

Cementitious and Geopolymer Composites with Lithium Slag Incorporation

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Lithium slag (LS)'s particle size distribution is comparable to fly ash (FA) and ground granulated blast furnace slag (GGBS), which suggests it can enhance densification and nucleation in concrete. The mechanical treatment of LS promotes early hydration by increasing the solubility of aluminum, lithium, and silicon. LS's compositional similarity to FA endows it with low-calcium, high-reactivity properties that are suitable for cementitious and geopolymeric applications. Increasing the LS content reduces setting times and flowability while initially enhancing mechanical properties, albeit with diminishing returns beyond a 30% threshold. LS significantly improves chloride ion resistance and impacts drying shrinkage variably.

Keywords: lithium slag ; cement ; concrete ; composite ; durability ; strength ; hydration ; carbon footprint

1. Introduction

Lithium, known for its exceptional electrochemical properties, is extensively used across various industries, including pharmaceuticals ^[1], lubrication ^[2], glass ^[3], and ceramics ^[4]. The late 1990s marked a significant surge in lithium demand due to its crucial role in portable electronic battery production ^[5]. These batteries have become indispensable for modern life, thereby providing clean energy for smart devices and vehicles ^[6].

During the production of lithium carbonate from spodumene, chemical treatment yields lithium slag (LS) and metal oxides ^{[7][8]}. Approximately 9–10 tons of LS are generated per ton of lithium carbonate produced, thus posing disposal challenges ^{[9][10]}. In 2022, Australia contributed 68,500 tons to global lithium production, and demand has been projected to increase from 114,400 tons in 2022 to 248,400 tons in 2026 ^[11]. Improper disposal of LS could harm the environment due to the leaching of fluorine (F^-) and sulphate (SO_4^{2-}) ions, thereby requiring careful management ^[12].

LS, which is rich in silicon, aluminum, and calcium oxides, has garnered interest as a substitute for ordinary Portland cement (OPC) in concrete production, thereby offering a reduced carbon footprint and costs ^{[13][14]}. However, its sulfuric acid extraction origins with elevated SO_3 levels necessitate caution in its use to maintain cement stability ^{[15][16][17][18]}. Adopting a prudent approach is vital to leverage LS sustainability without compromising the cement matrix.

Considerable research efforts have been dedicated to the utilization of LS in cementitious composites, particularly in China and Australia. Recent investigations conducted by ^{[19][20][21]} have collectively affirmed LS's potential as a supplementary cementitious material and as a partial precursor for the formulation of alkali-activated geopolymers. This is underpinned by the pozzolanic activity exhibited by LS, as well as its capacity to enhance the pore structure of cementitious materials.

To gain insight into the potential applications of LS and to enhance its utilization efficiency, it is imperative to comprehensively understand the physicochemical properties inherent to LS itself. Presently, numerous studies have embarked on an exploration of the physicochemical properties of LS. Research has demonstrated that particle size, chemical composition, and amorphous content (silica and alumina) are key determinants of the pozzolanic activity exhibited by noncrystalline materials ^[22]. Therefore, investigations into the physical characteristics of LS encompass parameters such as the specific surface area (SSA) ^{[14][23]}, specific gravity ^[24], density ^{[25][26]}, moisture content ^[23], and particle size distribution ^{[27][28]}. In parallel, conventional analytical methods have been employed to scrutinize the chemical composition of LS, including X-ray diffraction (XRD) ^{[28][29]} and scanning electron microscopy with energy-dispersive spectroscopy (SEM/SEM-EDS) ^{[28][30][31]}, as well as specialized techniques such as Fourier transform infrared spectroscopy (FTIR) ^{[30][32]}, nuclear magnetic resonance (NMR) ^{[33][34]}, X-ray photoelectron spectroscopy (XPS) ^[33], and thermogravimetry (TG)/thermogravimetric and derivative thermogravimetric analysis (TG-DTG) ^{[19][26][35]}.

Due to its analogous physicochemical properties to SCMs, LS has been widely adopted as a cementitious material additive in mortar and concrete. Extensive research has been conducted on the performance of cementitious composite materials incorporating LS, including aspects such as setting times [36][37], flowability [38], and rheology [16][39]. Furthermore, the mechanical properties of cementitious materials with LS incorporation have been thoroughly investigated, thus encompassing parameters like compressive strength [39][40], tensile strength [41][42], elastic modulus [43], and flexural strength [38].

2. Physiochemical and Microscopic Analysis of LS

2.1. Physical Properties of Raw LS

2.1.1. Particle Size Distribution

PSD is a crucial physical parameter used to assess the activity index of LS. PSD is typically characterized by its 10% (D10), 50% (D50), and 90% (D90) percentiles, representing the respective sizes finer than the nominated percentile. Variations in LS particle sizes are evident across different studies. For instance, the D10 of LS has been observed to range from 0.84 to 2.74 μm , with an average value of 2.01 μm . Similarly, the D50 (mean particle size) exhibited a range of 4.53–30.39 μm and an average value of 17.04 μm . Furthermore, the D90 was reported in the range of 28.00–83.65 μm , with an average size of 58.86 μm . These variations primarily depend on the source of the LS, roasting temperature, and the grinding methods and procedures [44]. Unfortunately, there is limited literature on systematic studies of the grinding methods and grinding parameters that can enhance the reactivity of LS. In response to the inherent characteristics of raw LS particles, which are characterized by their large size and low amorphous content, limited research endeavors have undertaken the examination of the grinding processes, both dry [14] and wet [33][36], to reduce particle size and enhance amorphousness. Wet grinding, in particular, has been effective in producing finely ground LS particles that facilitate ion dissolution. The incorporation of wet-ground LS has shown promise in improving the early strength of sulfoaluminate cement (SAC) [36].

2.1.2. Density, Specific Surface Area, and Moisture Content

In addition to analyzing the PSD of LS, various physical properties, including density, SSA, and moisture content, have also been examined. The SSA of LS, as reported by different researchers [14][25][45], exhibited a range from 265 to 1362 m^2/kg , with an average value of 698 m^2/kg , which is attributed to the porous surface texture of LS [46]. It is generally finer than the SSA of cement (350 m^2/kg), fly ash (350 m^2/kg), and GGBFS (400 m^2/kg) [47][48][49]. The density of LS was found to fall within the range of 2480–2500 kg/m^3 [26][50]. The moisture content in LS varied from 2.5% to 28.3%, with an average value of 17.5% based on different studies [30][46]. These physical properties of LS are closely linked to the preparation methods and grinding parameters of LS. To achieve good quality control and high use efficiency of LS, further research into the manufacturing methods and production parameters of LS is essential.

2.2. Chemical Properties of LS

2.2.1. Chemical Composition

The chemical composition analysis of LS serves to provide fundamental data for assessing its potential as a cementitious material. Numerous studies have investigated the chemical composition of LS through XRF testing. The analysis reveals that the primary chemical components of LS are SiO_2 , Al_2O_3 , and CaO . SiO_2 and Al_2O_3 are predominantly present in the form of $\text{H}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 4\text{SiO}_2$ (aluminum silicate), while CaO primarily exists as CaSO_4 (anhydrous gypsum) [51]. Furthermore, LS exhibits a notable sulfur trioxide (SO_3) content, thereby typically existing in the form of $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, which, upon reaction with calcium hydroxide, can lead to expansion. This limitation prevents the substantial inclusion of LS as a mineral admixture [52].

2.2.2. XRD Results

XRD analysis provides a quantitative assessment of the mineral composition of LS. According to references [30][37][53], the primary mineral phases in LS have been identified through XRD patterns, as are illustrated in **Figure 1**. These phases mainly include gypsum, quartz, and lithium aluminum silicate.

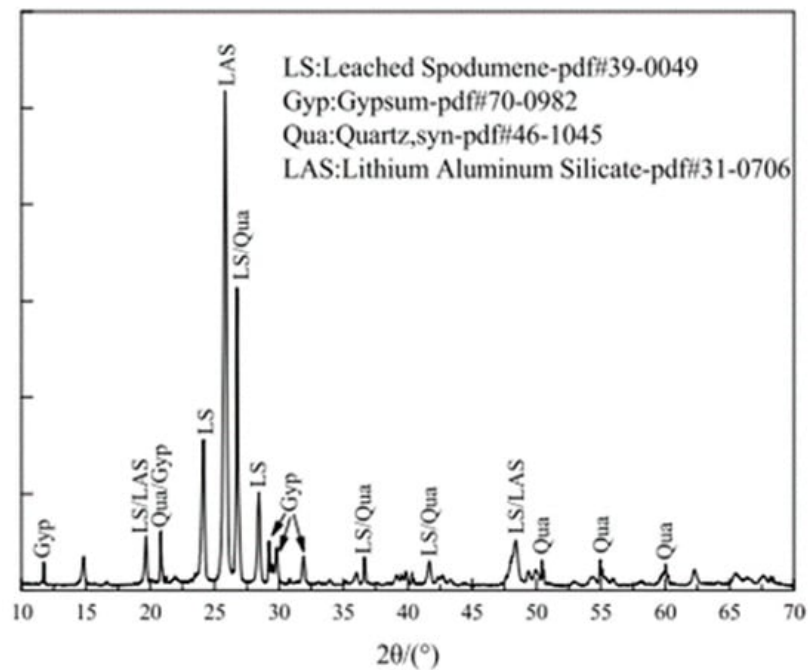


Figure 1. XRD patterns of LS powder [30].

Quantitative analysis conducted by Rahman et al. [19] revealed the relative compositions of the crystalline mineral phases in LS. The analysis indicated that LS comprises 2.1% β -spodumene ($\text{LiAlSi}_2\text{O}_6$), 6.6% bassanite ($\text{CaSO}_4 \cdot 0.5\text{H}_2\text{O}$), 5.4% calcium magnesium carbonate ($\text{Ca}_{0.845}\text{Mg}_{0.155}(\text{CO}_3)$), 23.8% quartz (SiO_2), 28.2% anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$), 2.3% albite ($\text{NaAlSi}_3\text{O}_8$), and 31.6% amorphous phase.

2.3. Microscopic Analysis of LS

2.3.1. SEM-EDS Analysis

Javed et al. [29] employed SEM-EDS to explore the phase and morphological change between raw LS and calcined LS. LS comprises angular particles that are rich in aluminosilicate minerals, while the prismatic/elongated particles consist of gypsum, as is evidenced by the SEM/EDS analysis depicted in **Figure 2a**. The EDS points B and C on the micrograph exhibited peaks corresponding to aluminum, silicon, and oxygen EDS spectra, thereby confirming the presence of aluminosilicate particles. Conversely, EDS analysis of the prismatic particles shown in **Figure 2b** displayed peaks of calcium, sulfur, and oxygen, thus indicating the presence of gypsum. This observation supports the notion of sulfation occurring during lithium extraction in refineries [54]. These prismatic particles possess a size exceeding 50 μm . Through the calcination of LS, the aluminosilicate components undergo a transformation into agglomerated amorphous (glassy) phases due to sintering. Particle fragmentation is a consequence of the lithium refining process being applied to spodumene ore, which induces a certain level of reactivity within LS. Notably, the crystalline phase transformation within LS initiates from temperatures exceeding 800 $^\circ\text{C}$ [55]. Thus, a calcination temperature of 700 $^\circ\text{C}$ is considered to be suitable for the production of amorphous aluminosilicates [29].

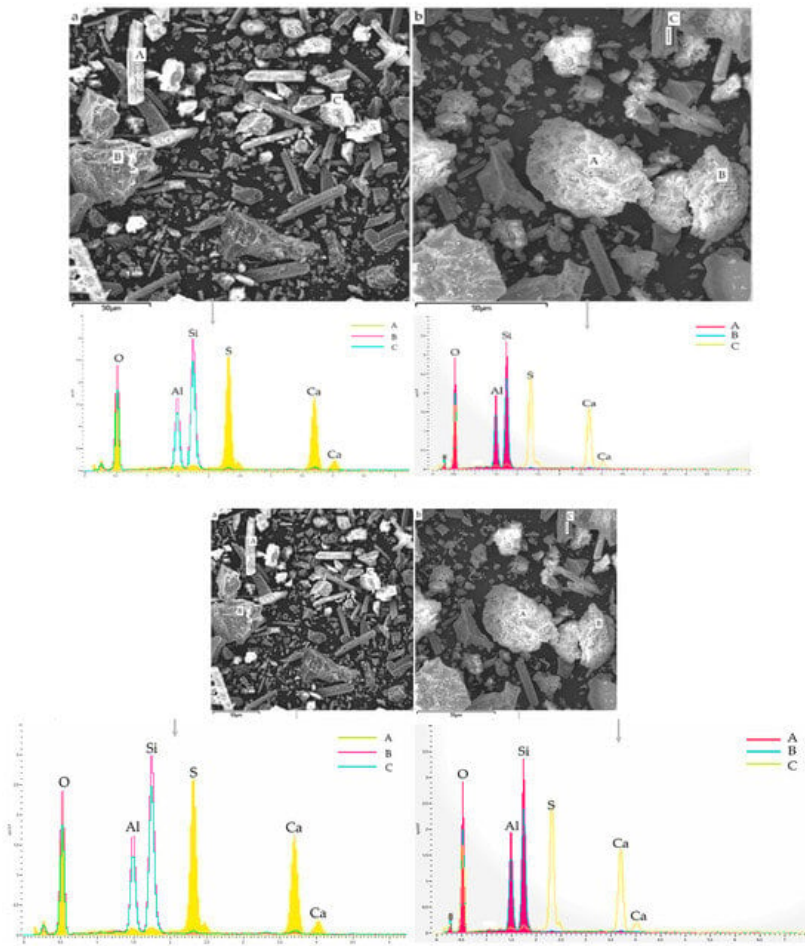


Figure 2. SEM-EDS results: (a) raw LS and (b) calcined LS [29].

2.3.2. TG-DTG Analysis

TG-DTG testing provides valuable insights into the thermal behavior of LS. It allows researchers to understand how LS reacts to changes in temperature, thus helping to identify important thermal transitions and decomposition processes. TG-DTG curves for LS compared with cement were obtained by Rahman et al. [19], as are depicted in **Figure 3**.

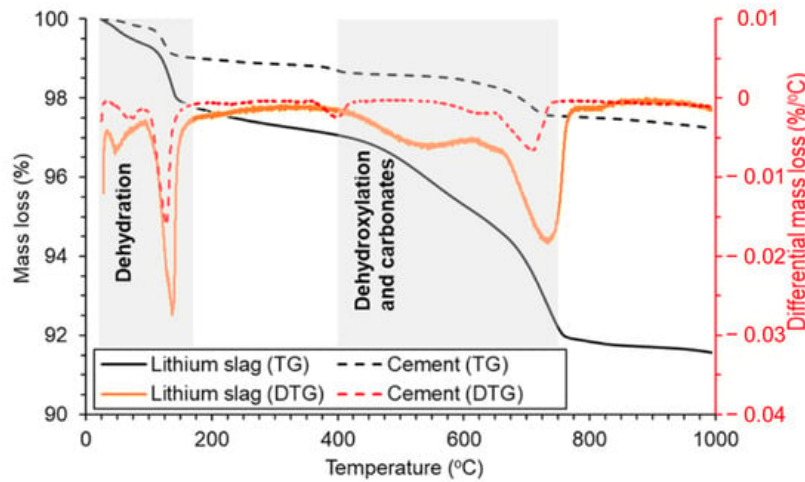


Figure 3. TG-DTG curves of LS and cement from room temperature to 990 °C [19]. Note: the shaded areas in the TG-DTG curves show the mass loss due to presence of moisture and gypsum, portlandite, and carbonates, respectively.

The observed mass loss in the TG-DTG curves of the LS, occurring between room temperature and 170 °C, primarily resulted from the removal of moisture and bassanite water [56]. It is evident that the moisture content of LS was notably higher in comparison to conventional cement. Subsequent mass losses in the temperature range of 400–750 °C can be attributed to the dehydroxylation of the zeolite phases [57] and the decomposition of carbonates [58]. These processes contribute to the reduction in the crystallinity of pozzolans. During this stage, there is a substantial decrease in the mass of LS, thus indicating a continuous augmentation of the amorphous phases within LS, which can significantly enhance its pozzolanic reactivity [59]. The most optimal temperature for enhancing the reactivity of LS is approximately around 700 °C

[29][32]. It is noteworthy that the higher moisture content in LS compared to cement, coupled with the more pronounced amorphous reactions, resulted in a total mass loss of the LS reaching 8.4% at 990 °C.

2.3.3. NMR and XPS Analysis

NMR patterns provide insights into the chemical environment surrounding silicon-oxygen tetrahedra, while XPS analysis yields binding energies that also reflect the chemical environment of the atoms. Therefore, Tan et al. [33] employed microscopic techniques such as XPS and NMR to investigate the chemical composition changes in LS as it transformed into micro-LS through wet grinding.

3. Fresh State Properties of Cementitious Composites with LS Incorporation

3.1. Setting Time

Figure 4 compiles data from various references on the changes in initial and final setting times with increasing LS content. The setting time ratio is defined as the ratio of the setting time of the cementitious composite with added LS to the setting time of the corresponding sample without LS (control group), with the subsequent parameter ratios described later (including flowability, strength characteristics, chloride resistance, and shrinkage) being defined in the same manner. It is evident that the majority of cementitious composites incorporating LS exhibited a setting time ratio of less than one, thereby indicating that the addition of LS reduces the setting time of cementitious composites.

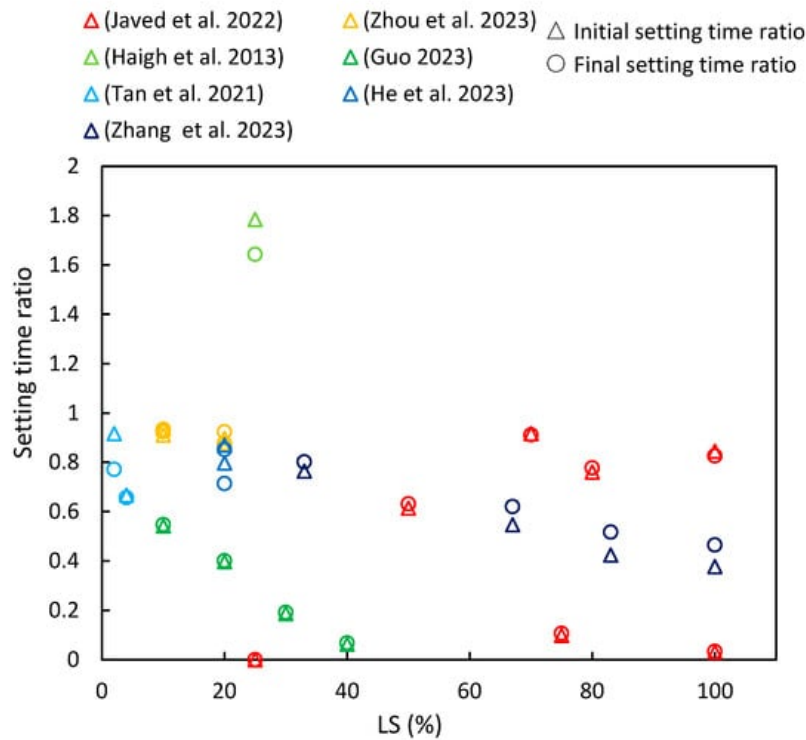


Figure 4. The setting time ratios of cementitious composites with varying levels of LS substitution [16][23][29][53][60][61][62].

Zhang et al. [60] observed that as the proportion of LS replacing LP increased, both the initial and final setting times of the paste decreased. In LS-UHPC (ultra-high-performance concrete), the increased formation of ettringite (Aft) with higher LS ratios contributed to reduced setting times by facilitating the solid network's overlap reduction [63]. Javed et al. [29] investigated the setting times of geopolymer pastes containing LS and increasing amounts of FA. Their findings indicated an increase in both the initial and final setting times with higher FA replacement. The abrupt setting observed in the LS geopolymer was attributed to false setting due to the presence of over 5% gypsum/anhydrite in the LS. This phenomenon was linked to the precipitation of interlocked needle-shaped gypsum (anhydrite), which contributed to self-desiccation in the geopolymer paste matrix due to reduced water content in the aluminosilicate gel [64].

Furthermore, various researchers have conducted in-depth investigations into the influence of LS on the setting time in special cementitious systems. Guo et al. [16] conducted a study in which ternary alkali-activated materials (AAMs) were synthesized using a combination of LS, metakaolin (MK), and GGBS. They explored the impact of LS replacing MK on the setting time. As the LS content increased, there was a significant reduction in the setting time. This phenomenon was

attributed to the LS requiring less time for the dissolution of (Si, Al)O₄ tetrahedra and Ca²⁺ ions in the same alkali activator, thereby creating more favorable conditions for polymerization and resulting in a shortened setting time.

3.2. Flowability

The flowability of fresh paste is primarily characterized through slump tests and flow table tests. **Figure 5** summarizes the flowability ratios of cementitious composites with varying proportions of LS based on different literature sources. It is evident from most of the literature that flowability decreases with an increase in LS content. Specific details of these changes are as follows.

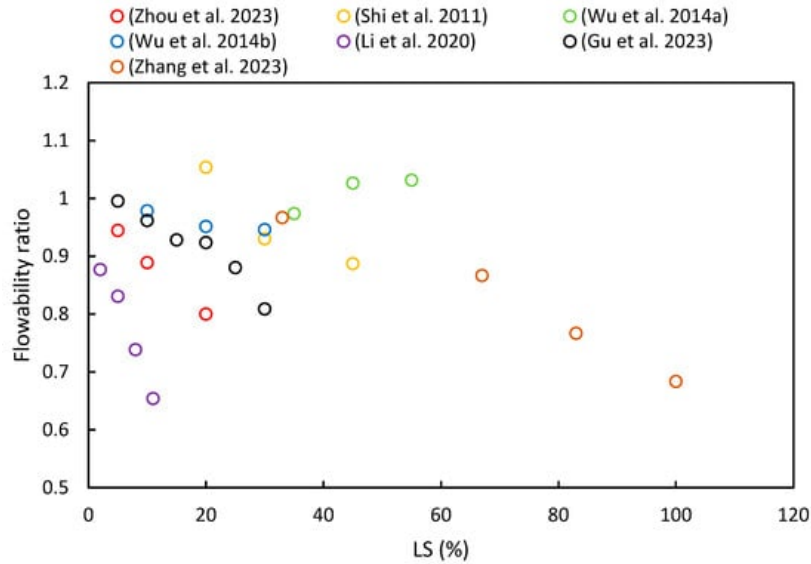


Figure 5. The flowability ratios of cementitious composites with varying proportions of LS [38][41][42][60][61][65][66].

Zhang et al. [60] noted that the addition of LS reduced the flowability of UHPC due to its irregular shape and strong water absorption. Additionally, LS led to increased Aft formation in the initial stages of hydration, which has been attributed to its high sulfate and aluminate content [30].

However, some studies have shown different patterns. Wu et al. [41] conducted research on concrete slumps using water-to-binder ratios ranging from 0.27 to 0.35 and a ternary combination of 25–65% OPC, 15–35% LS, and 20–40% SS content. The sand ratio and superplasticizer dosage were adjusted to maintain the concrete mixture's slump within the range of 190 ± 20 mm. Due to the numerous variables involved, the slump values did not exhibit a certain trend with the LS contents.

3.3. Rheology

Rheology, which is a branch of science focused on the deformation and flow of matter, explores the relationships between the stress, strain, and shear rate. In the context of cementitious materials, the rheological behavior of fresh mixtures plays a significant role in determining their optimal mixing, casting, and stacking properties. This is especially crucial in specialized construction techniques like 3D printing and self-compacting concrete [67][68][69].

He et al. [53] noted that an increased dosage of synthetic calcium silicate hydrates–polycarboxylate (CSH-PCE) led to a notable enhancement in the rheological properties of fresh LS–cement paste. This enhancement was evident in decreased viscosity and yield stress. It resulted from the physically combined PCE in C-S-H-PCE, which could dissolve in the solution and then adsorb onto minerals, thereby improving the flowability of the fresh paste. However, the addition of TEA had a negative impact on the rheological properties of the LS–cement paste. The incorporation of 1% C-S-H-PCE into the LS–cement binder, along with increasing the TEA content (from 0% to 0.5%), progressively increased the yield stress and viscosity values of the fresh binder.

4. Mechanical Properties of Cementitious Composites with LS Incorporation

4.1. Compressive Strength

Figure 6 summarizes the findings from numerous studies investigating the compressive strength ratios under different age and LS substitution conditions. A recurring pattern in most of the literature indicates that as the LS content increases,

the compressive strength ratio initially rises before gradually declining, thereby implying an optimal LS content. He et al. [24] observed that an LS content exceeding 20% had an adverse effect on the compressive strength of concrete, while Luo et al. [20] noted that the geopolymer compressive strength significantly improved with LS-based geopolymer contents below 70%. Furthermore, as the curing period increases, there is a growing trend in compressive strength ratios. This phenomenon can be attributed to the late-stage pozzolanic activity of LS.

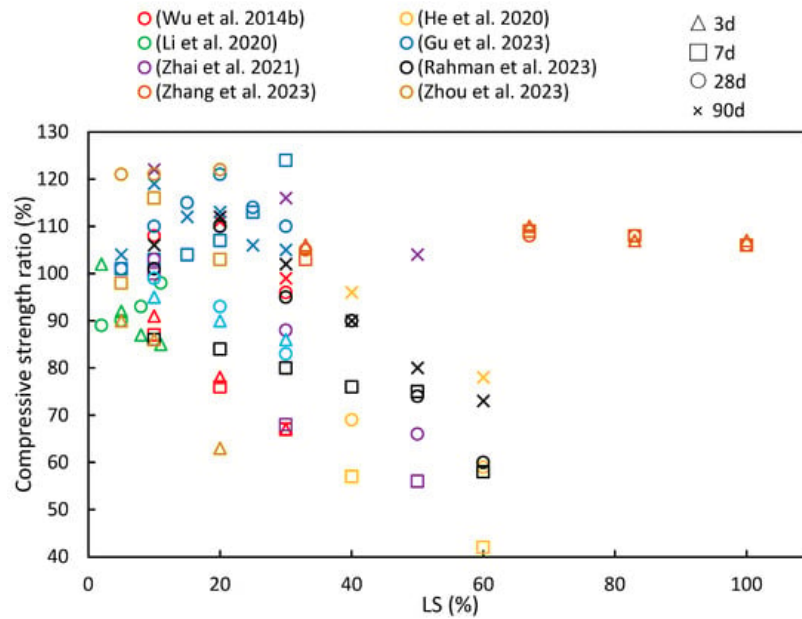


Figure 6. The compressive strength ratios of cementitious composites at various ages and levels of LS substitution [19][30][38][60][61][66][71][72].

In the study by Zhou et al. [61], experiments were conducted to evaluate the reactivity of LS and SS as SCMs. The addition of 10% LS powder resulted in substantial hydration product formation at 28 days. LS exhibited a superior advantage over SS in enhancing the paste compressive strength. The pozzolanic reactions of LS and SS contributed to the development of the compressive strength at later stages. Additionally, in an effort to enhance the performance of ultra-high-performance concrete containing limestone powder (LP-UHPC), Zhang et al. [60] explored the partial replacement of LP with LS. The compressive strength of the LP-UHPC initially decreased and then increased with increasing LS content at all ages. Replacing 5% or 10% of the LP with LS led to improved compressive strength in the LP-UHPC across all age groups. However, the pozzolanic reaction of the LS exhibited limited the activity at the early stages of hydration.

4.2. Flexural Strength

Figure 7 provides a summary of the flexural strength ratios reported in various studies at different levels of LS incorporation and testing ages. The general trend of the flexural strength ratios closely resembles that of compressive strength. In most cases, the strength ratio tends to increase initially and then decrease with increasing LS content, with early-age strength ratios being slightly lower than later-age ratios. This behavior can be attributed to several factors: Firstly, when a small quantity of LS (e.g., 10% or 20%) is mixed with cement paste, it is initially considered inert, thereby not actively participating in the early-age hydration reactions. This results in a higher retention of water, thereby promoting the formation of gel and subsequently increasing the strength. Additionally, LS contains gypsum, which enhances the reactivity of silica and alumina, thereby leading to the production of more C–S–H and hydrated calcium aluminate.

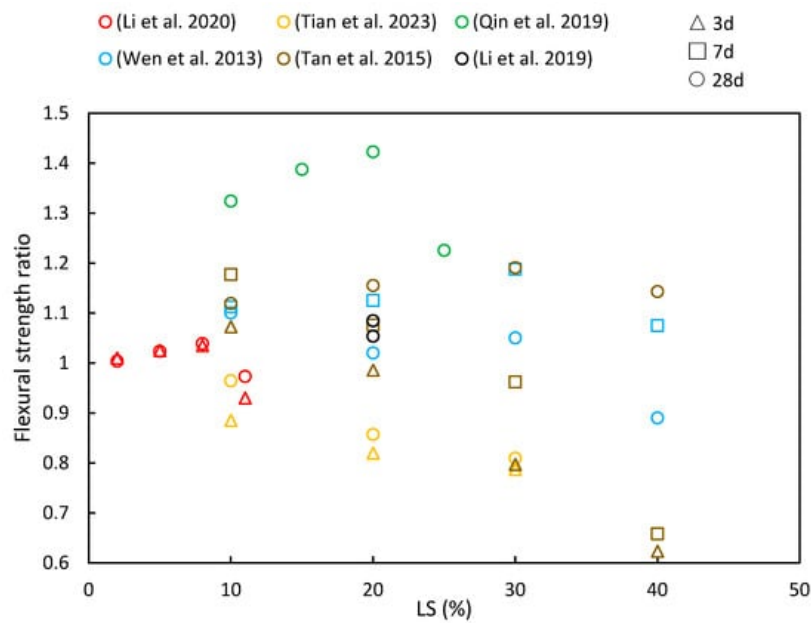


Figure 7. The flexural strength ratios of cementitious composites at various ages and LS substitution ratios [14][35][38][40][73][74].

4.3. Splitting Tensile Strength and Elastic Modulus

The splitting tensile strength ratios of cementitious composites at various ages and LS substitution ratios are depicted in **Figure 8**. It is evident from various literature sources that as the LS content increases, both increases and decreases in the splitting tensile strength ratios have been observed. However, with an increase in the curing period, the splitting tensile strength ratios were shown to consistently rise. At 28 days, the cementitious composites incorporating LS outperformed the control groups in the following studies, thereby highlighting the favorable long-term pozzolanic activity of LS. Qin et al. [40] employed LS derived from industrial waste to replace cement, thus aiming to enhance concrete's mechanical properties. The results indicated that the splitting tensile strength initially increased and then decreased with a rising LS content, thereby reaching its peak improvement when the LS content substitution was at 25%.

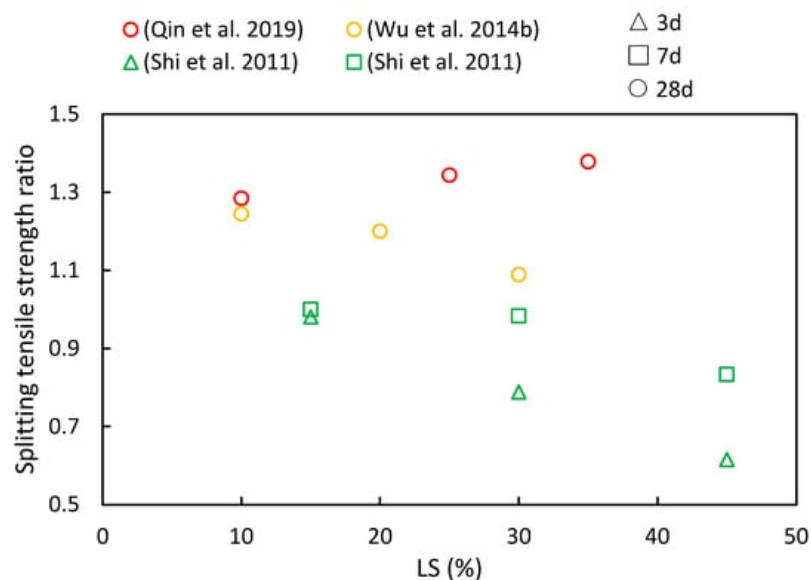


Figure 8. The splitting tensile strength ratios of cementitious composites at various ages and LS substitution ratios [40][42][66].

5. Durability of Cementitious Composites with LS Incorporation

5.1. Chloride Resistance

Figure 9 summarized the impacts of varying the LS content and the age of LS-based cementitious composites on chloride ion migration. The consistently observed chloride resistance ratio below one indicates that the LS enhanced the chloride ion migration resistance of cementitious systems. Furthermore, with an increase in the composite age, the resistance of

the cementitious system to chloride ions has been shown to strengthen. Specific reasons for these findings are elucidated upon synthesizing the results from diverse studies.

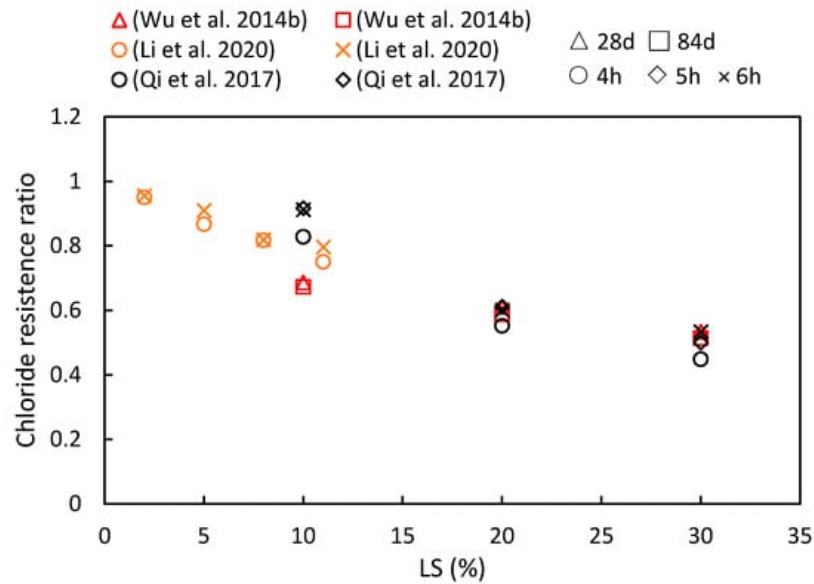


Figure 9. The chloride resistance ratios of cementitious composites at various ages and LS substitution ratios [38][66][75].

5.2. Shrinkage

Figure 10 presents a compilation of the shrinkage ratios in cementitious materials with varying LS contents and different ages, as have been reported in various studies. It is evident that increasing the LS content generally reduces the concrete's shrinkage values. However, with the progression of age, the pattern of drying shrinkage exhibits various trends. The specific research findings are detailed below. Qi et al. [75] investigated concrete prepared by replacing cement with LS at ratios of 10%, 20%, and 30%. They found that the drying shrinkage remained steady, with a slow increase observed after 60 days. As the LS content increased, the drying shrinkage values decreased.

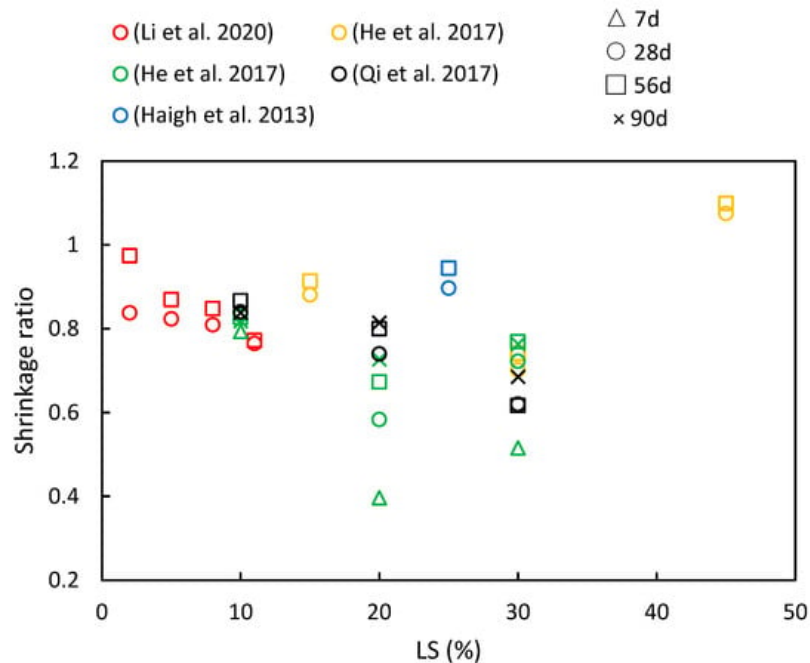


Figure 10. A compilation of shrinkage ratios in cementitious composites with varying LS contents and different ages [23][38][39][75][76].

5.3. Sulfate Attack and Carbonation

The investigation of sulfate attack and carbonation in cementitious composites with LS incorporation is vital for assessing their durability. Understanding how LS affects resistance to sulfate attack and carbonation is crucial for ensuring the long-term performance and sustainability of these materials when they are subjected to different exposure conditions.

Li et al. [35] conducted a study involving 20% LS mortar samples that were subjected to wet curing and steam curing, which was then followed by partial immersion in a 99% pure sodium sulfate solution for 720 days. Their findings indicate that, regardless of the initial curing conditions, the presence of LS enhances mortar properties in the face of sulfate attack. In addition to the role of leached $\text{LiAlSi}_2\text{O}_6$, the pozzolanic reaction of LS reduces the calcium hydroxide content within the mortar. Moreover, steam-cured LS mortar exhibited superior sulfate resistance compared to steam-cured PC mortar.

6. Chemical and Microstructural Investigations of Cementitious Composites with LS Incorporation

6.1. Hydration Heat

The evolution of hydration heat plays a fundamental role in the initial setting, hardening, and ultimate strength development of cement blends. The effective control of hydration heat is imperative to ensure the overall quality and durability of concrete structures. Upon contact with water, cementitious materials undergo a series of chemical reactions, thereby resulting in a distinct hydration heat evolution curve that typically encompasses several stages, including the initial period, induction period, acceleration period, deceleration period, and retardation period [77][78][79].

He et al. [80] noted that the incorporation of 20 wt% LS into a composite binder led to a notable reduction in the evolution of hydration heat and a significant delay in the induction period when compared to pure cement paste. This phenomenon can be attributed to the reduced clinker content resulting from the substitution of Portland cement with LS. In addition, during the early stages of hydration, LS was found to exhibit inert behavior and exhibited limited participation in the initial hydration reactions.

6.2. Pore Structure

The construction of pores, specifically porosity and pore size distribution, significantly influences concrete properties [81]. He et al. [39] examined the impact of LS on pore size distribution at 7 days and 90 days, which was measured via MIP. As the age increased, the pore size and volume decreased. The introduction of LS initially increased the porosity of larger pores. However, including 10% and 20% LS in the binder refined the pore structure in later stages. In addition, He et al. [74] observed that replacing 40% of their cement sample with LS reduced the porosity in the later curing stages, thereby correlating with improved mechanical properties. However, the use of 60% LS increased the total porosity, particularly for pores exceeding 100 nm in diameter. This was attributed to the incomplete stimulation of the LS pozzolanic reaction due to the limited calcium hydroxide amount generated during Portland cement hydration.

6.3. XRD Analysis

XRD analysis provides insights into the crystalline phases within samples. Zhou et al. [61] generated XRD patterns for specimens containing LS and SS following standard curing for 7 days, as are illustrated in **Figure 11**. The primary mineral composition of the cementitious materials was identified as portlandite, calcite, C_3S , $\text{Ca}_2\text{Fe}_2\text{O}_5$, $\text{LiAlSi}_2\text{O}_6$, and ettringite. Notably, the presence of $\text{LiAlSi}_2\text{O}_6$ was detected by a prominent peak between $2\theta = 25\text{--}30^\circ$, which was observed in the samples with the 10% LS replacement due to the introduction of spodumene ($\text{LiAlSi}_2\text{O}_6$) from the LS. The diffraction peak intensity of the lithium pyroxene was low, thereby indicating a predominance of amorphous Al_2O_3 and SiO_2 within it [82]. Furthermore, because spodumene was an inert component in the raw LS material and exhibited limited participation in the hydration process, traces of it persisted in the LS-mixed samples.

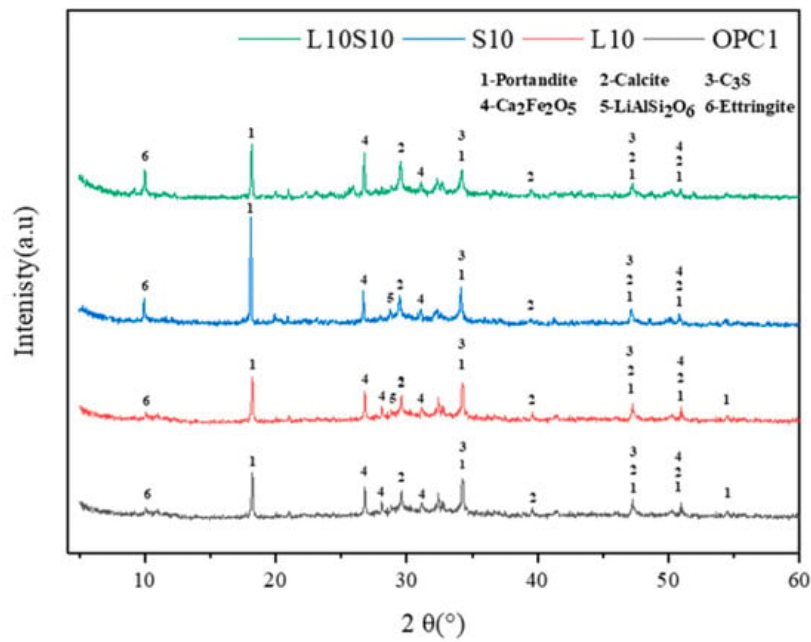


Figure 11. XRD patterns of cement pastes [61].

6.4. SEM Analysis

Investigating the microstructure of cementitious materials aids in explaining the mechanical strength of the matrix, as well as the extents and modes of reaction of different raw materials, thus elucidating the reaction mechanism of LS within the matrix. In the study conducted by Gu et al. [72], it was found that in the C-LS-SS cementitious system that SS and LS replaced 10% and 20% of the cement, respectively (10SS-20LS), $\text{LiAlSi}_2\text{O}_6$ and RO phases were also observed, and the layered spodumene ($\text{LiAlSi}_2\text{O}_6$) in the LS underwent partial leaching, as is depicted in **Figure 12a**. With an increase in the LS content, there was a significant consumption of portlandite, thereby leading to a reduction in the degree of LS reaction. In addition, the addition of SS had a detrimental impact on the pore structure of the pure cement system, thereby increasing the number of pores and fractures, which is consistent with previous findings [83]. However, the addition of SS-LS improved the pore structure of the composite system, thereby resulting in a denser surface. Notably, evident voids and large cracks were observed in the hydration products of 5SS-25LS (SS and LS replacing 5% and 25% of the cement, respectively) shown in **Figure 12b**. These results indicate the formation of delayed ettringite due to the high SO_3 content in the LS [84], which subsequently led to the development of microcracks.

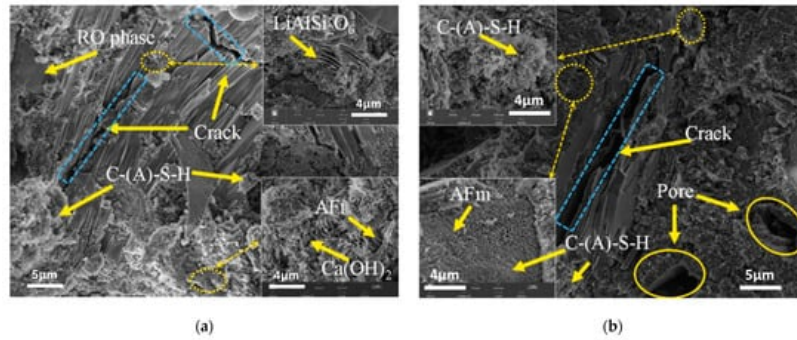


Figure 12. SEM images of (a) 10SS-20LS and (b) 5SS-25LS at 28 d [72].

6.5. FTIR and TG Analysis

SCMs were produced by blending SS and LS, and their synergistic effect was investigated by Gu et al. [72]. The characterization of the hydration products was conducted using FTIR, as is depicted in **Figure 13**. The faint peaks at 3644 cm^{-1} originated from the bending vibration of the OH groups in portlandite [85]. These spectral peaks associated with portlandite exhibited a reduction in intensity with an increasing LS content. Moreover, the absorption peaks at approximately 978 cm^{-1} were linked to the asymmetric stretching vibrations of Si–O–Si (Si–O–Al) related to C–S–H gels [86]. Among the composite systems, the 10SS-20LS group exhibited the highest peak strength, thus slightly surpassing that of the PC and signifying that the SS-LS demonstrated more pronounced pozzolanic activity at suitable proportions.

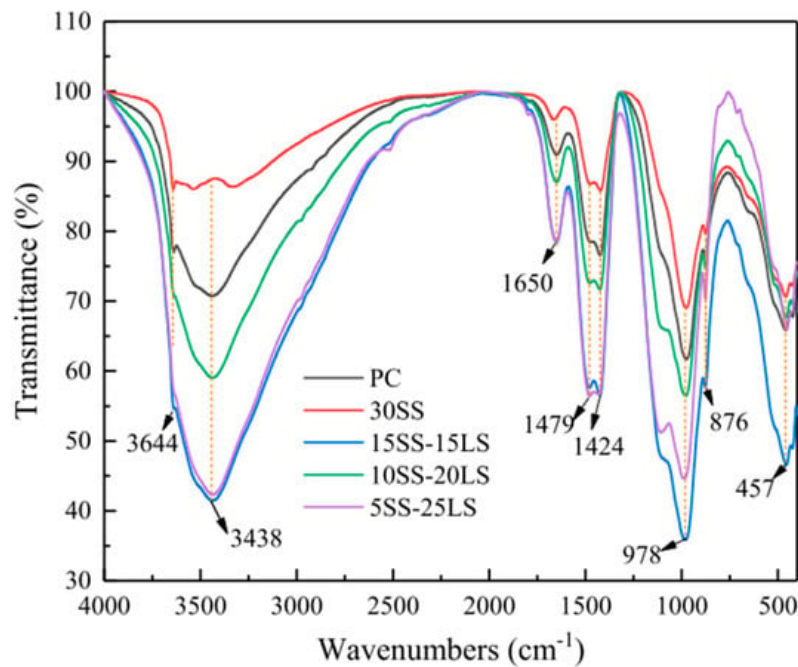


Figure 13. FTIR spectra of pastes at 28 d [72].

7. Cost, Energy, and Carbon Emission Comparisons

The ineffective disposal of LS can pose a serious environmental threat due to the leaching of fluorine and sulfate ions, which can lead to pollution of both land and water. Therefore, it is imperative to explore methods for the disposal and efficient utilization of LS to support the sustainable development of the lithium carbonate and construction materials industries. In order to facilitate the widespread application of LS in the construction materials sector, it is equally important to assess its commercial viability.

It is evident that LS has the lowest cost (10 USD/t) compared to other construction materials. Not only does LS exhibit similar pozzolanic activity to FA, but it also comes at just one-fifth of the price of FA. This makes LS an economically attractive option for concrete or geopolymer production, thereby significantly reducing manufacturing costs. However, it is worth noting that Ali et al. [5] reported the cost index values for a novel LS-based geopolymer, and it is interesting to observe that the geopolymer with a higher LS content resulted in higher cost index values. This is attributed to the very low strength of the binders prepared from higher LS contents. Furthermore, it is important to mention that the cost index values for all geopolymer mixes are higher than the 1.75 USD/m³.MPa cost index value reported for a previously studied OPC mix [87]. The higher cost of geopolymer binders can be attributed to the expensive sodium silicate and recent price increases in industrial waste materials (FA and slag) due to government-imposed limitations on coal and steel production.

Regarding the energy index, Guo et al. [16] demonstrated that the energy consumption of LS (2230 MJ/t) is slightly lower than that of MK (2500 MJ/t), thereby implying that replacing MK with LS can reduce energy consumption to some extent [16]. In AAM pastes with LS replacing MK, the higher strength of LS-containing AAM compared to AAM without LS does not provide a clear advantage in terms of the energy index. However, this substitution strategy holds strong potential for improving the construction environment and conserving natural resources.

8. Conclusions

(1) The PSD of LS closely resembles that of FA and GGBS. This similarity suggests that LS can exhibit similar effects related to densification and nucleation when integrated into concrete, thus resembling the behavior of FA and other SCMs. Mechanical treatment of LS enhances the dissolution of aluminum, lithium, and silicon in LS, thereby expediting early hydration in LS–cement systems.

(2) LS exhibits variations in SiO₂ + Al₂O₃ and Ca/(Si + Al) within the ranges of 70.29–80.77% and 0.02–0.14%, respectively. This composition aligns LS with FA, which is characterized by high SiO₂ and Al₂O₃ contents and a low CaO content. This similarity categorizes LS as a low-calcium precursor with chemical reactivity akin to that of FA.

(3) In most of the literature examined, an increase in LS content was shown to lead to a reduction in the initial and final setting times of LS–cement and LS–geopolymer systems. Moreover, the studies determined that flowability decreased with an increase in LS content due to its irregular shape, strong water absorption characteristics, and elevated formation

of AfT in the initial stages of hydration.

(4) As LS content increases, the compressive strength, flexural strength, and splitting tensile strength ratios initially increase, with diminishing returns beyond a 30% threshold. This suggests an optimal LS content for achieving favorable mechanical properties. Additionally, with longer curing periods, there is a noticeable upward trend in the compressive strength, flexural strength, and splitting tensile strength ratios.

(5) LS plays a crucial role in enhancing chloride ion migration resistance and reducing shrinkage in cementitious systems. Furthermore, as the composite ages, the resistance of the cementitious system to chloride ions becomes more robust. However, the behavior of drying shrinkage exhibits various trends.

(6) The mechanisms through which LS operates within cementitious composites can be classified into three main categories. Firstly, there is the filling effect: the fine-grained nature of LS improves particle packing, and its fine particles act as pore blockers, thereby reducing interconnectivity between pores and effectively lowering porosity. Secondly, there is the pozzolanic effect: LS reacts with calcium hydroxide to generate additional hydration products, thereby refining large pores and bridging the gap between the paste and aggregates. Thirdly, there is the nucleation effect: LS provides nucleation sites, thereby promoting the preferential production and development of hydration products in these specific locations.

(7) LS not only exhibits similar pozzolanic activity to FA, but it also comes at just one-fifth of the price of FA. This makes LS an economically attractive option for concrete or geopolymer production, thereby significantly reducing manufacturing costs. Moreover, the embodied CO₂ and embodied energy of LS are comparable to GGBS, slightly higher than those of FA and SF, and significantly lower than those of MK, LP, and cement. Therefore, LS exhibits the potential for solid waste recycling and sustainable development.

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