

Hydrothermal Synthesis Of Calcium Silicate Hydrate From Waste Glass: Effect Of CaO/SiO₂ Ratio On Phase Formation And Properties

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Environmental pollution has become increasingly severe due to multiple contributing factors, among which the rapid growth of industrial waste is particularly significant. In practice, the glass industry generates substantial waste daily, primarily in the form of grinding glass powder. While cullet can be recycled through remelting, the ground glass powder is often directly discharged into the environment or disposed of in landfills. Therefore, the treatment and valorization of industrial waste, particularly glass powder, have become research topics for mitigating environmental pollution. In this study, waste glass was used as a silica source to synthesize calcium silicate hydrate and investigate the effect of the CaO/SiO₂ ratio on phase formation. Waste glass powder was mixed with lime, pelletized, and subjected to hydrothermal treatment at 180 °C for 10 hours. The formation of Xonotlite and Tobermorite was confirmed by XRD, FTIR, SEM, and EDX analyses, demonstrating the feasibility of synthesizing calcium silicate hydrate from waste glass powder. Mechanical and physical properties, including diametral tensile strength, water absorption, and bulk density, indicated that the optimal CaO/SiO₂ ratio for synthesizing calcium silicate hydrate from waste glass is 0.9.

Keywords: Calcium silicate hydrate; CaO/SiO₂ ratio; Waste glass; Hydrothermal synthesis; Recycled materials

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1. Introduction

The recycling of waste glass has become increasingly important for environmental protection and sustainable development in the materials industry. In principle, glass can be recycled through remelting to produce new glass products. Commercial glass is typically manufactured from silica sand, ash, limestone, soda, and various additives. The incorporation of recycled glass into glass production offers significant benefits, including energy savings [1], reduced production costs, and decreased consumption of natural raw materials [2] while also mitigating environmental impacts [3]. Previous studies have shown that each kilogram of recycled glass used in production can reduce CO₂ emis-

sions by approximately 31 g [4]. Additionally, the inclusion of recycled glass improves energy efficiency by 2.5 - 3.0% for every 10 wt.% added [5].

However, in practice, waste glass often contains impurities such as metals, ceramics, organic matter, or mixed glass types, making direct recycling through remelting challenging. Consequently, alternative recycling approaches to valorize waste glass have attracted increasing attention. Due to its high SiO₂ content and amorphous structure, which confer high reactivity, waste glass is considered a promising silica source for synthesizing value-added materials, particularly calcium silicate hydrate (CSH).

CSH is widely studied and applied, mainly existing in two crystalline forms: Xonotlite and Tobermorite. These

materials have attracted considerable interest due to their superior properties and broad applications. In concrete, CSH plays a crucial role in cement hydration and is the primary phase responsible for concrete's mechanical strength [6]. Additionally, crystalline CSH phases, such as Xonotlite and Tobermorite, have been investigated for wastewater treatment due to their ability to adsorb heavy metal ions, such as Cr(III) and Cu(II) [7]. Notably, Xonotlite exhibits a fibrous structure with thermal stability up to approximately 1000 °C, making it suitable for thermal insulation and refractory applications [8].

CSH can be synthesized through the reaction between SiO_2 and CaO under appropriate conditions. Temperature and pressure are key factors influencing the synthesis process, as they govern the reaction kinetics between CaO and SiO_2 . Temperature, in particular, plays a critical role in phase transformation and crystallinity. At relatively low temperatures (150 - 180 °C), the product is mainly a poorly crystalline CSH gel. When the temperature increases to 180 - 220 °C, the formation of crystalline phases such as Tobermorite and Xonotlite is significantly enhanced [9]. Reaction time also strongly affects crystallinity and crystal size [10]. Furthermore, the initial shaping method can influence reaction kinetics, crystal morphology, phase evolution, and the final mineral composition. Therefore, optimizing these parameters is essential for controlling CSH formation in the CaO - SiO_2 system.

Recent studies have investigated the influence of the CaO/SiO_2 ratio on the structure and mechanical properties of CSH. Pelisser et al. (2012) reported that increasing the Ca/Si ratio significantly reduces the elastic modulus and hardness due to changes in silicate chain structure [11]. Wu et al. found that higher C/S ratios increase CaO conversion but decrease crystallinity and formation rate of CSH [12]. Other studies indicate that lower C/S ratios (0.8 - 1.0) promote the formation of highly crystalline Tobermorite and Xonotlite, thereby improving mechanical properties [13]. However, when waste glass is used as the silica source, most studies focus on hydrothermal conditions and adsorption properties, while the influence of the C/S ratio on phase formation and physico-mechanical properties remains limited [14–17].

Furthermore, the relationships among phase evolution, microstructural development and physico-mechanical properties of waste glass derived CSH have not been fully clarified. Therefore, this study aims to systematically investigate the effect of the CaO/SiO_2 ratio on phase composition, morphology, water absorption, bulk density and mechanical strength under identical hydrothermal conditions. The findings provide further insight into the for-

mation behavior of calcium silicate hydrate synthesized from recycled glass and contribute to the development of sustainable value-added materials from waste resources.

From the above review, it can be observed that although the effects of parameters such as temperature, pressure, and reaction time on the formation of CSH have been extensively investigated, studies on the role of the CaO/SiO_2 ratio in the formation of Xonotlite and Tobermorite phases - particularly with respect to mechanical properties such as bulk density, water absorption, and diametral tensile strength of CSH materials synthesized from waste glass - remain relatively limited. Therefore, this study aims to fill this gap by investigating the effect of the CaO/SiO_2 ratio on phase formation and properties of CSH synthesized from waste glass under hydrothermal conditions at 180 °C for 10 hours. The optimal ratio is determined through structural analyses (X-ray Diffraction, Fourier Transform Infrared Spectroscopy, Scanning Electron Microscopy, and Energy-dispersive X-ray spectroscopy) combined with mechanical and physical property evaluations, including strength, water absorption, and bulk density.

2. Experimental method

2.1. Materials

The primary raw materials used in this study were waste glass (WG) and calcium oxide (CaO). Waste glass was collected from glass-processing factories and preliminarily cleaned to remove mechanical impurities such as dust, metals, and organic contaminants adhering to the surface. The cleaned glass was then ground using a ball mill for an appropriate duration to obtain fine powder. The resulting glass powder was dried at 105 °C for 24 hours to eliminate residual moisture. After drying, the powder was sieved through a 90 μm mesh to ensure a relatively uniform particle size, facilitating solid-phase reactions in the CaO - SiO_2 system.

The chemical composition of the waste glass was determined by X-ray fluorescence and is presented in Table 1. The results indicate that SiO_2 is the dominant component, along with minor oxides such as CaO , Al_2O_3 , and alkali metal oxides. The high SiO_2 content and amorphous structure make waste glass a promising silica source for CSH synthesis.

Calcium oxide (CaO) powder was used as the calcium source. Prior to use, the CaO powder was dried and sieved through a 125 μm mesh to ensure uniform particle size and enhance the reaction surface area. Under hydrothermal conditions, CaO reacts with SiO_2 from waste glass to form calcium silicate hydrate phases.

Table 1. Chemical composition of waste glass.

SiO ₂	CaO	Na ₂ O	MgO	Al ₂ O ₃	K ₂ O	Fe ₂ O ₃	SO ₃	LOI
73.38	10.48	9.41	0.55	1.05	0.38	0.20	4.31	0.25

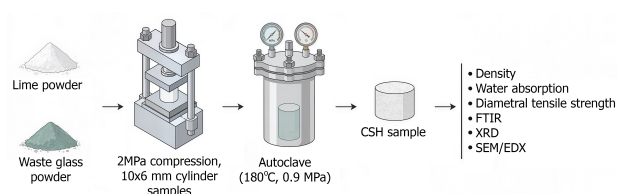
2.2. Sample Preparation

The sample preparation procedure is illustrated in Fig. 1. Initially, waste glass powder was homogeneously mixed with CaO powder at different CaO/SiO₂ molar ratios (denoted C/S). The batch compositions and sample designations are presented in Table 2. The variation of the C/S ratio was designed to investigate the effect of CaO content on phase formation and material properties. The selected C/S ratio range (0.8-1.2) was based on the reported phase stability diagrams showing the formation domains of Tobermorite and Xonotlite according to the Ca/Si ratio [15, 18]. Previous studies have shown that Tobermorite is the predominant phase at relatively low Ca/Si ratios whereas increasing the Ca/Si ratio promotes the formation and stabilization of Xonotlite [13, 15]. Therefore, the investigated compositions were selected the Tobermorite-Xonotlite transition region, allowing the influence of the C/S ratio on phase evolution, microstructure, physical and mechanical properties to be evaluated.

Table 2. Ingredient composition of samples.

Samples	Glass content (wt.%)	CaO content (wt.%)	C/S ratio
R1.2	58.81	41.19	1.2
R1.1	61.50	38.50	1.1
R1.0	64.39	35.61	1.0
R0.9	67.52	32.48	0.9
R0.8	70.90	29.10	0.8

After determining the batch composition, an amount of water equivalent to 9% of the dry mixture mass was added to provide sufficient moisture for shaping. The mixture after water addition was mixed uniformly for 5 minutes to ensure homogeneous distribution of components.

**Fig. 1.** A diagram of the prototyping process.

The mixture was then pressed into shape using a steel mold to form cylindrical samples measuring 10 mm in diameter and 6 mm in height. The pressing process was

conducted at 2 MPa to create initial bonding between powder particles and to ensure stable shape and dimensions before hydrothermal treatment.

The pressed samples were then placed in a 250 mL Teflon autoclave containing 45 mL of water. The hydrothermal process was conducted at 180 °C and 0.9 MPa for 10 hours. No stirring was performed during the hydrothermal treatment. These hydrothermal conditions were selected based on previous studies that reported that temperatures around 180 °C and reaction times of approximately 10 hours are favorable for the crystallization of calcium silicate hydrate phases, particularly Tobermorite and Xonotlite [9, 15]. To isolate the effect of the CaO/SiO₂ molar ratio on phase evolution, microstructure, physical and mechanical properties, all other synthesis parameters, including hydrothermal temperature, reaction time, water content, pressure, and forming conditions, were kept constant throughout the study. No stirring was performed during the hydrothermal treatment. Under the high-temperature, high-pressure conditions of the hydrothermal environment, the reaction between CaO and SiO₂ is promoted, leading to the formation of CSH mineral phases. After the process was completed, the samples were naturally cooled to room temperature before further characterization.

2.3. Characterization Methods

After hydrothermal treatment, the material samples were analyzed using various methods to evaluate their structure, phase composition, and physical properties.

The crystalline phase analysis was performed using X-ray diffraction (XRD) with a Bruker D8 Advance instrument. The diffraction patterns were recorded in the 2θ range from 5° to 75° with a step size of 0.02027°. The XRD results were used to identify the mineral phases formed during hydrothermal synthesis and to evaluate phase evolution. Semi-quantitative phase analysis was performed using the integrated areas of characteristic diffraction peaks assigned to Tobermorite and Xonotlite after background correction with Origin software. Only the principal reflections of each phase were selected, and overlapping peaks were deconvoluted prior to integration. The semi-quantitative phase ratio (%A) was calculated according to Eq. (1), where SA is the integrated area of the characteristic peaks of phase A and ΣS is the total area of the XRD

pattern.

$$\% A(\%) = \frac{S_A}{\sum S} \times 100\% \quad (1)$$

In addition, the average crystallite size was estimated from the selected characteristic diffraction peaks using the Scherrer equation (Eq. (2)), where D is the average crystallite size (nm), K is the shape factor (taken as 0.9 [19]), λ is the wavelength of the X-ray used (nm), β is the full width at half maximum (FWHM) after correction (rad), and θ is the Bragg diffraction angle ($^\circ$).

$$D = \frac{K \times \lambda}{\beta \times \cos \theta} \quad (2)$$

The lattice parameters (a , b and c) were calculated from selected characteristic diffraction peaks using Bragg's law (Eq. (3)) and the crystallographic relationships of the monoclinic crystal system (Eq. (4)). The unit cell volume (V) was estimated using the Eq. (5), where a , b and c are lattice parameters ($\text{\AA}$), β is the monoclinic angle ($^\circ$) and V is the unit cell volume ($\text{\AA}^3$) [20, 21].

$$n\lambda = 2d \sin \theta \quad (3)$$

$$\frac{1}{d^2} = \frac{1}{(\sin \beta)^2} \left(\frac{h^2}{a^2} + \frac{k^2(\sin \beta)^2}{b^2} + \frac{l^2}{c^2} - \frac{2hl \cos \beta}{ac} \right) \quad (4)$$

$$V = abc \sin \beta \quad (5)$$

Fourier Transform Infrared Spectroscopy (FTIR) analysis was performed on a Nicolet 6700 instrument over $4000 - 450 \text{ cm}^{-1}$ to identify characteristic functional groups of CSH materials, particularly Si – O – Si and Si – O – Ca bonds in the silicate structure.

The morphology and microstructure of the samples were observed using scanning electron microscopy (SEM) combined with energy-dispersive X-ray spectroscopy (EDX) on a JSM-IT 200 instrument at an accelerating voltage of 10 kV. The obtained results allow evaluation of crystal morphology, microstructure, and elemental distribution in the material.

In addition to structural analyses, physical properties, including bulk density, water absorption, and splitting tensile strength, were determined. Bulk density and water absorption were measured according to ASTM C20-00 using Eqs. (3) and (4). For each C/S ratio, six specimens were tested. Results are reported as mean $\pm$ standard deviation, and differences among samples were analyzed by one-way ANOVA ($p < 0.05$). The diametral tensile strength (DTS) was determined according to ASTM C1323-10 and calculated using Eq. (5), where B is bulk density (g/cm^3); W_d is dry mass (g); S is saturated mass (g); M is suspended mass; V is bulk volume (cm^3); W is water absorption (%); DTS is

splitting tensile strength (MPa); P is applied load (N); D is sample diameter (mm); and L is sample height (mm).

$$B = \frac{W_d}{V - (S - M)} \quad (6)$$

$$W = \frac{S - W_d}{W_d} \times 100 \quad (7)$$

$$\text{DTS} = \frac{2 \times P}{\pi \times D \times L} \quad (8)$$

The combined use of structural analysis and physical property testing enables evaluation of the effect of the CaO/SiO₂ ratio on phase formation and properties of CSH materials synthesized from waste glass. From this, the optimal C/S ratio can be determined based on the correlation between crystal structure, microstructure, and physico-mechanical properties.

3. Results and discussion

3.1. Effect of CaO/SiO₂ Ratio on Mechanical and Physical Properties

The results in Fig. 2 show that the DTS of the material varies significantly with the C/S ratio, reaching a maximum at C/S = 0.9 and decreasing as the C/S ratio further increases. This trend directly reflects changes in the bonding structure of the CSH phase at the atomic and nanoscale.

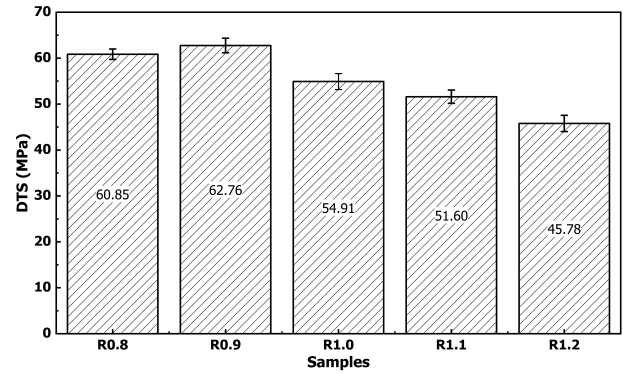


Fig. 2. Effect of the C/S ratio on the DTS of the samples after hydrothermal treatment.

One-way ANOVA confirmed that the C/S ratio significantly affected the measured properties ($p < 0.05$). From the perspective of silicate structure, CSH is described by a silicate chain model with Q^n units ($n = 0 - 4$), where n represents the number of bridging oxygen atoms between $[\text{SiO}_4]^{4-}$ tetrahedra. At low C/S ratios (0.8-0.9), the system tends to form $Q^2 - Q^3$ units, corresponding to long silicate chains with a high degree of polymerization [22, 23]. In this case, Ca^{2+} acts as a charge-balancing ion without significantly disrupting the silicate network, allowing the

formation of highly ordered Tobermorite structures. The continuous silicate network enables efficient stress transfer, resulting in the highest mechanical strength at $C/S = 0.9$.

In contrast, when the C/S ratio increases (>1.0), the excess Ca^{2+} increases the density of nonbridging oxygen sites, leading to a structural shift from $Q^2 - Q^3$ to $Q^1 - Q^2$ units [22, 24]. This reduces the length of silicate chains and disrupts the continuity of the bonding network, thereby decreasing the material's load-bearing capacity. At the same time, the formation of secondary phases, such as $Ca(OH)_2$ or a Ca-rich CSH gel with low structural order, further weakens the bonding system [25]. These results are consistent with nanoindentation studies showing that the elastic modulus and hardness of CSH decrease with increasing C/S ratio [26]. Fig. 3 shows the physical properties includ-

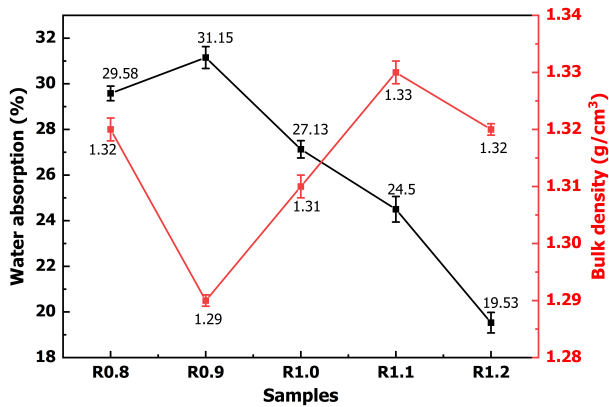


Fig. 3. Water absorption and bulk density of the samples after hydrothermal treatment.

ing water absorption and bulk density. Statistical analysis using one-way ANOVA indicated that the differences among the investigated compositions were significant ($p < 0.05$). The bulk density of the samples ranges from 1.29 to 1.33 g/cm^3 , while the water absorption of the hydrothermally treated samples ranges from 19.53% to 31.15%. Water absorption reaches its maximum and bulk density its minimum at $C/S = 0.9$, which may be associated with the development of an interconnected, open-pore system. This result may be related to the coexistence of CSH gel (which forms mesopores) and crystalline Tobermorite (which forms the structural framework). As the C/S ratio increases, fibrous Xonotlite contributes to a denser microstructure by filling pores and reduce open porosity, thereby decreasing water absorption [27].

Another important factor is the role of Na_2O in waste glass. Na^+ ions act as network modifiers, reducing the activation energy required to break $Si - O - Si$ bonds, thereby increasing the dissolution rate of silica in the initial stage

[28, 29]. This promotes the formation of CSH.

3.2. Phase Composition and Structural Analysis

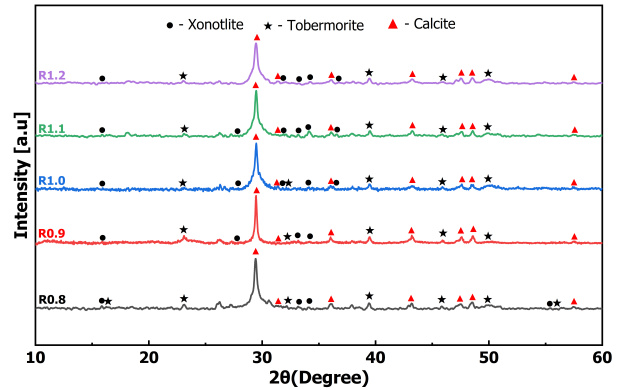


Fig. 4. XRD patterns of the samples after hydrothermal treatment.

To clarify the nature of the variation in physical properties with the C/S ratio as presented in Figs. 2 and 3, XRD (Fig. 4), phase content (Fig. 5, and FTIR (Fig. 6) analyses were employed.

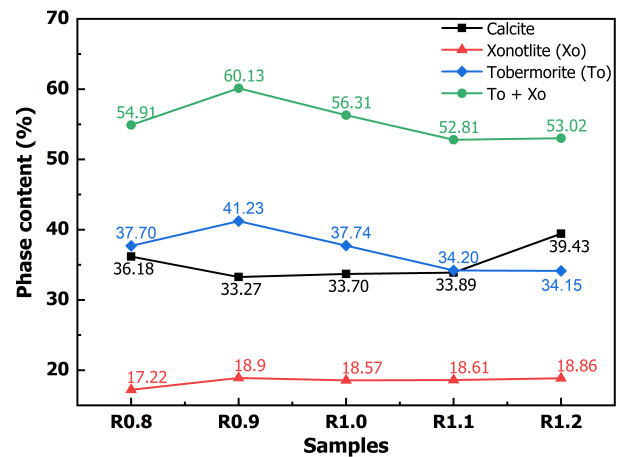


Fig. 5. Relative phase of the samples after hydrothermal treatment calculated from XRD patterns.

The X-ray diffraction patterns (Fig. 4) show the formation of main phases, including Tobermorite, Xonotlite, and Calcite, in all samples. Specifically, the characteristic diffraction peaks of Tobermorite appear at 2θ angles of approximately $16.2^\circ, 23.5^\circ, 29.0^\circ, 32^\circ, 39.8^\circ, 49.4^\circ, 45.3^\circ, 49.5^\circ$, and 55.8° [30], consistent with JCPDS Card No. 45-1480. For Xonotlite, the characteristic peaks are observed at $2\theta \approx 15.6^\circ, 27.5^\circ, 31.7^\circ, 33.1^\circ, 36.3^\circ$, and 55.4° [31], corresponding to JCPDS Card No. 23-0125. In addition, peaks at $2\theta \approx 29.4^\circ, 31.4^\circ, 36.0^\circ, 43.2^\circ, 47.5^\circ, 48.5^\circ$, and 57.4° [32] are

assigned to Calcite (JCPDS Card No. 05-0586), indicating the presence of carbonation products. Furthermore, the relative intensities of diffraction peaks vary with the C/S ratio. When the C/S ratio increases from 0.8 to 0.9, the intensity of Tobermorite peaks increases significantly, indicating an increase in the crystallinity of this phase. However, when the C/S ratio continues to increase, the intensity of Tobermorite decreases while the peaks of Xonotlite become sharper and more intense, reflecting a shift in phase dominance. This result is more clearly quantified in Fig. 5.

In Fig. 5, the relative phase of Tobermorite reaches a maximum at $C/S = 0.9$ and then gradually decreases, while Xonotlite increases almost linearly with increasing C/S ratio. This variation can be explained in terms of thermodynamic equilibrium and the chemical conditions of the system. At low C/S ratios, the system is relatively rich in SiO_2 , favoring the formation of long silicate chains and highly polymerized Tobermorite structures. As the Ca^{2+} concentration increases, the system becomes Ca-rich, reducing the degree of polymerization and promoting the formation of Xonotlite—a phase that is more stable under Ca-rich and high-temperature conditions [33–35]. Thus, the C/S ratio directly controls the phase balance between these two phases.

To further support the above observations, the FTIR spectra suggest changes in bonding structure (Fig. 6). The main absorption band at $\sim 970 - 990\text{ cm}^{-1}$ is assigned to the stretching vibration of Si – O bonds in Q^2 units (silicate chains) [36]. As the C/S ratio increases, this band shifts toward lower wavenumbers, which may indicate a decrease in the degree of silicate polymerization. At the same time, the increase in the relative intensity of the band at $\sim 850 - 900\text{ cm}^{-1}$ (Si – O – Ca bonds) reflects the increasing role of Ca in the network structure [37]. The bands at $\sim 450 - 520\text{ cm}^{-1}$ (Si – O – Si bending) [36] and 3400 cm^{-1} (–OH) [38] indicate the presence of hydrated structures characteristic of CSH. The combination of FTIR and XRD suggests that increasing the C/S ratio reduces silicate polymerization, which may be associated with a phase transformation from Tobermorite (rich in $Q^2 - Q^3$) to Xonotlite (rich in $Q^1 - Q^2$). The proposed interpretation is consistent with the observed phase evolution, although further detailed structural characterization would be necessary to validate the distribution of Q^n silicate species.

In addition to phase composition, the crystallite size (Fig. 7) shows that Tobermorite crystallites reach a maximum at $C/S = 0.9$ and then decrease with increasing C/S. In contrast, the crystallite size of Xonotlite increases with increasing C/S ratio. This indicates that at $C/S = 0.9$, the nucleation and growth processes of Tobermorite reach

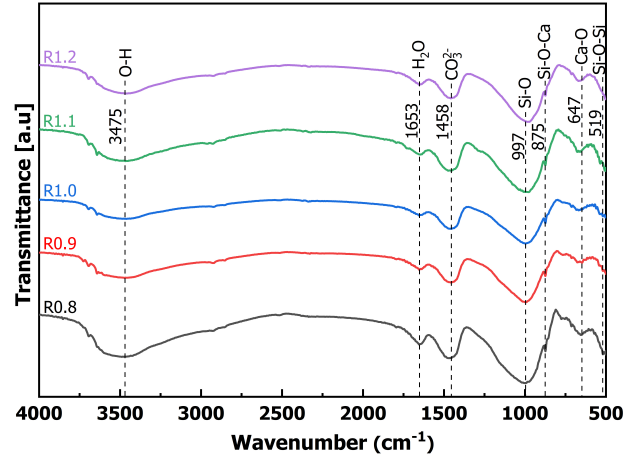


Fig. 6. FTIR spectra of the samples after hydrothermal treatment.

an optimal state, where the crystal growth rate exceeds the nucleation rate. As the C/S ratio increases, the higher concentration of Ca^{2+} accelerates the nucleation rate of Xonotlite, resulting in the formation of more nuclei with initially smaller sizes. However, since Xonotlite becomes the dominant phase, these crystals continue to grow, leading to an increase in crystallite size [39]. The decrease in Tobermorite crystallite size indicates that phase transformation limits its growth. The correlation between phase content (Fig. 5) and crystallite size (Fig. 7) shows that at $C/S = 0.9$, not only the content but also the crystallite size of Tobermorite reaches the highest value among the total crystalline phases. This confirms that this condition is optimal for crystallization and structural development of CSH, thereby directly explaining the highest mechanical strength observed in Fig. 2.

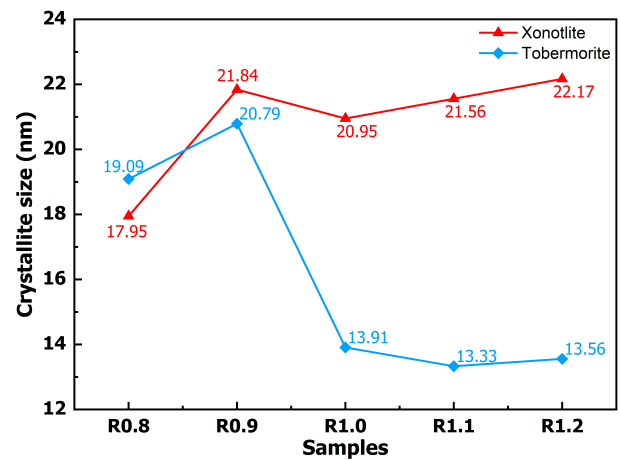


Fig. 7. Crystallite size of Tobermorite and Xonotlite.

Additionally, the presence of Calcite may influence crystallite size through growth inhibition or pore-filling effects. Overall, the results from Figs. 4 to 7 indicate that the C/S ratio not only controls phase composition but also affects bonding structure, crystallization kinetics, and crystallite size. This multiscale interaction ultimately determines the material properties, as shown in Figs. 2 and 3. The results in Table 3 show that only slight variations in the lattice constants of Xonotlite and Tobermorite are observed with changing CaO/SiO₂ ratio, indicating that the crystal structures of both phases remain relatively stable under the investigated hydrothermal conditions. These small changes may be attributed to variations in the degree of silicate polymerization and the local coordination environment within the CSH structure. This observation is consistent with previous studies, which reported that the Ca/Si ratio mainly influences the atomic arrangement, phase stability and phase evolution of calcium silicate hydrates rather than causing significant changes in the crystal structure of individual phases [13, 22, 23].

3.3. Microstructure and Crystal Growth Mechanism

As shown in Fig. 8, the morphological change exhibits a transition that is dictated by the C/S ratio. At lower C/S ratios (0.8-0.9), the microstructure is dominated by interconnected Tobermorite plates with observable lateral dimensions in the sub-micrometer range. These stacked plate-like networks provide multi-directional stress distribution, driving the mechanical properties. The crystallite size results (Fig. 7) show that Tobermorite reaches its maximum size at C/S = 0.9, indicating optimal growth conditions under this composition. Upon increasing the C/S ratio to 1.1-1.2, a 1D fibrous Xonotlite network is evident [40, 41]. While the high density and overlap of these ultra-fine, needle-like fibers preclude a strictly isolated statistical measurement of individual aspect ratios via 2D image analysis, the micrographs illustrate that these fibers extend over several micrometers while maintaining their diameters.

This morphological shift corroborates the XRD crystallite size analysis (Fig. 7), which indicates that the localized growth rate of Xonotlite along its principal crystalline axis is accelerated under Ca-rich conditions, leading to elongated fibrous frameworks that inherently increase the internal void volume between clusters. As the C/S ratio increases, nucleation of Xonotlite increases, leading to the formation of numerous smaller crystals with a dense distribution [41]. This microstructural difference directly affects material properties. The layered structure forms a compact bonding network, whereas the fibrous structure tends to create voids between fibers, especially under low

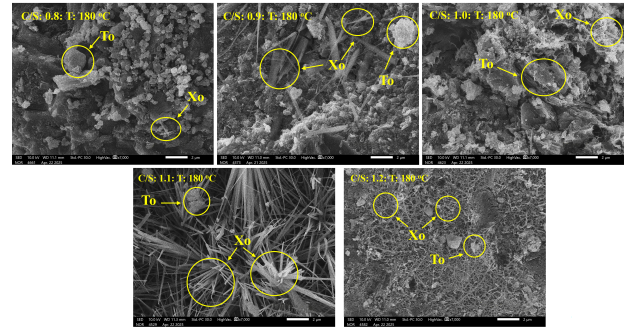


Fig. 8. SEM images of mineral phases of the samples after hydrothermal treatment.

compaction pressure. Consequently, the progressive increase in Xonotlite content is accompanied by a reduction in mechanical strength.

3.4. Elemental Composition and Distribution

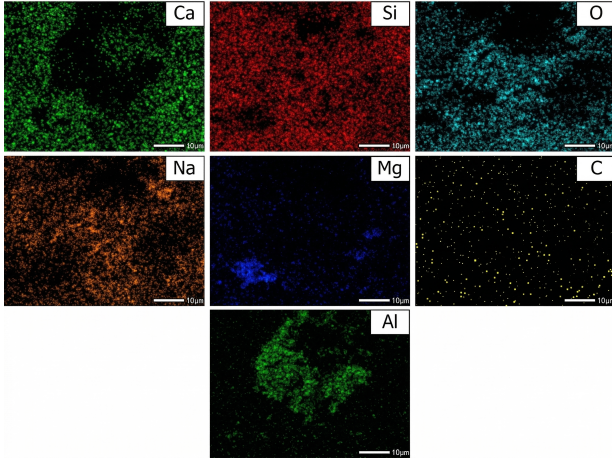
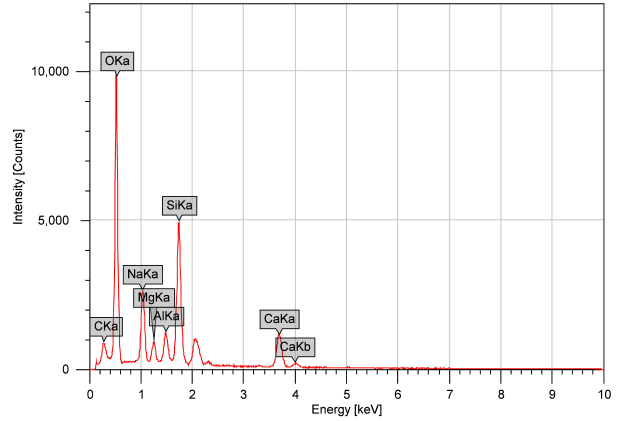
To complement phase and microstructure analyses, the EDX method was used. EDX mapping results (Fig. 9) show that Ca, Si, and O are relatively uniformly distributed across the sample surface at C/S = 0.9. This uniform distribution indicates that the hydrothermal reaction proceeded effectively via a dissolution-precipitation mechanism. Specifically, amorphous SiO₂ from waste glass dissolves to form silicate ions in solution, which then react with Ca²⁺ to form CSH gel. This gel subsequently restructures and crystallizes into stable phases such as Tobermorite or Xonotlite [42].

The EDX spectrum (Fig. 10) confirms that Ca, Si, and O are the dominant elements, consistent with the CSH composition. Minor elements such as Na, Mg, Al, and C reflect the initial composition and impurities present in the waste glass. These elements not only act as impurities but may also influence reaction kinetics. Specifically, Na⁺ can enhance silica dissolution by breaking the silicate network, while Mg²⁺ may participate in intermediate phases or retard crystallization [43].

The uniformity of elemental distribution plays an important role in material properties. A uniformly distributed Ca – Si – O network ensures continuity of the bonding phase, thereby enhancing the mechanical strength of CSH materials. The combined results from EDX, XRD, and SEM demonstrate that a relatively homogeneous material has been achieved in terms of both composition and structure, particularly at the optimal C/S ratio (C/S = 0.9). Overall, the results demonstrate that the C/S ratio plays a critical role in determining the phase change, microstructure, and resulting properties of CSH synthesized from waste glass. The correlation among phase composition (Tobermorite-

Table 3. Lattice parameters (a , b , c , β) and unit cell volume (V) of Xonotlite and Tobermorite at different CaO/SiO_2 ratios.

Sample	Lattice parameters (Å)									
	Xonotlite					Tobermorite				
	$a(\text{Å})$	$b(\text{Å})$	$c(\text{Å})$	$\beta(\text{Å})$	$V(\text{Å})$	$a(\text{Å})$	$b(\text{Å})$	$c(\text{Å})$	$\beta(\text{Å})$	$V(\text{Å})$
R0.8	16.87	7.31	7.14	90.44	880.48	11.25	7.73	22.90	89.30	1991.29
R0.9	16.92	7.35	7.05	90.06	876.75	11.33	7.48	22.90	89.55	1940.68
R1.0	16.91	7.34	7.06	90.12	876.28	11.29	7.57	22.94	89.43	1960.48
R1.1	16.88	7.32	7.10	90.31	877.27	11.26	7.65	22.88	89.35	1970.73
R1.2	16.84	7.30	7.12	90.36	875.26	11.21	7.71	22.83	89.28	1973.02

**Fig. 9.** EDX mapping results of sample R0.9 after hydrothermal treatment.**Fig. 10.** EDX spectrum of sample R0.9 after hydrothermal treatment.

Xonotlite balance), silicate polymerization degree, and microstructural development highlights the nature of the products. In particular, a C/S ratio of 0.9 is considered the best ratio under experimental conditions. This ratio promotes the formation of Tobermorite and achieves superior mechanical properties. These findings provide insight into the structure-property relationship in hydrothermally synthesized CSH and serve as a basis for optimizing material design using recycled silica sources.

4. Conclusions

Calcium silicate hydrate was successfully synthesized from waste glass via hydrothermal synthesis, and the effect of the CaO/SiO_2 ratio on phase, microstructure, and mechanical properties was investigated. The results demonstrate that the CaO/SiO_2 ratio is a key parameter governing phase formation and material properties. At a CaO/SiO_2 ratio of 0.9, the material exhibited the highest mechanical strength, corresponding to the predominant formation of layered Tobermorite and Xonotlite. Increasing the CaO/SiO_2 ratio promoted the transformation of Tobermorite into fibrous Xonotlite due to excess Ca^{2+} , thereby lowering mechanical strength. XRD and FTIR analyses confirmed varia-

tions in crystalline phases and silicate bonding structures, while SEM and EDX observations revealed corresponding changes in morphology and elemental distribution. These findings establish a relationship among silicate polymerization (Q^n units), phase assemblage, microstructural development, and macroscopic properties, providing insight into the mechanism of calcium silicate hydrate formation from recycled silica sources. The synthesized materials show potential for applications in lightweight construction materials, thermal insulation, and environmental remediation. Moreover, utilizing waste glass offers a sustainable route for producing value-added functional materials while reducing environmental burdens. Since this study focused on the influence of composition under fixed hydrothermal conditions, future work will investigate the effects of synthesis parameters, including temperature, reaction time, and waste glass composition. Additional studies on advanced mechanical characterization and environmental applications, such as heavy-metal adsorption, will further optimize material performance and broaden potential applications.

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