineral processing, primarily composed of SiO_2, CaO, MgO, B_2O_3, and other metal oxides. Zanelli et al. [14] incorporated boron mud (containing 6.4% B_2O_3) as a fluxing agent into ceramic raw materials, demonstrating its ability to improve powder grindability and green density while reducing sintering temperature. However, excessive addition (>10%) decreased product bulk density and increased closed porosity. The resulting ceramic phases consisted mainly of quartz, mullite, and a glassy matrix. While boron mud had negligible effects on quartz and mullite phase composition, it notably reduced SiO_2 content in the glass phase while increasing B_2O_3/CaO/MgO concentrations. Controlled addition of boron mud in quartz-based ceramic production proves effective for both energy savings and waste recycling solutions.

4. Desert Sand

Deserts occupy one-third of the total land area, and desert sand, as a kind of natural wasteland, is very rich in sources, with fine particles, low water content, narrow range of particle sizes, and stable chemical compositions [15], which make it naturally advantageous to prepare ceramics by using desert sand instead of pure SiO_2.As shown in Table 1, taking the Kubuqi Desert as an example, the main composition of desert sand is SiO_2, and some other impurity components such as Al_2O_3, CaO, MgO, Fe_2O_3, K_2O, etc. are present. Compared with high-purity quartz sand, desert sand contains a large number of alkali metal and alkaline earth metal oxides, which belongs to low-quality quartz sand. In the process of synthesizing ceramics using desert sand, the low melting point impurities will melt into the liquid phase during the sintering process, which can effectively reduce the sintering temperature of ceramics [16,17], however, the glassy phase formed in this process will lead to the reduction of the toughness of desert sand ceramics while enhancing the densification of ceramics, which will have some negative effects.

Table 1. Chemical Composition of Desert Sand in Kubuqi Desert.

Consist of	SiO_2	Al_2O_3	CaO	MgO	Fe_2O_3	K_2O	Na_2O	Bal.
Content/wt%	78.91	9.50	2.33	0.86	2.26	2.08	2.00	2.06

To further expand the application scope of desert sand and improve the overall performance of ceramics, introducing reinforcing phases is an effective approach. By adding MgO and SiC to desert sand, followed by ball milling, wet pressing, and sintering at 1300–1400℃, a $MgSiO_3$-SiC composite ceramic with a porosity of less than 5% can be obtained, as shown in Figure 4 [18]. When sintered at 1350℃, the sample containing 30 wt% SiC exhibits the highest flexural strength of 173.18 MPa, significantly higher than that of the ceramic sample without SiC (approximately 100 MPa). With the addition of 50 wt% SiC and sintering at 1400℃, the ceramic achieves the highest hardness of 804.70 HV and the lowest thermal expansion coefficient of 6.09×10^{-6}/℃. The SiO_2-ZrO_2 ceramic prepared

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using desert sand and ZrO_2 as raw materials primarily consists of α-quartz, $ZrSiO_4$, and a glass phase. $ZrSiO_4$ initially forms on the outer layer of ZrO_2 particles and then grows inward, creating a core-shell structure [19]. The addition of ZrO_2 significantly enhances the flexural strength and hardness of the ceramic. When 50 wt% ZrO_2 is added and sintered at 1500℃ for 2 h, the ceramic achieves the highest flexural strength of 109.3 MPa and hardness of 835.7 HV. Compared to the sample without ZrO_2, the flexural strength and hardness increase by 51.8% and 28.6%, respectively.

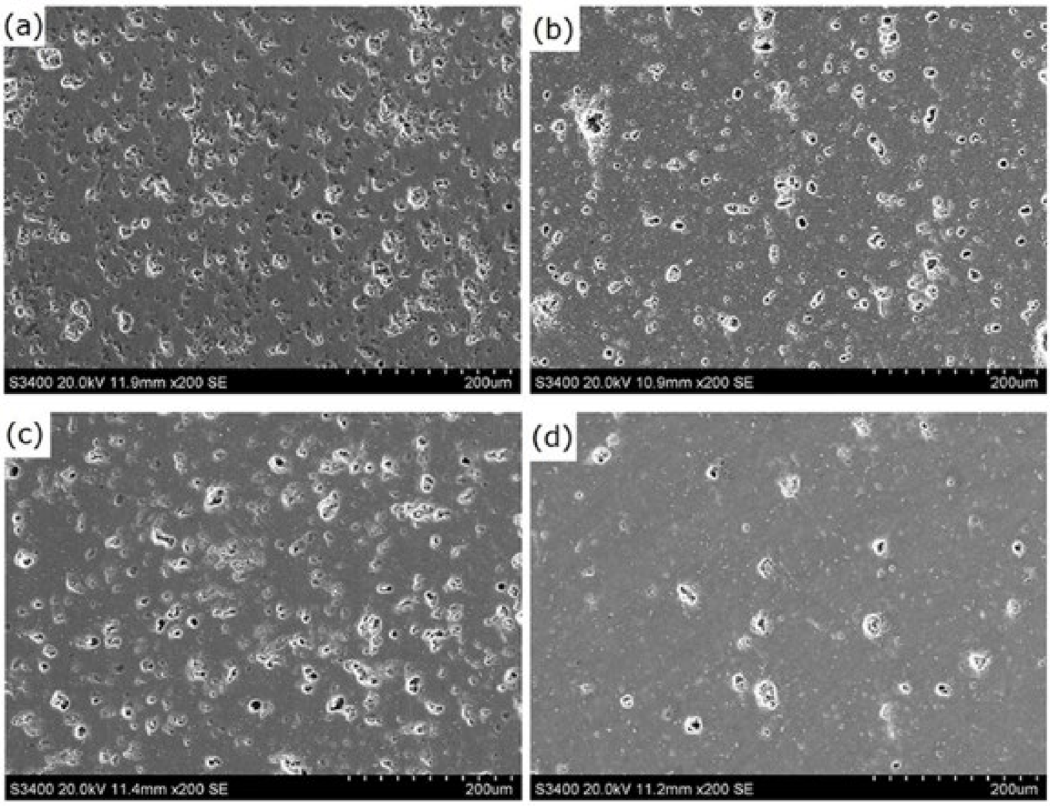

Figure 4. Surface morphology of samples prepared by adding 30 wt% SiC at different sintering temperatures: (a)1250°C, (b)1300°C,(c) 1350°C and (d) 1400°C.

5. Mining Tailings

Tailings are industrial solid wastes generated during ore processing through crushing, screening, grinding and flotation. Their chemical compositions vary depending on ore types, primarily consisting of SiO_2, Al_2O_3, Fe_2O_3, CaO and MgO, demonstrating their potential for manufacturing traditional ceramic products like bricks, tiles and concrete. Fernandes et al. [20] successfully produced eco-friendly ceramic samples containing mullite, quartz, calcite and anorthite as primary phases by incorporating quartzite and scheelite tailings into conventional ceramic raw materials. Performance tests confirmed that adding less than 8% scheelite tailings had no significant impact on product properties, indicating these tailings can effectively substitute traditional ceramic materials. Composite ceramics sintered at 1200℃ using iron ore tailings, charcoal blast furnace slag, foundry sand and natural clay as raw materials exhibited a bending strength of 12.19 MPa, water absorption of 2%, and density of 2.03 g/cm^3. With iron ore tailings and blast furnace slag accounting for up to 70% of the raw materials, this formulation shows great potential for large-scale tile production [21].

6. Coal-Smelting Slag Solid Waste

Steel slag, another form of industrial solid waste, primarily consists of CaO, Fe_2O_3, and SiO_2-components that complement coal gangue in composition. Researchers have successfully developed coal gangue-steel slag ceramics using these industrial solid wastes as raw materials [22]. When the steel

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slag to coal gangue ratio ranges between 8:2 and 2:8, the resulting ceramics form an anorthite ($CaAl_2Si_2O_8$)-diopside ($CaMgSi_2O_6$) eutectic structure. The controlled addition of steel slag significantly enhances porosity while reducing bulk density, demonstrating promising potential for porous material applications. Furthermore, using aluminum chips as a reducing agent at 1000-1100°C enables the reduction of coal gangue into silicon, which subsequently forms nitride whiskers through a vapor-solid mechanism to reinforce the ceramic matrix. This process markedly improves the ceramic's fracture toughness while maintaining excellent thermal stability, positioning these materials as highly promising for thermal energy storage applications [23].

7. Desert Sand-Coal Solid Waste Composites

High-quality quartz-anorthite composite ceramics can also be synthesized using desert sand and fly ash [24]. Within the sintering temperature range of 1100–1200°C, bulk densification and anorthite phase transformation occur simultaneously, with the phase transition temperature increasing as the proportion of fly ash rises. When the fly ash content reaches 40%, samples sintered at 1100°C exhibit optimal mechanical properties (flexural strength of 115.6 MPa, Vickers hardness of 668.3 HV), while samples with 100% fly ash demonstrate the lowest thermal expansion coefficient (5.1×10^{-6}/K) and the highest bulk density (2.5 g/cm^3). The material forms an anorthite phase through solid-state reactions, combined with β-tridymite formation and liquid-phase sintering mechanisms, ultimately resulting in a composite system composed of α-quartz, β-tridymite, anorthite, and an amorphous phase. All samples show water absorption rates below 1.8%. This technology effectively utilizes industrial solid waste to produce construction ceramics, thermal insulation materials, and other civil engineering products, offering significant economic and environmental benefits.

8. Conclusion

In summary, environmentally friendly composite ceramics produced from solid waste have been successfully applied in various fields, including thermal insulation, heat storage, seismic resistance, and ceramic tiles. With the growing trend of solid waste resource utilization, they hold broad prospects for future development.

However, several challenges remain in achieving the industrialization and large-scale application of solid waste-derived materials:

(1) Variability in composition: Solid waste from different sources (e.g., fly ash, coal gangue, smelting slag) exhibits significant differences in chemical composition. Even the same type of solid waste may vary due to origin or processing methods, making ceramic formulation design difficult and leading to inconsistent product performance.

(2) Preprocessing and pollution risks: Pretreatment processes such as crushing, screening, and impurity removal are required. Additionally, residual chemical agents or heavy metals in smelting slag and tailings may release harmful gases (e.g., SO_2, heavy metal vapors) during high-temperature sintering. These preprocessing steps and hazardous gas treatments increase production costs, hindering industrial-scale development.

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Conflict of Interest: The authors declare that there is no conflict of interest regarding the publication of this article.

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