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Eco-Friendly Cement Clinker Manufacturing Using Industrial Solid Waste and Natural Idle Resources as Alternative Raw Materials

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Abstract: The production of silicate cement clinker consumes a huge amount of non-renewable mineral resources, resulting in both land degradation and environmental issues. Utilizing alternative silica sources is an effective way to achieve sustainable cement production. The present work reviews the use of industrial solid waste (such as iron tailings, steel slag, red mud, and ceramic waste) and natural idle resources (such as pumice, basalt, and desert sand) as sustainable substitutes for traditional SiO_2 sources in silicate cement clinker synthesis. These alternative sources are characterized by their wide availability and low cost, and they are capable of alleviating resource shortages and environmental pressures. The characteristics of clinker calcination, cement hydration, and hardening properties were elaborated. The results demonstrated that by optimizing the dosage of alternative sources and improving the calcination process, the properties of cement clinker can be effectively enhanced, while achieving efficient resource utilization and sustainable environmental development. This review provides a theoretical basis and technical support for promoting the application of industrial solid waste and natural idle resources in the cement industry, and has important guiding significance for promoting the green manufacturing in cement industry.

Keywords: Industrial Solid Waste; Natural Idle Resources; Cement Clinker; Green Manufacturing

1. Introduction

Silicate cement clinker, as the primary component of silicate cement, consumes substantial quantities of quartz mineral resources (including clay, river sand, and silica) during its production. With global annual cement production reaching approximately 4 billion tons, the demand for industrial quartz is nearly 800 million tons [1, 2]. This has led to extensive resource extraction, severe land degradation, and environmental issues. To solve this problem, utilizing industrial solid waste (such as iron tailings, steel slag, red mud, and ceramic waste) [3-8] and natural idle resources (such as pumice, basalt, and desert sand) [9-13] as substitutes for traditional SiO_2 sources in the preparation of cement clinker has been investigated. Therefore, the clinker calcination, cement hydration, and hardening properties of utilizing

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industrial solid waste and natural idle resources in the preparation of silicate cement clinker were reviewed, providing a theoretical basis for the application of alternative raw materials in the cement industry to achieve green manufacturing.

2. Advances in Clinker Calcination and Hydration Using Alternative Silica Sources

The primary mineralogical constituents of silicate cement clinker are tricalcium silicate ($3CaO{\cdot}SiO_2$, C_3S), dicalcium silicate ($2CaO{\cdot}SiO_2$, C_2S), tricalcium aluminate ($3CaO{\cdot}Al_2O_3$, C_3A), and tetracalcium aluminoferrite ($4CaO{\cdot}Al_2O_3{\cdot}Fe_2O_3$, C_4AF) [14]. Among these, C_3S and C_2S play a crucial role in the hardening of cement. According to EN 197-1:2011, the total content of C_3S and C_2S must account for two-thirds of the cement clinker. C_3S demonstrates higher early strength than C_2S [15, 16], leading to a preference for clinker with higher C_3S content. Domestic and international research on alternative silica sources for cement clinker has mainly focused on industrial solid waste and natural idle resources.

2.1. Industrial Solid Waste

2.1.1. Iron Tailings

Iron tailings are waste residues from iron ore processing. Their major composition is similar to clay, with high contents of SiO_2, Al_2O_3, and Fe_2O_3, all of which are the main raw materials for silicate cement clinker formation. Additionally, the high content of MgO and trace K_2O and Na_2O in iron tailings act as fluxes during calcination, facilitating the phase transformation of calcium silicates. Experiments showed that adding 10 wt% iron tailings (CaO: 16.07 wt%, SiO_2: 35.48 wt%, Al_2O_3: 6.1 wt%, Fe_2O_3: 20.71 wt%, MgO: 19.81 wt%, K_2O: 0.5 wt%, Na_2O: 1.34 wt%) and calcining at 1420 °C for 1 h obtained clinker with 57.45 wt% C_3S, higher than the control sample without addition (55.13 wt%). The main mineral phases (C_3S, C_2S, C_3A and C_4AF) remained unchanged (Figure 1). Compressive strengths at 3 d and 28 d were 24.1 MPa and 52.3 MPa, respectively, with increases of 13.68% and 3.36%. Adding 20 wt% iron tailings, C_3S reduced to 48.32 wt% and compressive strength decreased significantly (by 21.7% at 3 d and 24.11% at 28 d) [4]. This can be explained by the fact that excess MgO in the raw meal forms free MgO, producing $Mg(OH)_2$ during hydration, which hinders calcium silicate hydration [17]. Replacing clay entirely with iron tailings lowered the formation temperature of C_2S by 30 °C. Meanwhile, clinker from calcining at 1450 °C for 1 h showed an earlier hydration time of C_3S (6 h) and compressive strengths of 20.8 MPa and 48.6 MPa at 3 d and 28 d, respectively, with increases of 0.48% and 5.19% compared to the control [8]. In conclusion, the proper use of iron tailings can promote the phase transformation of clinker, accelerate hydration, and enhance mechanical properties.

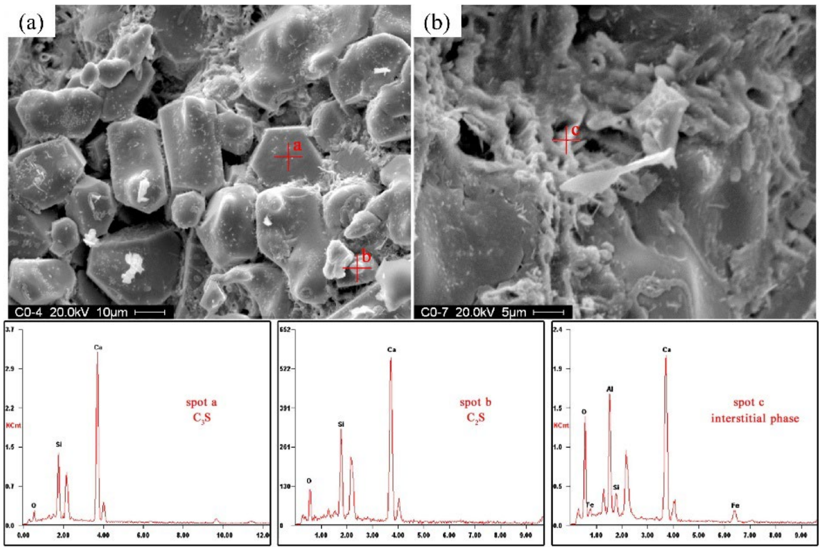

Figure 1. SEM images of iron tailings cement clinker: (a) C_3S and C_2S, (b) interstitial phase.

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2.1.2. Red Mud

Red mud, a by-product of alumina production, primarily consists of SiO_2, Al_2O_3, and Fe_2O_3, and CaO, all of which are essential for forming the main mineral phases in cement clinker. Incorporating 3.5 wt% red mud (CaO: 15.15 wt%, SiO_2: 8.17 wt%, Al_2O_3: 23.98 wt%, Fe_2O_3: 49.04 wt%, MgO: 0.24 wt%，K_2O: 0.17 wt%，Na_2O: 3.25 wt%) into the raw meal and calcining at 1450 °C for 30 min resulted in the formation of the clinker with well-formed, uniformly fine-grained C_3S, C_2S, C_3A and C_4AF. The total content of C_3S and C_2S met the standard EN 197-1:2011, although the C_3S content is 3.92 wt% lower than that of the reference sample without red mud. The compressive strengths of the obtained cement paste at 2 d and 7 d were comparable to the reference sample, with 22 MPa and 32 MPa, respectively. Furthermore, the 28 d and 90 d strengths are 45 MPa and 56 MPa, respectively, increases of 4.6% and 6.86% [6]. The above results indicate that red mud can be used as a raw material for cement clinker production, yielding clinker with satisfactory strength.

2.1.3. Steel Slag

Steel slag is a solid waste generated during the conversion of iron to steel. Its chemical composition includes 45–60 wt% CaO, 10–15 wt% SiO_2, 1–5 wt% Al_2O_3, 10–29 wt% Fe_2O_3, and 3–13 wt% MgO [18]. Adding 10.5 wt% steel slag to the raw meal and calcining at 1450 °C for 30 min does not affect the clinker microstructure or the formation of main mineral phases. Meanwhile, the C_3S content increased from 61.75 wt% to 62.24 wt%. The compressive strengths of the mortar at 2 d, 7 d, 28 d, and 90 d were comparable to those of Ordinary Portland Cement (OPC) (Figure 2a) [3]. Additionally, incorporating 14 wt% ladle metallurgy stainless steel slag (CaO: 56.4 wt%, SiO_2: 32.5 wt%, Al_2O_3: 1.3 wt%, Fe_2O_3: 0.4 wt%, MgO: 7.5 wt%) lowered the transformation temperature of C_2S. This is attributed to the presence of F and Cr elements in the ladle metallurgy slag, which act as mineralizers and accelerate the crystallization of C_2S. However, the hydration time of C_3S in the clinker was prolonged by 7 h, and the hydration rate was reduced by 8.43% (Figure 2b). Nevertheless, the 7 d and 28 d strengths were comparable to those of OPC (Figure 2c) [19].

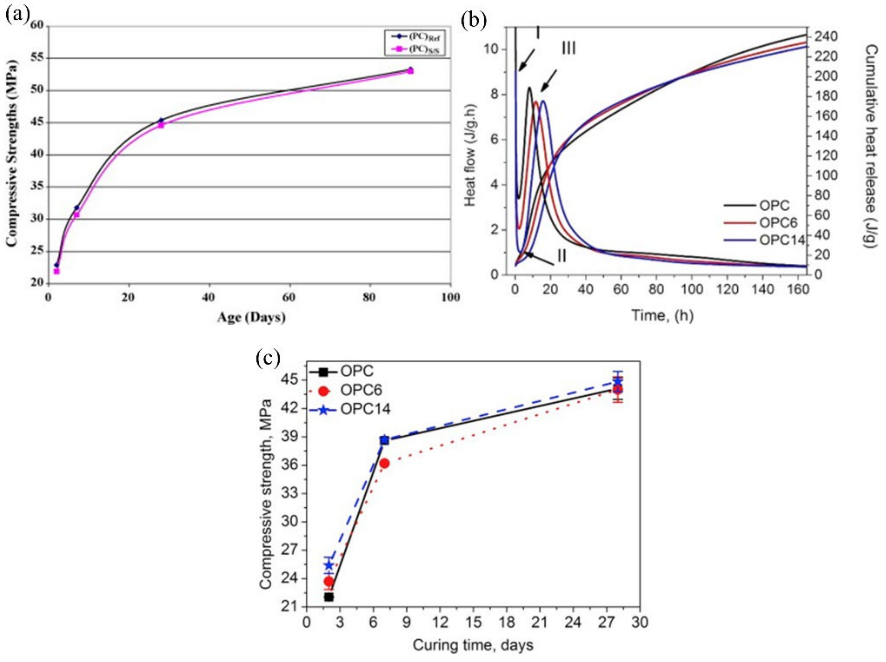

Figure 2. Compressive strength and hydration heat of steel slag-based cement: (a) compressive strength of cement with steel slag, (b) hydration heat of cement with ladle metallurgy stainless steel slag, (c) compressive strength of cement with ladle metallurgy stainless steel slag.

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2.1.4. Ceramic Waste

Ceramic waste, which is generated as scrap during tile production, has a chemical composition (CaO: 6.37 wt%, SiO_2: 68.94 wt%, Al_2O_3: 14.83 wt%, Fe_2O_3: 3.46 wt%, MgO: 1.91 wt%，K_2O: 3.51 wt%，Na_2O: 0.97 wt%) [20] similar to clay and can be used as an alternative raw material. Clinker prepared from 84.84 wt% $CaCO_3$, 13.86 wt% ceramic waste, and 1.3 wt% Fe_2O_3 at 1450 °C for 30 min showed a decrease of 13.24 wt% in C_3S and an increase of 145.24 wt% in C_2S compared to commercial cement clinker, with more abundant C_2S phases observed under optical microscopy (Figure 3). This indicates that ceramic waste addition hindered the transformation of C_2S to C_3S. Additionally, the hydration time of C_3S was delayed by 2 h, and the hydration rate decreased by 22.22%. The compressive strengths at 7 d and 28 d were 40.27 MPa and 68.99 MPa, respectively, meeting the requirements of EN 197-1:2011, but 15.74% and 1.03% lower than those of commercial cement [5]. This is attributed to the fact that the components ZnO, ZrO_2, and B_2O_3 in tile glaze form hydroxides on the surface of the unhydrated cement particles, which hinder the hydration process and reduce the strength of matrix [5, 21].

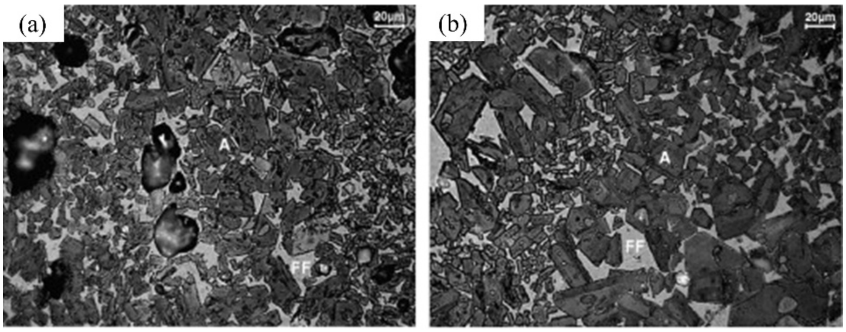

Figure 3. SEM images of ceramic waste cement clinker: (a) reference sample, (b) ceramic waste cement clinker. (A: C_3S, FF: interstitial phase).

The above analysis shows that using appropriate amounts of industrial solid waste to produce cement clinker can promote the phase transformation of calcium silicates. However, impurities such as MgO, ZnO, ZrO_2, and B_2O_3 can partially hinder the hydration of calcium silicates, resulting in less significant improvements in mechanical properties. Nevertheless, the performance is still sufficient for practical applications.

2.2. Natural Idle Resources

2.2.1. Pumice

Pumice, which forms from cooled volcanic lava, has a chemical composition (CaO: 2.16 wt%, SiO_2: 64.7 wt%, Al_2O_3: 12.9 wt%, Fe_2O_3: 7.15 wt%, SO_3: 0.126 wt%) also similar to that of clay. Experiments indicate that clinker produced with a raw material ratio of $CaCO_3$/pumice/Al_2O_3/Fe_2O_3:75.8/22.9/0.65/0.65 wt% contains the same mineral phases as silicate cement clinker, including C_3S, C_2S, C_3A, and C_4AF. When calcined at 1300 °C for 90 min, the compressive strengths of the clinker were 7 MPa, 11 MPa, and 27.5 MPa at 3 d, 7 d, and 28 d, respectively [9].

2.2.2. Basalt

Basalt, the volcanic igneous rock, has high contents of Al_2O_3, Fe_2O_3, and MgO that act as fluxes during calcination, making it a potential substitute. Using 50 wt% weathered basalt (CaO: 10 wt%, SiO_2: 51.43 wt%, Al_2O_3: 15.29 wt%, Fe_2O_3: 13.04 wt%) to replace clay, clinker with C_3S and C_2S contents meeting the standard EN 197-1:2011 was obtained, with a high C_3S content of 59.44 wt% and well crystallinity (Figure 4). The compressive strengths of mortar prepared from this clinker were 20.5 MPa, 35.2 MPa, and 46.7 MPa at 2 d, 7 d, and 28 d, respectively [11]. Using 12 wt% basalt (CaO: 10.29 wt%, SiO_2: 48.04 wt%, Al_2O_3: 17.92 wt%, Fe_2O_3: 13.47 wt%) instead of clay increased the C_3S content by 5.34 wt% and decreased the C_2S content by 5.38 wt% compared to the reference sample using clay. The compressive strengths of cement paste prepared from this clinker were 0.2 MPa and 1 MPa lower than those of the reference with clay at 7 d and 28 d, respectively, and 23.4 MPa and 19 MPa lower than those of industrial cement, respectively [12].

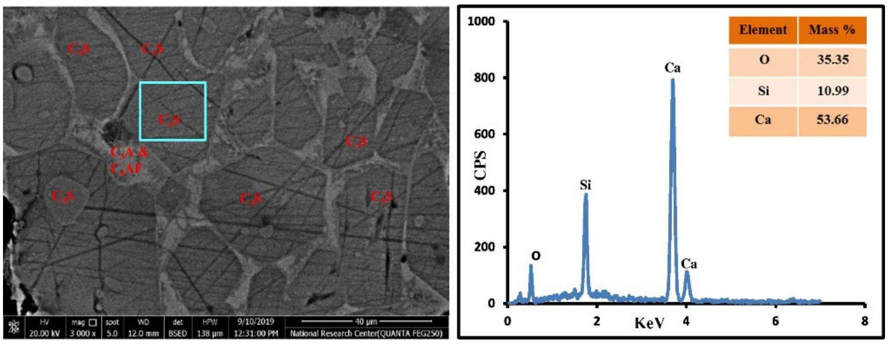

Figure 4. SEM images of basalt cement clinker.

2.2.3. Desert Sand

Desert sand, a fine-grained, chemically stable quartz sand rich in SiO_2, quartz sand, is found in many industrialized countries. Its main chemical composition (CaO: 1.76–3.62 wt.%, SiO_2: 80.01–83.55 wt.%, Al_2O_3: 7.65–9.62 wt.%, Fe_2O_3: 1.87–2.85 wt.%, MgO: 0.92–1.72 wt.%， K_2O: 0.03–0.93 wt.%， Na_2O: 0.08–0.96 wt.%) is similar to industrial quartz. Among these, SiO_2, CaO, Al_2O_3, and Fe_2O_3 play a crucial role in forming useful phases in cement clinker, while Na_2O and K_2O can form a liquid phase at lower calcination temperatures to promote phase transformation. Results showed that with a raw material ratio of desert sand/ $CaCO_3$/ Al_2O_3/ Fe_2O_3: 17.31/77.35/2.56/2.79 wt% to produce cement clinker lowered the phase transformation temperatures of C_2S and C_3S by 9 °C and 6.2 °C, respectively, compared to the reference sample prepared by industrial quartz. Calcining at 1350 °C for 30 min produced clinker with a C_3S content of 60.8 wt%, 9.4 wt% higher than the control sample, and Figure 5a showed uniformly fine distributions of C_2S and C_3S crystal [10]. Additionally, the hydration behavior of desert sand cement is similar to that of silicate cement, while the C_3S hydration time is 4.15 h earlier and the hydration rate is 3.7% faster in desert sand cement. The hardened paste exhibited a dense microstructure (Figure 5b), with strengths increased by 19.06% at 3 d and 17.01% at 28 d [22].

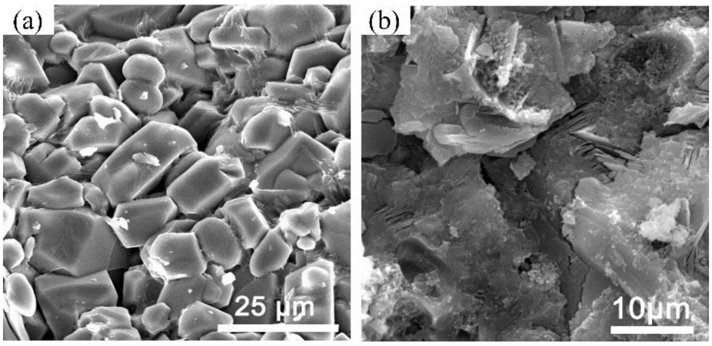

Figure 5. SEM images of desert sand cement clinker and hydration product: (a) clinker, (b) hydration product.

The natural idle resources discussed above, formed from high-temperature cooled melts, exhibit low activation energy during clinker calcination, thereby promoting phase transformation. It is essential to evaluate their chemical suitability for raw materials and to optimize their addition to achieve high-performance clinker.

3. Conclusion

The contradiction between the decreasing mineral resources and the increasing demand for silicate cement, along with issues of land degradation, environmental pollution, and high energy consumption, highlights the necessity of using industrial solid waste and natural idle resources for cement production. Discussions on the clinker calcination, cement hydration, and hardening properties of cement clinker prepared with industrial solid waste and natural idle resources as silica sources have demonstrated their feasibility for cement production. The synthesis of cement clinker using industrial solid waste and natural idle resources notably integrates low-cost green manufacturing, resource and land conservation, environmental protection, and socio-economic sustainability, highlighting its broad prospects and significant potential for cement production.

Future research should focus on the development of industrial solid waste and natural idle resources for cement production, incorporating new technologies and processes to improve cement performance. Moreover, additional study is essential for the mechanisms and control methods of cement synthesis using these materials to establish a comprehensive theory for the green manufacturing of cement.

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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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