



Novel cementitious encapsulation method for salt hydrates in building applications

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ABSTRACT

Phase change materials (PCMs) offer significant potential for improving building energy efficiency by moderating indoor temperature fluctuations and increasing the thermal inertia of building components. This study investigates a cementitious encapsulation approach for salt-hydrate PCMs, in which expanded vermiculite (EV) is impregnated with sodium sulfate decahydrate (Glauber's salt, GS) and subsequently coated using calcium sulfoaluminate (CSA) or magnesium oxy-sulfate (MOS) cement. The compatibility of GS with the selected binders was evaluated through isothermal calorimetry, compressive strength testing, X-ray diffraction (XRD), and thermophysical characterization. The results show that GS significantly alters CSA hydration, resulting in increased porosity and reduced mechanical performance. In contrast, MOS exhibited substantially greater compatibility with GS, maintaining its hydration behaviour and mechanical properties throughout the investigated curing period. MOS-coated EV-GS pellets were successfully produced and incorporated into mortar composites. The resulting composites exhibited reductions in thermal conductivity and thermal diffusivity of up to 70% relative to the reference MOS matrix while maintaining a comparable volumetric heat capacity. Overall, the results demonstrate the feasibility of producing MOS-coated PCM-containing pellets and highlight their potential for enhancing the thermal buffering characteristics of cementitious building materials.

1. Introduction

Maintaining stable indoor temperatures is essential for comfort and productivity, yet it remains one of the most energy-intensive aspects of building operation [1–5]. As climate change drives increasingly extreme temperature fluctuations, the energy required for daily thermal regulation continues to rise [6]. In this context, passive thermal management using phase change materials (PCMs) has emerged as a promising strategy to moderate indoor temperature swings and reduce reliance on mechanical heating and cooling systems [7]. By absorbing and releasing latent heat within a narrow temperature range, PCMs help stabilize indoor environments and lower overall energy demand [8–14]. When integrated into building components, they also offer an environmentally sustainable alternative to conventional construction practices, contributing to reduced carbon emissions [15]. Despite these advantages, the use of salt-hydrate PCMs presents several integration challenges [16].

A variety of PCM incorporation methods have been explored. Sleeve-based systems place PCM-filled plastic containers within wall cavities [17,18], while external thermal storage units coupled with HVAC

systems transfer heat to and from an outdoor reservoir. For new construction, encapsulating PCMs within porous hosts—such as foams or clays—allows them to function as structural aggregates [19]. In cementitious applications, PCMs can replace either coarse (>4 mm) or fine (<4 mm) aggregates. The present study focuses on developing functional fine aggregates (<4 mm) coated with a cementitious layer, enabling seamless incorporation into mortar while providing a thermally responsive alternative to conventional fine aggregates [20–23]. Because salt hydrates can alter the chemistry and mechanical properties of cementitious binders [24], identifying compatible coating materials is essential. Fig. 1 illustrates the conceptual structure of the proposed pellets.

However, existing encapsulation approaches introduce their own limitations. Polymeric coatings and glass bead methods raise environmental concerns, require energy-intensive processing, and often suffer from microleakage or fatigue under repeated thermal cycling [25–34]. Cement-based encapsulation offers a more sustainable alternative, yet two major failure mechanisms must be addressed. First, the volumetric expansion of the PCM during melting and solidification can generate

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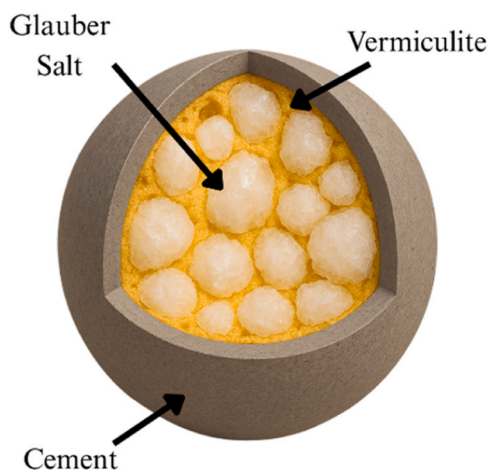


Fig. 1. Schematic representation of the EV-GS pellets. The yellow region denotes the vermiculite matrix, whose pores are filled with Glauber's salt crystals and enclosed by a gray cementitious coating.

internal stresses that exceed the tensile capacity of the cement shell, causing cracking or debonding. Second, chemical reactions between salt hydrates and cement phases—particularly in systems containing Glauber's salt ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$)—can form expansive minerals such as ettringite or gypsum, compromising mechanical integrity and PCM reversibility [35].

These challenges highlight the need for encapsulation strategies that minimize chemical incompatibility, accommodate PCM expansion, and remain cost-effective and environmentally sustainable [36,37]. A suitable matrix must be chemically inert, compatible with both salt hydrates and cement, capable of limited expansion, widely available, and economically viable. Expanded vermiculite (EV) satisfies many of these requirements. Its layered, highly porous structure enables strong retention of polar liquids, making it an effective host for PCMs [38]. EV is already widely used in construction for insulation, fire protection, and acoustic control [20,21,39–44]. Its low thermal conductivity ($0.06 \text{ W/m}\cdot\text{K}$), high specific heat capacity ($1.08 \text{ kJ}/(\text{kg}\cdot\text{K})$), and exceptional liquid absorption capacity allow it to store melted PCM within its pores, reducing volumetric expansion and limiting leakage through microcracks. Nonetheless, EV alone cannot prevent chemical interaction with cement, necessitating a protective coating.

The PCM selected for this study is Glauber's salt (GS), which offers a high phase-change enthalpy (250 kJ/kg) and a melting point of 32°C —ideal for research-scale thermal energy storage [45,46]. Despite its favorable thermal characteristics, GS reacts adversely with Ordinary Portland Cement (OPC) and CEM III binders, as confirmed in previous studies [24]. Therefore, a chemically compatible coating is required.

Two sulfate-resistant binders—calcium sulfoaluminate (CSA) [47, 48] and magnesium oxy-sulfate (MOS)—were selected due to their inherent tolerance to sulfate environments [49–57]. Unlike OPC, which can form gypsum and experience setting retardation upon sulfate exposure [35], CSA and MOS exhibit lower alkalinity and reduced susceptibility to deleterious reactions with GS [49,52,53]. CSA provides rapid setting and high early strength, while MOS offers strong wear resistance, low pH, and excellent durability in sulfate-rich conditions [58,59]. Their comparatively low pH further decreases the likelihood of harmful reactions with salt hydrates [60].

While porous-host PCM systems and coated aggregates have been widely investigated, the majority of reported encapsulation strategies rely on polymeric, silica-based, or glass shells [61–63]. These approaches can involve energy-intensive manufacturing routes, generate additional non-mineral waste streams, and may suffer from mechanical damage during mixing and service life. Direct cementitious encapsulation of salt-hydrate PCMs has received comparatively limited attention

because conventional binders are highly susceptible to chemical degradation in sulfate-rich environments [64].

The novelty of the present study lies in evaluating whether alternative cementitious binders can act as chemically compatible encapsulation systems for Glauber's salt. Rather than treating the coating solely as a physical barrier, this work investigates how binder hydration chemistry governs compatibility with sulfate-bearing PCMs. By comparing CSA and MOS binders, the study demonstrates that sulfate resistance alone is insufficient to predict PCM compatibility and identifies MOS as a promising mineral-based encapsulation material. Beyond reducing reliance on polymeric coatings, such systems may also offer advantages for future circular construction strategies through the development of fully mineral-based PCM composites.

This study investigates a cement-based encapsulation approach for salt-hydrate PCMs using EV as the absorbent core and CSA or MOS as protective coatings. The goal is to produce fine aggregate pellets ($<4 \text{ mm}$) that are compatible with cementitious mortars, limit direct contact between the PCM and surrounding cementitious materials, and minimize chemical degradation. Chemical interactions between GS and the coating binders are evaluated through isothermal calorimetry, compressive strength testing, and X-ray diffraction (XRD). After coating and curing, EV-GS pellets are incorporated into MOS-based mortar, and their thermal performance is assessed to determine their suitability for passive thermal regulation in building materials [65–68].

The following section details the materials, fabrication procedures, and characterization methods used to develop and evaluate the EV-GS pellets coated with CSA and MOS. This includes material selection, mix design, pellet production, and the experimental techniques employed to assess chemical compatibility, mechanical behavior, and thermal performance, establishing the methodological framework for evaluating this encapsulation strategy in cementitious applications.

The primary objective of this study is not to quantify PCM energy-storage capacity, which is already well established for Glauber's salt, but rather to evaluate whether suitable cementitious encapsulation systems can be developed without detrimental chemical interactions.

2. Materials and methods

2.1. Materials

The pellet core was produced by impregnating expanded vermiculite (EV) [Primaterra, Grades 3 (0–5 mm) and 2 (0–3 mm)] with sodium sulfate decahydrate (Glauber's salt, GS) [AnalaR NORMAPUR 99.6%, VWR]. EV exhibits a bulk density of 0.9 g/mL , a thermal conductivity of $0.0635 \text{ W/m}\cdot\text{K}$, and a volumetric heat capacity of $0.972 \times 10^6 \text{ J}/(\text{m}^3\cdot\text{K})$ [69]. GS, used as the phase change material, has an enthalpy of fusion of 251.2 kJ/kg , a melting temperature of 32.4°C , and a volumetric heat capacity ranging from 3.06 to $4.91 \times 10^6 \text{ J}/(\text{m}^3\cdot\text{K})$ [70].

Two cementitious binders were selected as coating materials:

Calcium sulfoaluminate (CSA) cement [Alpenat UP, VICAT], with a thermal conductivity of $0.7 \text{ W/m}\cdot\text{K}$, a volumetric heat capacity of $1.55 \times 10^6 \text{ J}/(\text{m}^3\cdot\text{K})$, and a thermal diffusivity of $1.03 \text{ mm}^2/\text{s}$ [71].

Magnesium oxy-sulfate (MOS) cement, synthesized from magnesium oxide (MgO) [312/31212 Caustic Burnt, Magnesita Co.] and magnesium sulfate heptahydrate ($\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$) [99.5%, Acros Organics], following established hydration reactions [53,54,56]. According to Khalil et al. [72], MOS typically exhibits a thermal conductivity of $1.68 \text{ W/m}\cdot\text{K}$, a volumetric heat capacity of $0.898 \times 10^6 \text{ J}/(\text{m}^3\cdot\text{K})$, and a thermal diffusivity of $1.05 \text{ mm}^2/\text{s}$.

2.2. Material characterization

2.2.1. Particle size distribution (PSD)

The particle size distributions of MgO and CSA were measured using an Analysette 22 NeXT (Fritsch). Isopropanol (refractive index 1.39) served as the dispersion medium, and samples were added until a beam

obscuration of 10–18% was reached. Each material was analyzed in triplicate, and the results were averaged. The refractive index for both solids was set to 1.73.

The median particle sizes (D_{50}) were 23.54 μm for MgO and 9.99 μm for CSA (Fig. 2, Table 1).

2.2.2. X-ray diffraction (XRD)

XRD measurements were conducted using a Bruker D4 Phaser measure for from 10 to 90(2 θ) on sample powders sieved below 500 μm . For quantitative phase analysis, 10 wt% silicone was added as an internal standard. QXRD was performed using a Co-Anode ($K\alpha 1$: 1.7901 Å and $K\alpha 2$: 1.7929 Å) and a LynxEye detector. Samples were placed to rotate at 30 RPM, and measurement was 1 s per step. Qualitative analysis were conducted by X'pert HighScorePlus 2.2 (PANalytical), whilst quantitative x-ray diffraction analysis (QXDA) was performed using the software TOPAS Academic 5.1. Primary cif data was obtained from the Crystallography Open Database.

Quantitative phase analysis was performed using the Rietveld refinement method implemented in TOPAS Academic 5.1. The addition of 10 wt% silicone internal standard enabled estimation of the amorphous content through comparison between the measured and known standard concentrations. Refinements included scale factors, background fitting, peak-shape parameters, unit-cell parameters, and preferred-orientation corrections where necessary. Phase identification was initially conducted using HighScore Plus and subsequently refined quantitatively using TOPAS.

As shown in Fig. 3, EV consisted of 59.9% vermiculite [$((\text{Mg}, \text{Fe}^{2+}, \text{Fe}^{3+})_3(\text{Al}, \text{Si})_4\text{O}_{10}(\text{OH})_2 \cdot x\text{H}_2\text{O})$] and 40.1% phlogopite [$\text{KMg}_3(\text{AlSi}_3\text{O}_{10})(\text{F}, \text{OH})_2$], consistent with typical mineral assemblages [73,74]. MgO displayed high purity with minor carbonate impurities, while CSA contained the expected mixture of calcium carbonates, sulfates, aluminates, and silicates [35].

XRD was also performed on the CSA and MOS samples after mechanical testing at different curing ages. To stop hydration, crushed fragments were immersed in isopropanol or ethanol for 24 h, dried at 40 °C for two hours, and then finely ground with 10 wt% silicone internal standard prior to analysis.

2.2.3. X-ray fluorescence (XRF) and loss on ignition (LOI)

EV was heated at 1000 °C for four hours in a porcelain crucible to determine loss on ignition (LOI). Subsequently, 1 g of the residue was fused with 10 g of lithium tetraborate and 750 μL of lithium iodide using a PANalytical CLAISSE LeNEO at 1065 °C to form a fused bead. Table 2 summarizes the oxide compositions of EV, CSA, and MgO.

Table 1

Particle size distribution parameters (D_{10} , D_{50} , D_{90}) of MgO and CSA powders.

Material	D_{10} (μm)	D_{50} (μm)	D_{90} (μm)
MgO	2.76	23.54	55.43
CSA	2.77	9.99	32.45

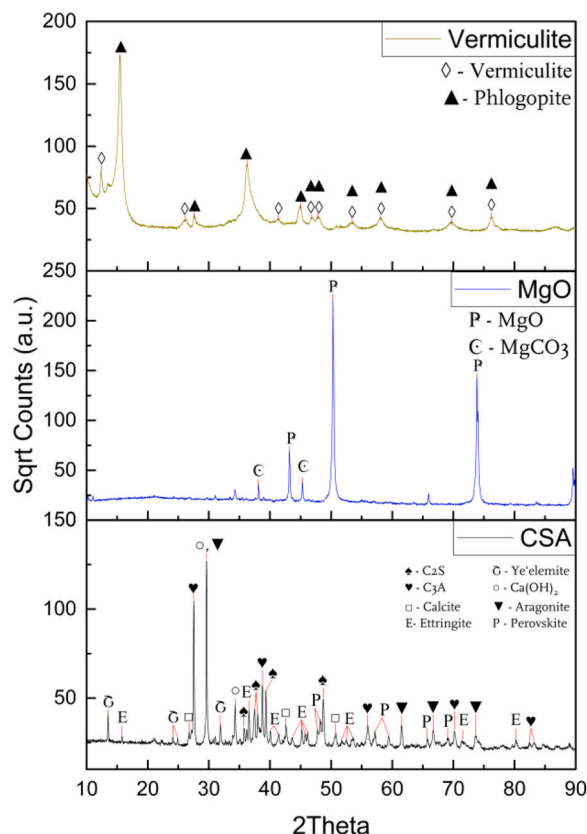


Fig. 3. XRD patterns of expanded vermiculite (top), MgO (middle), and CSA clinker powder (bottom).

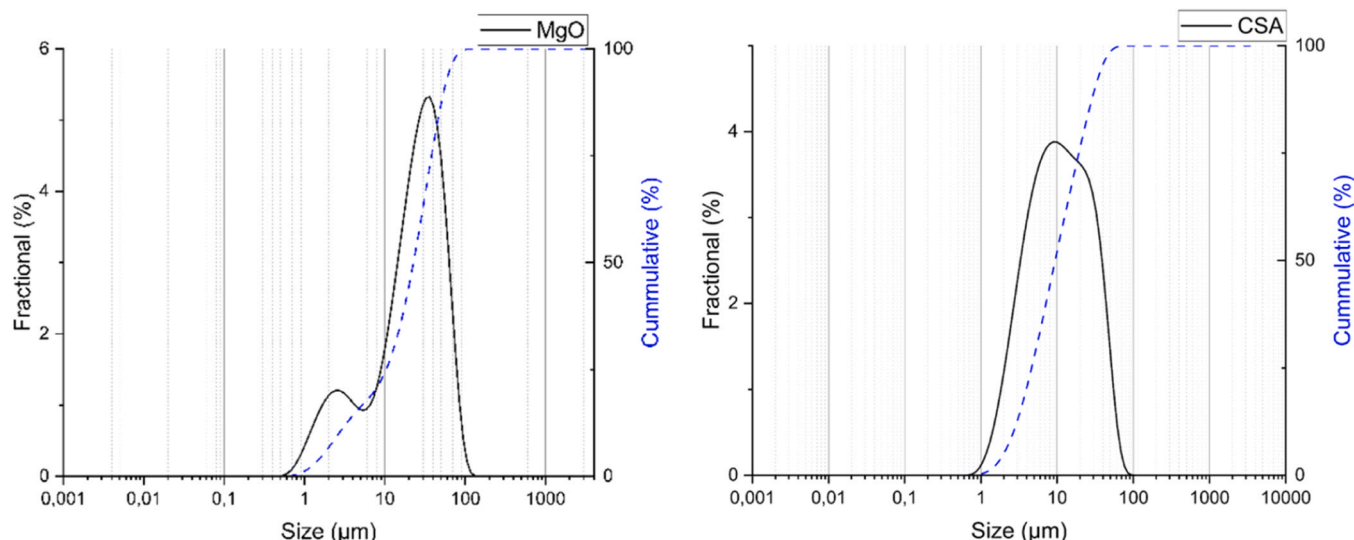


Fig. 2. Particle size distribution (PSD) of MgO and CSA powders.

Table 2

XRF quantified values of MgO, CSA, and EV.

Material	MgO	Al ₂ O ₃	SiO ₂	SO ₃	CaO	K ₂ O	TiO ₂	Fe ₂ O ₃	Other	LOI
Vermiculite	21.40	8.87	39.05	–	–	7.75	1.97	13.98	0.29	7.17
CSA	0.86	15.18	7.79	13.29	48.86	0.28	1.05	9.67	0.32	2.78
MgO	91.52	–	1.16	–	2.90	–	–	0.92	0.04	3.51

2.2.4. Gas pycnometry

Material densities were measured using a Micromeritics AccuPyc II 1340 gas pycnometer, which determines solid volume via gas displacement. Samples filled approximately 80% of either a 5 mL or 30 mL chamber. Ten measurements were collected per sample and averaged.

2.2.5. Manual pycnometry

Manual pycnometry was performed following ASTM C128-01 [75] to determine water absorption, particularly for EV. Glass pycnometers of 500 mL were used, and calculations followed the standard procedure.

2.3. Material preparation

2.3.1. CSA paste preparation

CSA pastes were prepared using a water-to-binder ratio (w/b) of 0.4, selected to balance mechanical performance with minimal free water. Higher w/b ratios can cause bleeding, which risks dissolving the PCM and forming deliquescent surface salts during curing. For reaction testing, GS was added as a percentage of binder mass and pre-dissolved in the mixing water. The sample compositions are listed in Table 3.

2.3.2. MOS paste preparation

MOS binders were produced using MgO, MgSO₄, and water in molar ratios of 10:1:20 and 15:1:20, which favor formation of the 5:1:20 phase known for its superior strength and water resistance [76,77]. Table 3 summarizes all mix designs.

2.3.3. Mortar mix composition

Mortar formulations followed BS EN 998-1/NHBC 2024 (6.1.14) guidelines, using 40 vol% MOS cement and 60 vol% fine aggregates [78]. CSA was initially considered but later excluded due to its inability to form a stable coating in preliminary trials. Mortar specimen mix presented on Table 4.

2.4. Pellet preparation

2.4.1. Impregnation

The EV-GS core was produced using a 1:5 mass ratio of EV to GS. First, 50 g of EV (dried at 130 °C overnight) was combined with 250 g of GS and heated to 45 °C in a water bath. Once melted, 17 g of deionized water was added to compensate for evaporation losses, and the mixture was mechanically stirred until homogeneous. After cooling to room

Table 3

Mix design samples and composition overview.

	CSA w/b ratio	MgO g	MgSO ₄ g	Water g	GS wt%	GS g
CSA 0 GS	0.4	-	-	-	-	-
CSA 5 GS	0.4	-	-	-	5	-
CSA 10 GS	0.4	-	-	-	10	-
CSA 15 GS	0.4	-	-	-	15	-
CSA 20 GS	0.4	-	-	-	20	-
MOS 10	-	403	120	360.3	-	-
MOS 15	-	604.5	120	360.3	-	-
MOS 15 5 GS	-	604.5	120	360.3	5	36.2
MOS 15 10 GS	-	604.5	120	360.3	10	72.5
MOS 15 15 GS	-	604.5	120	360.3	15	108.7
MOS 15 20 GS	-	604.5	120	360.3	20	145

Table 4

Composition of thermophysical test specimens.

Sample ID	MOS Binder (vol%)	MOS Coated EV Pellets (vol %)	EV-GS Pellets (vol%)	MOS-EV- GS Pellets (vol%)	Specimen Mass (g)
MOS	100	-	-	-	116.14
MOS (MOS- EV)	40	60	-	-	112.51
MOS (EV- GS)	40	-	60	-	103.15
MOS (MOS- EV-GS)	40	-	-	60	112.89

temperature, the material was sieved to remove agglomerates and left to crystallize for 24 h.

The EV:GS ratio of 1:5 was selected based on the high liquid absorption capacity of expanded vermiculite, which can absorb approximately four to five times its own mass in liquid. This ratio was therefore considered sufficient to maximize PCM loading while minimizing the presence of excess free liquid. The impregnation temperature of 45 °C was chosen to ensure complete melting of Glauber's salt, whose melting temperature is approximately 32 °C, while limiting excessive dehydration during processing. The addition of 17 g of deionized water was introduced to compensate for evaporation and dehydration losses occurring during the melting stage.

2.4.2. Pelletization

Pelletization was performed using an EIRICH TR04 mixer. A 1:13 molar MgSO₄:H₂O wetting solution was prepared one day in advance. The impregnated EV was added to the rotating drum, followed by alternating additions of MgO powder and sprays of the MgSO₄:H₂O wetting solution to achieve a double coating while maintaining the 15:1:20 MOS stoichiometry. The final MOS-to-core ratio was 1:6 (25 g MgO per 300 g impregnated EV), with a w/b ratio between 0.4 and 0.6. Pellets were cured for one month in a sealed, humid environment.

The MOS-to-core ratio of 1:6 was selected based on preliminary pelletization trials. Lower coating contents resulted in incomplete surface coverage of the EV-GS particles, whereas higher coating contents reduced the achievable PCM fraction in the final pellets. Consequently, the selected ratio was considered an appropriate balance between coating continuity and PCM loading for this proof-of-concept investigation.

2.4.3. Cross-sectional analysis

Cross-sectional analysis was performed by collecting 2–5 g of pellets, and using the Struers CitoVac. Each mold was prepared in a 7.5 mL cup, with 7 parts resin, and 1 part hardener. Once inside, allow 10/15 min of vacuuming, and 12 h for fully curing. Once samples are ready, they follow the step by step polishing of Struers Tegramin-30 sample polisher. Samples were polished using rough, coarse, and fine polishing disks. Once polished the samples were analysed using Zeiss Image.Z2m Microscope Using the Epiplan -NEOFLUAR 5X/0,13HD using Real Light-Dark Field settings, and image capture was done using the EDF Motorized Focus settings.

2.5. Thermal testing

Thermal properties were measured using an ISOMET 2104 (Applied Precision) via transient heat flow analysis. Samples were cast in 3D-printed PLA molds (height 2 cm, radius 3 cm) containing 60 vol% pellets and 40 vol% MOS binder (15:1:20). The instrument provided thermal conductivity (λ), volumetric heat capacity (C_p), and thermal diffusivity (α), following established procedures for PCM–cement composites [68]. It should be noted that the ISOMET method measures effective thermophysical properties and does not directly determine latent heat storage capacity, phase transition temperatures, or PCM reversibility.

Prior to testing, the specimens were cured for 7 days within the PLA molds under laboratory conditions (18 ± 2 °C). Thermophysical properties were measured using an ISOMET 2104 (Applied Precision) equipped with a surface probe operating at a maximum probe temperature of 50 °C. Each measurement required approximately 15–20 min to complete. The reported thermal conductivity, volumetric heat capacity, and thermal diffusivity values therefore represent the effective thermophysical properties of the composites under ambient laboratory conditions.

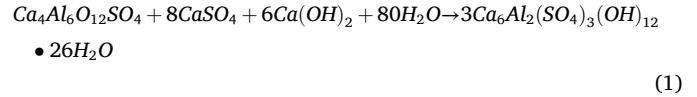
According to the manufacturer, the instrument provides a resolution of 0.001 W/(m·K) for thermal conductivity, 1×10^{-3} J/(m³·K) for volumetric heat capacity, and 0.01 °C for temperature measurements. The measurement reproducibility is $\pm 3\%$ of the measured value, while the stated accuracy is $\pm 5\%$ for thermal conductivity, $\pm 15\%$ for volumetric heat capacity, and ± 0.5 °C for temperature. All reported values represent the mean $\pm$ standard deviation of three independent measurements. The reported uncertainties in Table 7 correspond to the standard deviation of three independent measurements and should be interpreted separately from the instrument accuracy specified above.

3. Results and discussion

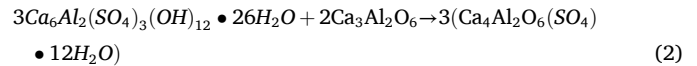
Understanding the chemical compatibility between a salt-hydrate PCM and cementitious binders is essential when developing encapsulation systems for building applications. The experiments involving the direct mixing of GS with CSA and MOS pastes were therefore designed to evaluate binder compatibility, the likelihood of salt leakage, and any chemical or physical degradation that could compromise structural performance over time. By replicating conditions of direct contact, the study assesses whether GS induces surface deterioration, strength loss, or volumetric instability within the cementitious coating.

3.1. Isothermal-calorimetry

Isothermal calorimetry was used to evaluate how GS influences the hydration behavior of CSA and MOS (Fig. 4). For CSA, the heat-flow profile over 24 h displayed two dominant peaks. Although four peaks are commonly reported in literature, overlapping reactions often reduce the number of distinguishable events. The first peak (2–6 h) corresponds to ye'elimite dissolution and ettringite formation (Eq. 1),



while the second peak (6–10 h) marks gypsum depletion and the conversion of ettringite to monosulfate (Eq. 2).



The introduction of GS increases the sulfate concentration in the pore solution, thereby prolonging the availability of SO_4^{2-} ions during hydration. As a result, Eq. 1 is promoted through enhanced ettringite formation at early ages. Simultaneously, the conversion represented by Eq. 2 becomes less pronounced because sulfate depletion is delayed. This interpretation is consistent with the observed increase in the first calorimetric peak and suppression of the second peak with increasing GS content.

Increasing GS content intensified the first peak and suppressed the second, indicating that additional sulfate from GS alters the balance between ettringite formation and subsequent conversion. As noted by Winter [79], high sulfate availability delays sulfate depletion and can shift the system toward aluminate-limited behavior. Taylor [35] further reports that C_3S reacts with even low GS concentrations (≈ 0.15 M), and excess GS can also react with $\beta\text{-C}_2\text{S}$, forming gypsum and producing a decalcified C–S–H gel with reduced Ca/Si ratio—ultimately weakening the material.

Furthermore, Wang [51] reported that ettringite formation is governed by the balance of AlO_2^- , OH^- , Ca^{2+} , and SO_4^{2-} ions. According to Wang, the addition of up to 3 wt% GS promotes ettringite formation, whereas higher GS contents favor the formation of monosulfate. Wang further proposed that sodium sulfate reacts with calcium hydroxide to generate sodium hydroxide, which subsequently reacts with monosulfate to form hydrogarnet (C_3AH_6), sodium sulfate, calcium hydroxide, and water, as shown in Eq. 3.

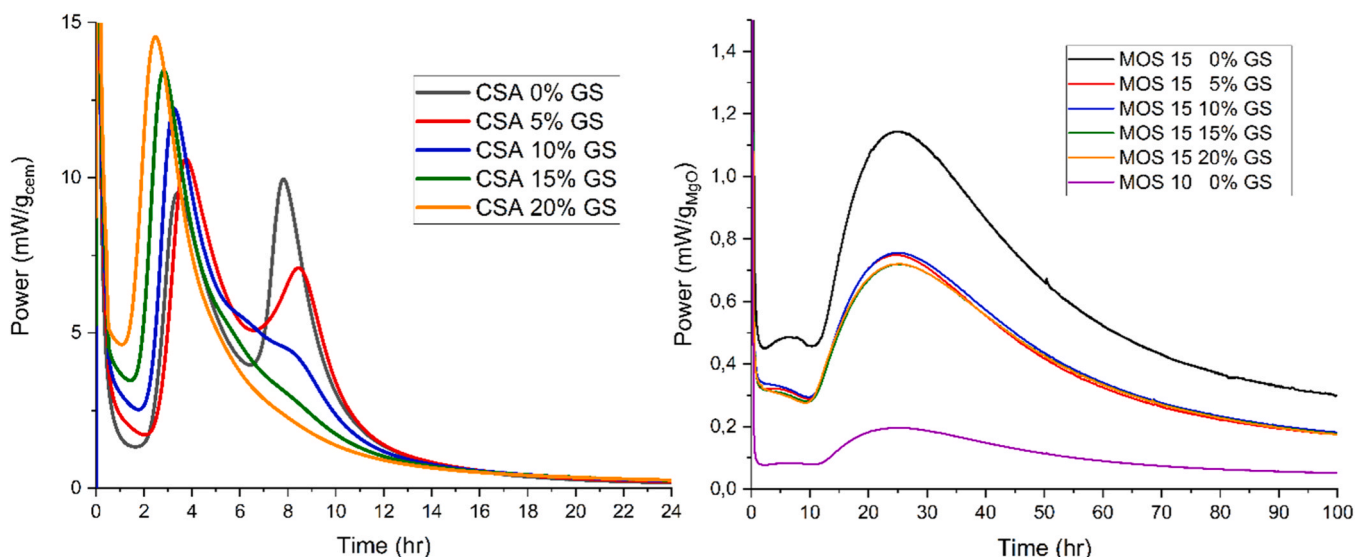
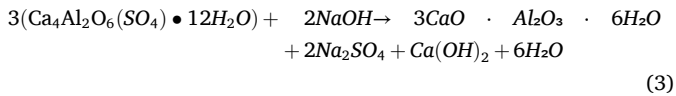
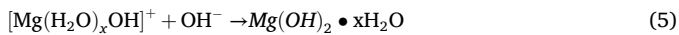
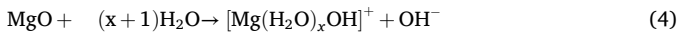


Fig. 4. Iso-calorimetry of CSA (left) and MOS (right) with variations of additional GS percent.

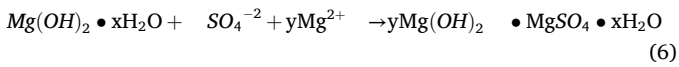


The GS addition also introduces sodium ions into the system. While sulfate primarily influences AFt/AFm (Aluminate-Ferrite-trisubstituted/monosubstituted) equilibria, sodium contributes to changes in pore-solution chemistry and alkalinity. These changes may destabilize monosulfate phases and promote the formation of secondary products such as hydrogarnet, contributing to the observed increase in porosity and reduction in strength.

For MOS, the calorimetry curves extended over 100 h and exhibited two broad peaks. No significant differences were observed between the 10:1:20 and 15:1:20 mixes. The first peak (0–10 h) corresponds to MgO hydration (4 and 5).



In contrast to CSA, the sulfate ion introduced through GS is chemically similar to the sulfate already present within the MOS binder system. Consequently, no major disruption of 4 and 5 is expected. The calorimetry results support this interpretation, as GS did not significantly alter the onset of hydration or the overall shape of the hydration profile. This is followed by a larger peak (11–45 h) associated with the formation of MOS phases (Eq. 6) [53].



The principal influence of GS appears to be the formation of secondary sodium-magnesium sulfate hydrates, including bloedite and minor konyaite, rather than any substantial modification of the primary MOS hydration pathway.

When the heat release was normalized to the MgO content, all GS-containing samples exhibited a lower total heat output, indicating that the MgO-to-sulfate ratio, rather than the absolute GS content, governs the heat evolution during hydration. Importantly, GS did not delay the onset of hydration, and the relative difference between MOS 10 and MOS 15 remained consistent regardless of GS addition.

Zhu [76] reported that the hydration behavior of MOS cement exposed to salts is primarily influenced by the cation with respect to setting time, whereas the anion largely governs the resulting strength and overall performance. Because the sulfate ion introduced by GS is identical to the sulfate already present in the MOS system, no significant

changes in the hydration reaction are expected. Regarding the cation, Ba [77] investigated the addition of sodium silicate and attributed the observed improvement in the formation of the desirable 5–1–7 phase to the hydrophobic behavior of the silicate ion rather than to the sodium cation itself. Consistent with these findings, the present results suggest that the sodium ion has a negligible influence on the hydration behavior and overall performance of the MOS system under the conditions investigated.

These reactions result in the phase compositions discussed in the following section.

3.2. Compressive strength and mineralogy

Compressive strength tests were performed on CSA and MOS pastes with and without 15% GS to assess whether GS exposure compromises mechanical performance (Fig. 5, Table 5). The 15% GS level represents the expected interfacial concentration between the coating and the EV-GS core. CSA was tested at 7 days due to its rapid setting, while MOS samples were evaluated at 28 days following ASTM C109/C349 guidelines [80,81].

The addition of 15 wt% GS significantly affected early-age CSA strength development. Averaged across all curing ages investigated (1, 5, and 7 days), the CSA-GS specimens exhibited an approximately 54% lower compressive strength than the corresponding reference specimens. However, the magnitude of the reduction varied with curing age, decreasing from 33% at Day 1 and 62% at Day 5 to approximately 12% at Day 7. Visual inspection revealed significantly higher porosity in CSA-15, likely caused by GS crystallization during early hydration followed by dissolution as curing progressed. This mechanism aligns with

Table 5

Compression test results of CSA and MOS.

Compression (MPa)					
Day	1	5	7	14	28
CSA	26.83 ± 3.83	35.13 ± 2.21	32.04 ± 0.76	-	-
CSA + 15% GS	17.87 ± 0.59	13.38 ± 0.80	28.07 ± 2.03	-	-
MOS 10	0.55 ± 0.04	-	6.30 ± 0.70	8.08 ± 1.39	13.17 ± 1.31
MOS 15	4.59 ± 0.15	-	15.40 ± 0.30	20.74 ± 0.48	28.34 ± 2.04
MOS 15 + 15% GS	0.69 ± 0.02	-	8.43 ± 1.45	13.07 ± 1.95	25.01 ± 2.14

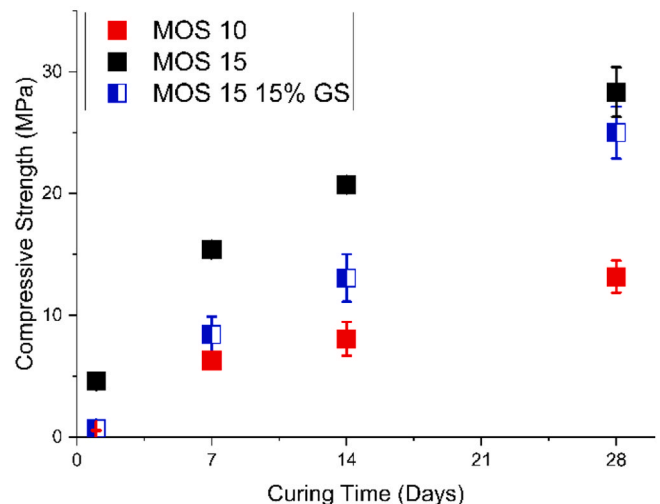
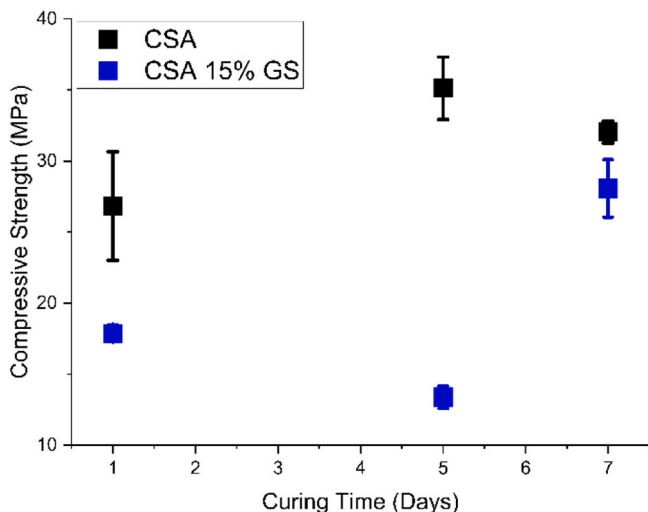


Fig. 5. Compression Testing of CSA (left) and MOS (right).

reports that high salt concentrations increase pore size and slow hydration kinetics in CSA systems [82,83].

Quantitative XRD (Table 6) confirmed substantial changes in phase composition. GS increased the amorphous fraction, likely due to dehydration of GS to thenardite [84]. Crystalline phases such as ettringite, calcite, dolomite, gibbsite, and anhydrite were reduced in the GS-containing samples. After aluminate depletion, gypsum and anhydrite became the dominant sulfate phases, while sodium likely entered the C–S–H gel [79]. The destabilization of AFm/Aft phases under high sodium concentrations explains the reduced strength and increased porosity.

The observed unusual porous structure is attributed to a complex mechanism initiated by the GS addition. The workability of the material was no different from the regular CSA, and there were no crystals in the liquid added to the CSA. While the GS was pre-dissolved in the mixing water, the fast-setting nature of the CSA paste (evident from calorimetry) likely forced the premature crystallization of the GS. Subsequently, during the curing period, the continued hydration of the cement paste pulls the water necessary for its reaction, causing the GS crystals to dehydrate and partially dissolve. This dehydration and dissolution process leaves behind the observed pores within the cement paste as well as thenardite. This increased porosity, combined with the phase compositional changes, plays a significant role in the material's overall mechanical strength (Fig. 6).

In contrast, MOS samples showed more robust performance. MOS 15 achieved the highest strength, while MOS 15 with 15% GS showed a moderate reduction but remained comparable to MOS 10. Literature reports strengths of 25–60 MPa for highly reactive MgO systems [85], and the observed values fall within this range. All MOS samples appeared visually similar (Fig. 7), with no GS-induced defects.

XRD analysis of the MOS material was performed at curing ages of 1, 7, 14, and 28 days, corresponding to the compression testing schedule. Prior to analysis, the samples were chemically dried via solvent exchange to arrest hydration and preserve the phases present at each age.

At Day 1, no crystalline MOS phases had formed. Only the initial constituents were detected: MgO (periclase) [P], magnesium sulfate hydrates ($\text{MgSO}_4 \cdot 7\text{H}_2\text{O}/6\text{H}_2\text{O}$) [S], and minor $\text{Mg}(\text{OH})_2$ (brucite) [B]. With increasing curing time, the XRD patterns showed a progressive decrease in MgO (periclase) accompanied by a corresponding increase in $\text{Mg}(\text{OH})_2$ (brucite), indicating the slow advancement of the hydration reaction. Gypsum [G] was also identified among the reaction products.

Only minor differences were observed between the MOS 10 and MOS 15 samples (Fig. 8). The primary MOS peaks were identified by region and labelled [M]. Because available databases did not contain sufficient reference patterns for precise phase assignment, literature sources were used to validate the general MOS peak regions [53,86].

The broad peaks near $2\theta = 20^\circ$ align with brucite, although this

region is also characteristic of early MOS phases such as 5-1-3 and 3-1-8. Analysis of samples containing GS (MOS 15 + 15% GS) showed minimal deviation from this overall pattern (Fig. 9). The continued conversion of MgO to $\text{Mg}(\text{OH})_2$ and the consistent appearance of MOS phases indicate that GS has little influence on the primary MOS crystallization process.

The only notable differences were the presence of bloedite ($\text{Na}_2\text{Mg}(\text{SO}_4) \cdot 4\text{H}_2\text{O}$), labelled [D], and small amounts (<5%) of konyaite ($\text{Na}_2\text{Mg}(\text{SO}_4) \cdot 5\text{H}_2\text{O}$) detected at $2\theta = 26.03^\circ$ (Fig. 9). In agreement with the calorimetry results, the hydration reaction proceeds similarly regardless of GS content, confirming that compressive strength is governed primarily by water content (w/b ratio) rather than salt addition.

Taken together, the calorimetry, compressive-strength, and XRD results indicate that the compatibility of salt-hydrate PCMs with cementitious coatings is governed primarily by hydration chemistry rather than sulfate resistance alone. Although CSA is generally considered sulfate resistant, additional sulfate and sodium introduced through Glauber's salt substantially altered hydration pathways and increased porosity. In contrast, the MOS system remained comparatively unaffected because sulfate is already an intrinsic component of its hydration chemistry. This finding provides an important design principle for future mineral-based PCM encapsulation systems and extends beyond the specific materials investigated in this study.

3.3. Pelletized material

Pellets were produced using various EV size fractions, with 1.4–2.0 mm EV yielding the most consistent final pellet size (<4 mm). CSA-coated pellets were unsuitable due to poor coating stability during fabrication and curing. Manual PSD measurements confirmed that most MOS-coated pellets fell within the 2.0–4.0 mm range (Fig. 10).

3.3.1. Pellet morphology and cross-sectional analysis

To evaluate the quality of the encapsulation process, representative MOS-coated EV-GS pellets were sectioned, mounted in epoxy resin, polished, and examined using optical microscopy. Fig. 11 presents a representative cross-section of the pellet structure.

The images reveal a distinct core-shell morphology consisting of an inner EV-GS region surrounded by a continuous MOS coating. The shell remained generally uniform around the pellet perimeter, indicating that the stepwise pelletization process successfully produced a cementitious encapsulation layer. Due to the sample-preparation procedure, which involved polishing with water- and ethanol-based media, a portion of the exposed Glauber's salt was dissolved from the sectioned surface. Consequently, the observed core morphology may not fully represent the original distribution of GS within the pellet. No large-scale coating discontinuities were observed within the analysed sections.

Table 6
Q-XRD of CSA and CSA GS 15% paste in 7-day ages (% by mass).

Phase Name	Composition	CSA –1	CSA –5	CSA –7	CSA GS-1	CSA GS-5	CSA GS-7
Anhydrite	CaSO_4	8.06 ± 0.18	8.02 ± 0.18	7.45 ± 0.17	5.14 ± 0.12	3.72 ± 0.12	4.56 ± 0.12
Bayerite	$\text{Al}(\text{OH})_3$	1.17 ± 0.33	1.44 ± 0.25	1.27 ± 0.26	1.17 ± 0.24	0.99 ± 0.24	0.85 ± 0.25
Calcite	CaCO_3	3.58 ± 0.17	4.17 ± 0.22	3.84 ± 0.16	2.68 ± 0.13	2.63 ± 0.18	2.83 ± 0.13
Dolomite	$\text{CaMg}(\text{CO}_3)_2$	2.80 ± 0.37	1.35 ± 0.24	1.45 ± 0.21	0.62 ± 0.15	0.58 ± 0.16	0.58 ± 0.16
Ettringite	$\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12} \cdot 26 \text{H}_2\text{O}$	18.21 ± 0.43	22.33 ± 0.50	19.23 ± 0.45	11.71 ± 0.28	14.60 ± 0.37	14.52 ± 0.38
Gibbsite	$\text{Al}(\text{OH})_3$	1.33 ± 0.19	3.63 ± 0.38	2.63 ± 0.34	0.85 ± 0.27	1.02 ± 0.27	1.13 ± 0.30
Kuzelite	$\text{Ca}_4\text{Al}_2(\text{OH})_{12}\text{SO}_4 \cdot 6 \text{H}_2\text{O}$	2.07 ± 0.16	0.87 ± 0.14	0.55 ± 0.12	-	-	-
Larnite	Ca_2SiO_4	18.37 ± 0.42	16.46 ± 0.41	15.06 ± 0.35	16.38 ± 0.33	15.28 ± 0.34	16.13 ± 0.36
Magnetite	Fe_3O_4	-	1.65 ± 0.14	1.67 ± 0.12	2.45 ± 0.15	1.51 ± 0.10	1.59 ± 0.11
Magnesite	MgCO_3	0.67 ± 0.12	0.47 ± 0.34	0.32 ± 0.25	-	0.18 ± 0.12	0.13 ± 0.17
Perovskite	CaTiO_3	7.78 ± 0.16	8.49 ± 0.17	7.49 ± 0.15	7.64 ± 0.13	6.78 ± 0.13	6.99 ± 0.13
Quartz	SiO_2	0.54 ± 0.10	4.20 ± 0.12	8.08 ± 0.14	0.50 ± 0.14	4.95 ± 0.10	8.34 ± 0.13
Thenardite	Na_2SO_4	-	-	-	5.01 ± 0.23	6.81 ± 0.24	5.09 ± 0.24
Yeelimite	$\text{Ca}_4(\text{Al}_6)\text{SO}_4$	1.59 ± 0.14	3.79 ± 0.17	3.92 ± 0.16	3.48 ± 0.13	1.99 ± 0.13	2.57 ± 0.13
Amorphous		33.84 ± 1.10	23.14 ± 1.25	27.06 ± 1.10	42.38 ± 0.88	38.87 ± 0.96	34.69 ± 1.00

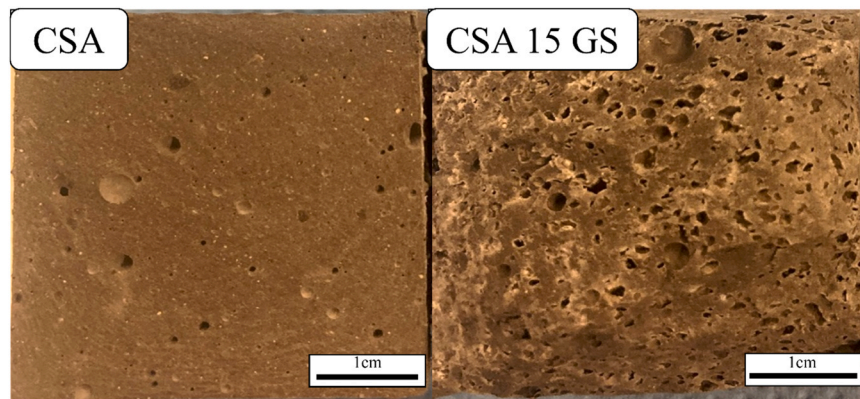


Fig. 6. Visible comparison of effects of GS on CSA cement at 7 days. Formation of pores on right image as well as a noticeable discoloration.

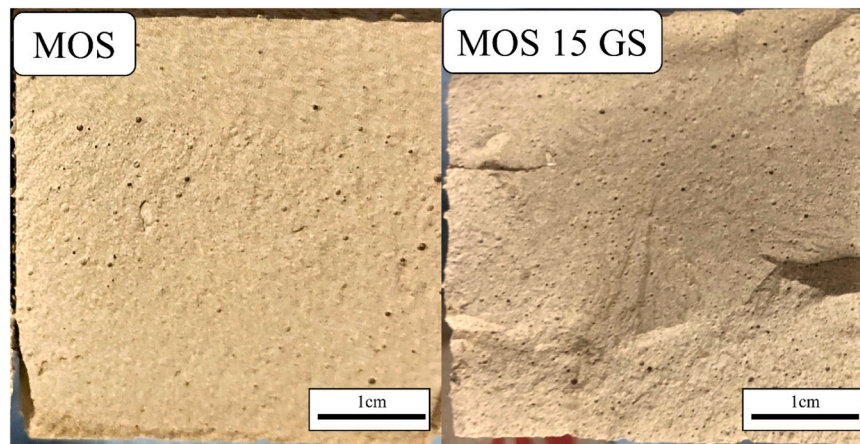


Fig. 7. MOS Prism Cross-section at 28 days.

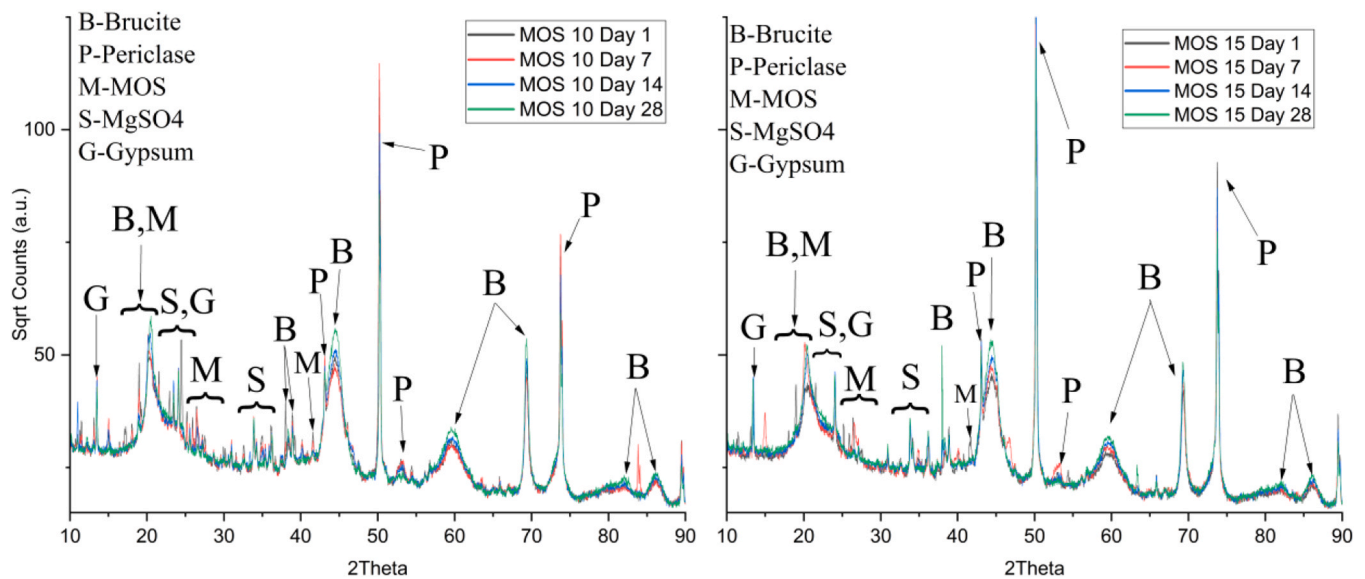


Fig. 8. XRD of MOS phases in both MOS and MOS 15 cement mixes.

Local variations in shell thickness were observed, which are attributed to the irregular geometry of the expanded vermiculite particles and the dynamic nature of the pelletization process. Small voids were occasionally present within the coating layer, which is typical for light-weight pelletized systems. Nevertheless, the overall continuity of the

MOS shell supports the proposed encapsulation mechanism by physically separating the PCM-containing core from the surrounding cementitious matrix.

The coating thickness measured from the pellet surface to the core-shell interface ranged from approximately 0.3–0.45 mm. Considering

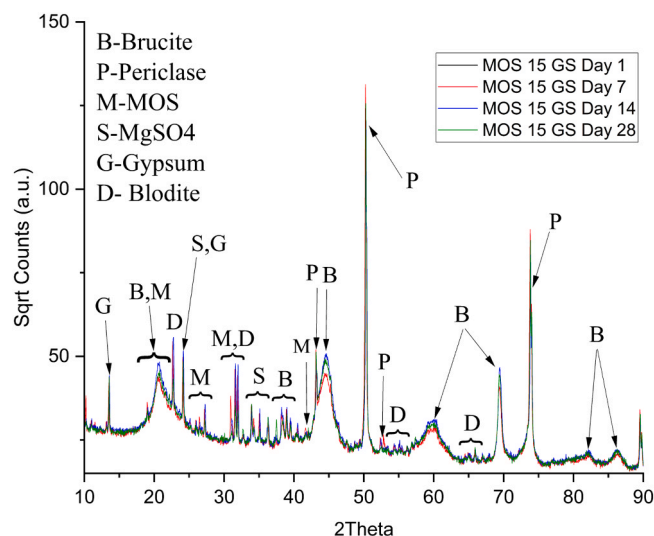


Fig. 9. XRD phases on MOS 15 with 15% GS.

both sides of the pellet, the total shell thickness was typically between 0.9 and 1.0 mm.

3.3.2. Density

Gas and manual pycnometry results (Table 7) showed that EV possesses high porosity (43–53%) and can absorb up to four times its weight in water. The 1.4–2.0 mm EV fraction was used for EV-GS impregnation. MOS-coated EV-GS pellets (2.0–4.0 mm) exhibited an average density of 1.18 g/mL, consistent with their composite structure.

3.3.3. Thermophysical properties of pelletized material

Thermal behavior was evaluated using four sample types: pure 15:1:20 MOS, MOS with empty EV pellets, MOS with uncoated EV-GS pellets, and MOS with coated EV-GS pellets. The MOS-EV-GS composite specimen contained approximately 26.4 wt% Glauber's salt relative to the total specimen mass. After one week, samples containing pellets exhibited slight expansion and superficial cracking, consistent with previous observations [87], but intact regions allowed reliable measurement. The render with such defects are shown in Fig. 12.

Thermal properties are summarized in Table 8. Pure MOS showed high conductivity and diffusivity, characteristic of dense cementitious

materials. Adding empty EV pellets reduced conductivity and diffusivity due to vermiculite's insulating properties. Incorporating uncoated EV-GS pellets further reduced conductivity and diffusivity while increasing heat capacity, reflecting the thermal buffering effect of the PCM.

The reported thermal properties were measured after 7 days of curing under laboratory conditions ($18 \pm 2^\circ\text{C}$). Consequently, moisture content, porosity, interfacial thermal resistance, and superficial cracking may all contribute to the effective composite properties measured by the ISOMET device. The reported values should therefore be interpreted as bulk thermophysical properties of the composite system rather than the isolated effect of PCM incorporation.

The reduction in thermal diffusivity cannot be attributed exclusively to the presence of the PCM. The incorporation of expanded vermiculite introduces a low-conductivity porous phase, while interfacial thermal resistance between the pellets and binder, local porosity, and the observed cracking may also contribute to the measured reductions. Consequently, the measured values should be interpreted as effective composite properties resulting from all constituent phases and microstructural effects. Furthermore, the ISOMET measurements quantify sensible thermophysical properties under transient heat-flow conditions and should not be interpreted as a direct measurement of latent heat storage capacity.

The coated EV-GS pellets produced the most desirable thermal profile: the lowest conductivity and diffusivity, combined with elevated heat capacity. Despite minor cracking, these results demonstrate that encapsulated PCM pellets significantly enhance thermal regulation. The measured MOS heat capacity ($1.477 \times 10^6 \text{ J/m}^3 \cdot \text{K}$) exceeded typical literature values of $0.898 \times 10^6 \text{ J/m}^3 \cdot \text{K}$ [72]. The MOS-EV-GS composite achieved the lowest diffusivity of all mixtures—an essential characteristic for passive thermal regulation [91–93]. As illustrated in Fig. 13, the coated-pellet composite aligns well with the expected behavior of an effective thermally buffering material.

The observed reductions in thermal conductivity and thermal diffusivity indicate an increased resistance to heat transfer through the composite. However, these measurements should not be interpreted as direct evidence of latent heat storage. The ISOMET technique measures effective thermophysical properties under transient heat flow conditions and cannot quantify phase transition enthalpy or supercooling behaviour. Consequently, the present results demonstrate improved thermal buffering characteristics rather than direct confirmation of retained PCM storage capacity following encapsulation.

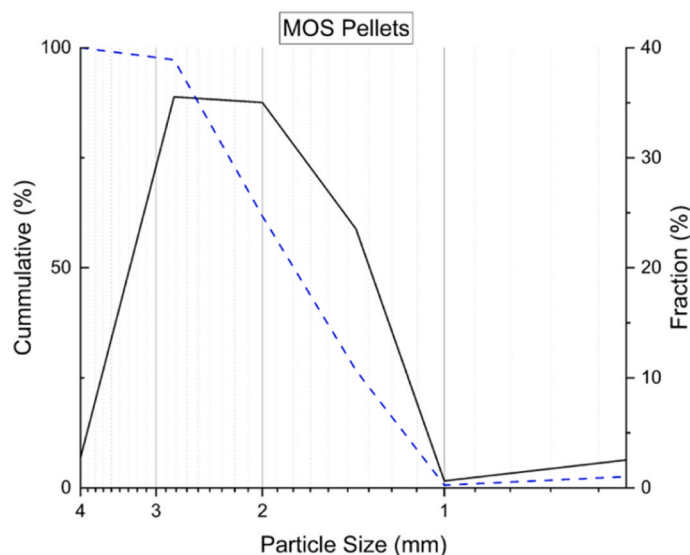


Fig. 10. PSD (left) and visual result (right) of MOS pellets.



Fig. 11. Cross-Section of polished pellet.

Table 7

Pycnometry results overview of materials and pellets.

Pycnometry	Size Fraction (mm)	Bulk Density	Skeletal Density	Porosity (%)	Absorption (%)
EV	1.0–1.4	0.52 ± 0.01	1.62 ± 0.01	53.60	440.10
	1.4–2.0		1.78 ± 0.01	43.07	402.04
EV-GS	1.0–2.8	1.39 ± 0.003	1.88 ± 0.005	-	-
CSA-EV-GS	1.0–1.4	1.63 ± 0.002	1.65 ± 0.1	-	-
	1.4–2.0	1.47 ± 0.003		-	-
	2.0–2.8	1.45 ± 0.003		-	-
MOS-EV-GS	2.0–4.0	1.18 ± 0.04	1.96 ± 0.001	-	-

4. Conclusion

This study developed a proof-of-concept for cement-coated phase change material (PCM) pellets designed for incorporation into mortar and render systems. The work focused on two sulfate-resistant binders—Calcium Sulfoaluminate (CSA) and Magnesium Oxy-Sulfate

(MOS)—selected for their potential compatibility with Glauber's salt (GS), a widely studied salt-hydrate PCM. Expanded vermiculite (EV) served as the absorbent matrix for stabilizing the PCM, after which the EV-GS cores were coated using MOS-based pelletization.

A key finding of this study is that sulfate resistance alone is insufficient to predict compatibility between salt-hydrate PCMs and cementitious encapsulation systems; instead, compatibility is governed

Table 8

Thermal properties of base materials and mortar MOS samples.

Material Sample	Thermal Conductivity (W/m•K)	Heat Capacity (10^6 J/m ³ •K)	Thermal Diffusivity (10^{-6} m ² /s)	Apparent Density (g/mL)	Ref.
GS	1.00 ± 0.02	1.62 ± 0.14	-	-	[88]
EV	0.06	1.08	-	-	[89]
CSA	0.70	0.97	-	-	[90]
MOS	2.193 ± 0.055	1.477 ± 0.012	1.487 ± 0.035	2.05	-
MOS (MOS-EV)	1.180 ± 0.026	1.407 ± 0.012	0.838 ± 0.014	1.99	-
MOS (EV-GS)	1.123 ± 0.032	1.433 ± 0.006	0.826 ± 0.025	1.82	-
MOS (MOS-EV-GS)	0.379 ± 0.007	1.460 ± 0.000	0.259 ± 0.005	1.99	-

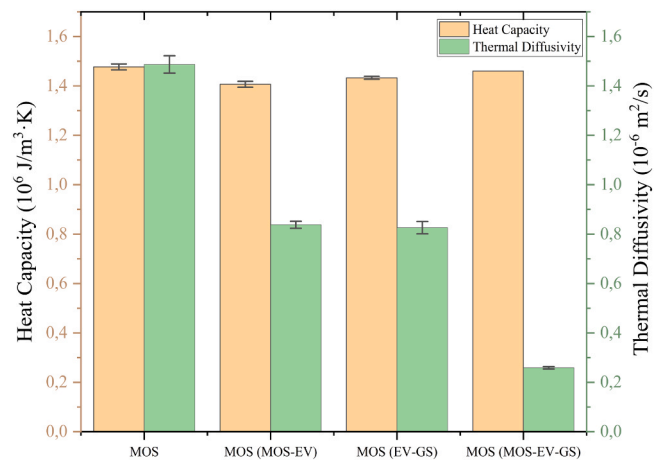


Fig. 13. Graphical representation of thermal data of MOS concrete render mixes.



Fig. 12. Observation of top (left) and bottom (right) view of puck render.

primarily by the interaction between PCM chemistry and binder hydration pathways.

Experimental findings demonstrated that CSA is unsuitable as a coating material in the presence of GS. Exposure to GS caused rapid dehydration and crystallization, producing a porous, thenardite-rich microstructure that compromised coating integrity almost immediately. In contrast, MOS—particularly the 15:1:20 formulation—showed far greater compatibility. Although GS reduced the total heat release in isothermal calorimetry and led to the formation of bloedite and konyaite, these effects did not significantly influence strength over the investigated curing period. After 28 days, MOS 15 with GS performed comparably to the GS-free reference. Both MOS formulations tested (10:1:20 and 15:1:20) maintained their mechanical performance in the presence of GS over the investigated curing period.

When incorporated into a render, the coated pellets reduced thermal conductivity and thermal diffusivity while maintaining a comparable volumetric heat capacity to the reference material. These changes indicate enhanced thermal buffering behaviour and suggest potential for thermal-energy-storage applications; however, direct quantification of latent heat storage requires future DSC investigation.

Despite containing only 26.4 wt% GS, the MOS-EV-GS composite exhibited a volumetric heat capacity of $1.46 \times 10^6 \text{ J/m}^3 \cdot \text{K}$, which is comparable to the literature value reported for pure GS ($1.62 \times 10^6 \text{ J/m}^3 \cdot \text{K}$). This indicates that the incorporation of PCM-containing pellets did not substantially reduce the overall heat-storage capacity of the composite while simultaneously lowering its thermal diffusivity. These improvements indicate that the pellets effectively enhance thermal buffering and slow heat transfer—key attributes for passive thermal regulation.

The broader implications of this work are significant for sustainable construction. Enhancing the thermal performance of building materials can reduce operational energy demand, lower carbon emissions, and improve indoor comfort in climates with substantial heating or cooling requirements. Although CSA is typically two to three times more expensive than OPC, the thin (~1 mm) coating layer required for PCM encapsulation keeps overall material costs manageable. MOS offers additional advantages—high strength, chemical resistance, and cost-effectiveness—though its sensitivity to prolonged moisture exposure limits its use to interior applications.

Overall, this research establishes a viable pathway for using specialty cements to encapsulate salt-hydrate PCMs, providing a foundation for thermally responsive and energy-efficient building materials. Future work should focus on quantifying latent heat storage capacity through DSC analysis, optimizing PCM-to-cement ratios, refining coating thickness, and evaluating leakage resistance and durability under repeated melting-crystallization cycles. Such investigations are necessary before long-term field application can be confirmed.

CRedit authorship contribution statement

C.A. Wesemann: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **H.J.H. Brouwers:** Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work, the authors used ChatGPT 5.5 to improve in clarity and readability of the material. After using this tool/service, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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