ABSTRACT: Phase change materials (PCMs) store latent heat energy as they melt and release it upon freezing. However, they suffer from chemical instability and poor thermal conductivity, which can be improved by encapsulation. Here, we encapsulated a salt hydrate PCM (Mg(NO\textsubscript{3})\textsubscript{2}·6H\textsubscript{2}O) within all-silica nanocapsules using a Pickering emulsion template. Electron microscopy analysis demonstrated robust silica–silica (RSS) shell formed inner silica layer of approximately 45 nm thickness, with silica Pickering emulsifiers anchored to the surface. The RSS nanostructured capsules are 300–1000 nm in size and have far superior thermal and chemical stability compared with that of the bulk salt hydrate. Differential scanning calorimetry showed encapsulated PCMs were stable over 500+ melt/freeze cycles (equivalent to 500+ day/night temperature difference) with a latent heat of 112.8 J·g\textsuperscript{-1}. Thermogravimetric analysis displayed their impressive thermal stability, with as little as 37.2% mass loss at 800 °C. Raman spectroscopy proved the presence of salt hydrate within RSS capsules and illustrated the improved chemical stability compared to non-encapsulated Mg(NO\textsubscript{3})\textsubscript{2}·6H\textsubscript{2}O. Energy capsule behavior compared with the bulk material was also observed at the macroscale with thermal imaging, showing that the melting/freezing behavior of the PCM is confined to the nanocapsule core. The thermal conductivity of the silica shell measured by laser flash thermal conductivity method is 1.4 ± 0.2 W·(m·K)\textsuperscript{-1}, which is around 7 times more than the thermal conductivity of the polymer shell (0.2 W·(m·K)\textsuperscript{-1}). RSS capsules containing PCMs have improved thermal stability and conductivity compared to polymer-based capsules and have good potential for thermoregulation or energy storage applications.

KEYWORDS: heat storage, salt hydrates, capsule, Pickering emulsion, silica shell, thermal energy
as an incentive to drive technological advancement in smart air conditioning. Developing zero-energy thermoregulation systems that automatically respond to environmental changes would simultaneously decrease energy requirements and improve user comfort.

Salt hydrates (also known as hydrated salts or crystallohydrates) are the most promising low-temperature PCM due to their low cost and high energy density. They have the formula MS·nH₂O, where MS is a metal salt and n is the hydration number. The level of hydration has a profound effect on the Tₓ. The major problem with salt hydrates is chemical instability, mainly due to two factors: low thermal conductivity in the liquid phase, leading to slow and unpredictable freezing behavior, and changes in hydration number, causing phase separation and an irreversible Tₓ change. For use in practical applications, they can be encapsulated into a core–shell structure. The encapsulation minimizes solid-to-liquid volume change, stabilizes the desired hydration number, protects from the external environment, and enhances heat transfer due to an increased surface area (SA) to volume ratio (SA/vol). To maximize beneficial PCM properties, nanostructured capsules (diameter <1 μm) are desirable due to their large SA/vol compared with macro- (1+ mm) or microcapsules (1–1000 μm) and increased structural stability. Capsules can be adapted to contain any active ingredient, and their miniaturization will help to achieve mass-market penetration. Few researchers have reported the fabrication of salt-hydrate-loaded capsules, with long-term stability especially challenging to achieve. However, in previous reports, we successfully synthesized salt-hydrate-loaded nanocapsules, confirming the predicted benefits of encapsulation with several interesting observations: (i) encapsulated salt hydrates were thermally and chemically stable over 100+ cycles with minimal latent heat storage loss; (ii) sonochemistry is highly beneficial for emulsion production; (iii) addition of extra water to the salt hydrate core conserves the salt hydration level; (iv) latent heat of the core material was higher than expected, possibly due to nanoconfinement effects which have been reported in the literature. Research into nanoconfinement effects on PCMs is in its infancy, recently summarized in a review by Aftab et al.

The fabrication of core–shell nanomaterials requires a template such as an emulsion—a dispersion of liquid droplets within another immiscible liquid. Particle-stabilized emulsions are known as Pickering emulsions. Solid particles have advantages over traditional emulsifiers (surfactants), including increased interfacial binding energy, which boosts template stability, and the ability to use different kinds of particles to stabilize single capsules, increasing functionality. Pickering emulsions can encapsulate either hydrophobic or hydrophilic materials, using oil-in-water (O/W) or “inverse” water-in-oil (W/O) emulsions, respectively. Pickering emulsions must be partially wetted by both phases to adsorb to the interface. As the emulsifier surface must be primarily hydrophobic (i.e., water contact angle >90°) to stabilize W/O emulsions, oxides such as silica prove a good choice due to their abundant surface OH groups allowing facile modification. For this study, we used commercial silica particles (LUDOX TMA) hydrophobized with octadecyltrimethoxysilane (OTMS) to stabilize W/O Pickering emulsions. Silica particles are the most commonly used Pickering stabilizer and are more thermally conductive than conventional organic surfactants, increasing heat transfer to the thermally responsive PCM core.

Fabricating nanomaterials using Pickering templates is difficult due to smaller particles having lower binding energy at the solid–liquid interface. The packing of the solid stabilizers, therefore, limits the minimum size of capsules. However, droplet sizes can be tailored by adjusting energy input. Essentially, the higher the energy delivered, the smaller the droplet size.

In this work, we demonstrate that PCM-loaded capsules with an all-silica shell can be produced via inverse Pickering emulsions. Ultrasonic energy was applied to form the silica-nanoparticle-stabilized Pickering emulsion template. A further silica layer was then deposited by the HCl-catalyzed condensation of tetraethyl orthosilicate (TEOS) to create a robust silica shell (RSS) with Mg(NO₃)₂·6H₂O loaded in the core. Thus, the Pickering emulsion was turned into the emulsion stabilized by the Pickering shell. The fabricated RSS capsules were 300–1000 nm in diameter. This size range appears optimum for thermal energy storage, as capsules of diameter <300 nm may see a decrease in latent heat due to low core-to-shell ratio. Silica is a good shell material due to its chemical inertness, abundance, low cost, and excellent thermal stability. We demonstrated solely inorganic PCM nanocapsules produced form the Pickering emulsion. Nanoconfinement of the PCM within RSS capsules results in them having excellent thermal and chemical stability for automatic local heat control/storage. The smaller volume of the capsules means the increase of the surface area, which enhances heat transfer and reduces supercooling of salt hydrates.

RESULTS AND DISCUSSION

Formation and Morphology. RSS capsules were formed through the interfacial self-condensation of TEOS using HCl as a catalyst, shown in Figure 1. The samples consisted of an oil phase (7.5 g overall) made up of 5 wt % hydrophobically modified LUDOX TMA (OTMS-LUDOX) in cyclohexane and an aqueous phase (1 g overall) containing Mg(NO₃)₂·6H₂O (50 wt %). The two phases were sheared using ultrasonication to form a Pickering emulsion. Ultrasonic treatment provides higher input of energy into the reaction
system compared to high-speed mixing resulting in formation of nanometer-sized capsules and partial melting of the nanoparticles forming a Pickering emulsion shell. To this were quickly added varying amounts of TEOS (1−3 mL) along with HCl (2 mL) and stirred overnight to give RSS nanocapsules. The samples are described in Table 1 as RSS$_n$, where $n$ is the amount of TEOS added in milliliters. Further details can be found in the Experimental Section.

As seen in the scanning electron microscopy/transmission electron microscopy (SEM/TEM) images in Figure 2, the RSS capsules are 300−1000 nm in diameter, with most in the 500−750 nm size range. Sonication parameters delivering high-energy input were important in fabricating such small materials. The volume of TEOS used had minimal effect on capsule diameter. Assuming a completely smooth surface, a capsule with a diameter of 500 nm will have a 20-fold increase in SA/vol compared to a capsule with a diameter of 10 μm (SA/vol for sphere = 3/$r$). The rough surface of these submicrometer capsules further increases their SA/vol. It is also reasonable to suggest some nonhydrophobic LUDOX particles will be present in the core, which can potentially build a thermally conductive network within the capsules.55 The composition of the capsule shell made of silica reduces the supercooling effect, a notorious problem of salt hydrates, keeping the PCM core composition as compared to Mg(NO$_3$)$_2$·6H$_2$O without capsule shell.

**Figure 2.** (a−c) SEM and (d) TEM images of (a,b) RSS1.5 and (c,d) RSS3 samples. Please refer to size distribution of the capsules and shell for RSS3 sample in Supporting Information, Figures S3−S5.

**Table 1. List of RSS Samples**

| sample | oil phase | aqueous phase | TEOS added (mL) | HCl added (mL) |
|--------|-----------|--------------|----------------|---------------|
| RSS1   | cyclohexane (7.135 g, 9.16 mL), OTMS-LUDOX (0.375 g) | Mg(NO$_3$)$_2$·6H$_2$O (0.5 g), H$_2$O (0.5 g, 0.5 mL) | 1              | 1.5           |
| RSS1.5 |            |              | 2              | 2             |
| RSS2   |            |              | 3              | 3             |

**Figure 3.** (a) TGA curves for RSS nanocapsules. (b−e) TEM images of (b) RSS1, (c) RSS1.5, (d) RSS2, and (e) RSS3 before TGA measurements. Scale bars are 200 nm for all TEM images. Mass losses are 28% for RSS3, 30% for RSS1.5, 33% for RSS2 and RSS1 samples at 200 °C; 3% for RSS3, 4% for RSS1.5, 3% for RSS2, and 7% for RSS1 samples between 200 and 580 °C. The thickness of the capsule shells is between 20 and 45 nm, as shown in Figures S5−S8 in the Supporting Information.

From 50 to 200 °C, all RSS samples lose between 28 and 33% mass attributed to free water in the capsule cores. Above 200 °C, the remaining mass reaches a plateau with minimal further losses. By 800 °C, RSS samples have lost between 37.2 and 43.7% mass due to the water chemically bonded to the silica shell. By comparison, polymer shell capsules will lose 70−100% mass upon heating to 800 °C.45,57

Although all RSS capsules have similar thermal characteristics, the minor differences can be rationalized. From RSS1 to RSS3, the increasing dosage of TEOS stabilizes the Pickering emulsion better, resulting in less waste LUDOX in the final product due to collapsed capsules. RSS1 loses mass slowest until 130 °C. By 200 °C, it has the least mass (67%) remaining alongside RSS2. Above this temperature, it loses the most mass of all samples. The initially slow rate of mass loss can be ascribed to it containing the lowest amount of Mg(NO$_3$)$_2$·6H$_2$O and the highest amount of silica particles not involved in shell formation, which lose almost zero mass at low temperatures. Differential scanning calorimetry (DSC) results (Figure 4) confirmed the instability of the RSS3 sample after 500 heat uptake/release cycles. Reduced shell strength when in emulsification or detach from the interface. These free particles comprise silica and do not absorb latent heat in the studied temperature range.56

**Thermal Uptake/Release Characteristics.** Thermogravimetric analysis (TGA) results show the thermal stability of the RSS capsules, all of which have similar curves (Figure 3a).
using only 1 mL of TEOS leads to RSS1 having the lowest remaining mass at 800 °C (56.3%).

RSS2 loses mass fastest until 200 °C, where 68.4% remains. This is a result of 2 mL of TEOS giving a relatively durable shell, with an encapsulation efficiency better than that for RSS1. Better encapsulation results in initial rapid water loss. By 800 °C, RSS2 has a mass remainder of 59.1%—an intermediate value between that of RSS1 and RSS3. RSS1.5 behaves similarly to RSS2; however, it loses mass more slowly at all temperatures.

Above 130 °C, RSS3 loses mass slowest of all samples, with 72% and 63% mass remaining at 200 and 800 °C, respectively. This is due to the high amount of TEOS added, resulting in denser SiO2 shell material with high thermal stability. The durable shell also provides excellent protection for the core material, so water is lost at a more gradual rate. With such good shell stability at increased temperatures, the nanocapsules are also excellent candidates for high-temperature latent heat storage for concentrated solar power (CSP) applications. CSP plants require components stable at very high temperatures, as increased heat energy leads to improved efficiency.58 Dehydrated inorganic salts are the most promising high-temperature PCMs, so it should be possible to encapsulate them within RSS capsules. The RSS3 sample, with the densest silica shell formed, demonstrated the best stability during heat uptake and release.

Close-up TEM images in Figure 3b−e show the structure of the capsules fully, with silica formed from TEOS deposited at the inner interface. OTMS-LUDOX particles are layered on top of this inner shell. On the images, circular dark “halos” can be observed for some capsules, indicating the inner shell, approximately 20−30 nm thick regardless of the volume of added TEOS. The fact the capsules are loaded with solid material makes the shell thickness difficult to measure. Cao et al. also observed that increasing the amount of TEOS added had minimal effect on shell thickness, explaining that increasing TEOS volume causes a more compact and, therefore, more durable shell to form.59 This durability causes the improved thermal stability of RSS3 above the other samples, whereas shell thickness remains constant.

DSC results in Figure 4 demonstrate that the silica shell can increase cycling stability, which depends on the shell thickness. Bulk Mg(NO3)2·6H2O has lost its salt hydrate structure after 20 heat uptake/release cycles. Encapsulation into the silica shell stabilized the salt hydrate structure due to the prevention of the water loss. The RSS1 sample demonstrated considerable mass loss of water during the first cycles (40%), which can be related to the thin shell layer and defects in the shell. Increasing the amount of TEOS used for the shell formation increases the stability of the heat uptake/release with the latent heat capacity of 77.2 J g−1 for the RSS2 sample after 500 cycles (water loss is 22% during first cycles). The overcooling remained at a low value for all energy capsules, which can be explained by the high thermal conductivity of silica shell (1.4 ± 0.2 W·(m·K)−1) compared to the 0.2 W·(m·K)−1 for the polymer (like polyurethane) shell.

The best DSC stability has been achieved for the RSS3 sample with a dense silica shell (Figure 5). The sample demonstrated high stability for over 500 heat uptake/release cycles, keeping a stable enthalpy of 112.8 J g−1. The sample demonstrated an increased supercooling related to the denser capsule shell. First cycles demonstrated the loss of unbound water, 32% mass, which is higher than that for the RSS2 sample because of the thicker capsule walls resulting in more defected capsules after synthesis. However, thermal properties have been stabilized during further cycling. Figure S9 in the
Supporting Information demonstrates that the loss of water from defected capsules stops after the fifth cycle, and the next heat uptake/release cycles remain stable.

A comparison of the behavior of RSS3 capsules and the bulk salt hydrate is shown in Figure 6. Thermal images of Mg(NO$_3$)$_2$·6H$_2$O and RSS3 capsules are shown in Figure 6A,B. Both were subjected to direct heating with an IR lamp at a distance of 10 cm. Bulk Mg(NO$_3$)$_2$·6H$_2$O melted before it reached its $T_m$ at only 82.3 °C (Figure 6A). In contrast, RSS3 capsules remain as a white powder far beyond the $T_m$ at 113 °C (Figure 6B), with no volume change and no liquid observed. The apparent absence of melting proves that Mg(NO$_3$)$_2$·6H$_2$O phase transitions are confined to the core of the RSS3 capsules.

When heated in an oven at 120 °C for 6 h and cooled to room temperature (Figure 6C,D), Mg(NO$_3$)$_2$·6H$_2$O becomes a compact solid block, and much water is lost (30.6% mass), which is approximately 72.5% of the total water in the sample (H$_2$O makes up 42.2% of Mg(NO$_3$)$_2$·6H$_2$O). RSS3 capsules, in contrast, remain as a white powder with seemingly no volume change despite losing around 22.3% mass. Mass loss from RSS capsules is mainly free water from the core. SEM images of the capsules (Figure 6E,F) before and after heating to 120 °C show how the capsule morphology remains unchanged at the nanoscale and robust to changes in temperature. This structural maintenance explains why the RSS3 capsules in Figure 6C,D appear to have no change in volume despite the mass loss.

Chemical Analysis. To probe any thermally induced changes to the chemical structure, we employed Raman spectroscopy (Figure 7). We registered Raman spectra for both Mg(NO$_3$)$_2$·6H$_2$O and RSS3 before and after heating to 120 °C for 6 h under air. This procedure simulates the practical applications where PCM would be exposed to both the atmosphere and prolonged heating times. Mg(NO$_3$)$_2$·6H$_2$O (Figure 6A) displays two peaks for nitrate stretching at 729 cm$^{-1}$ and an intense peak at 1058 cm$^{-1}$.60 The peak at 729 cm$^{-1}$ shifts and splits after cycling. Asymmetric stretching modes are seen at 1358 cm$^{-1}$ as two peaks, both of which split after heating. It also has three peaks corresponding to H$_2$O stretching at 3250, 3452, and 3575 cm$^{-1}$. Similar peaks were described by Zhang et al.61 After 6 h of heating (Figure 6B), the H$_2$O peaks have lower intensity due to dehydration, and some peaks have shifted, now appearing at 3250, 3484, and 3571 cm$^{-1}$. An additional H$_2$O stretching peak is formed at 3402 cm$^{-1}$. The cause of the peak shifting/splitting is likely due to changes in the hydration number.62 Salt hydrates have a specific mechanism of melting—as the water is released from the crystal structure, it dissolves the salt. Changes in crystalline water structure affect melting and, therefore, thermal performance.63

In contrast, RSS3 capsules have minimal change in their Raman spectra after thermal cycling (Figure 7c,d). The nitrate stretching peak at 1049 cm$^{-1}$ clearly shows the presence of Mg(NO$_3$)$_2$·6H$_2$O, although the peak intensity is decreased due to the silica shell. This Raman shift is confirmed to be from the nitrate ion rather than any shell material, as hollow capsules containing no salt did not display this peak. Peaks at 1366 and
1452 cm\(^{-1}\) combine character from the salt hydrate nitrate anions and C–H bending from the OTMS-LUDOX. The triple peak at 2883 cm\(^{-1}\) is assigned to OTMS-LUDOX C–H stretching. H\(_2\)O stretching appears as a broad double peak at 3257 and 3421 cm\(^{-1}\). Unlike bulk Mg(NO\(_3\))\(_2\)·6H\(_2\)O, there is no peak shifting after heating, suggesting the hydration number is unaffected due to nanoconfinement of the hexahydrate structure. It also suggests water loss from the core is water not fully bonded to Mg(NO\(_3\))\(_2\), that is, from the 50 wt % water added to the aqueous phase during synthesis. These observations are consistent with DSC measurements, with improved chemical stability and maintenance of the Mg(NO\(_3\))\(_2\)·6H\(_2\)O hydration number, leading to excellent thermal cycling ability.

**CONCLUSIONS**

Thermal storage nanocapsules containing salt hydrate phase change materials were fabricated by the HCl-catalyzed interfacial condensation of tetraethyl orthosilicate. Pickering emulsions stabilized by silica nanoparticles and formed by ultrasonication were used as capsule templates. TEOS was condensed to form a thin layer (25–45 nm depending on the preparation conditions) of silica at the water–cyclohexane interface of the Pickering emulsion droplets with silica nanoparticles anchored to the silica shell, giving a robust silica–silica shell. The nanocapsules were 300–1000 nm in diameter, with the majority being 500–750 nm. SEM and TEM images revealed their morphology, with the large surface area to volume ratio of the nanocapsules providing excellent thermal transfer, further improved by the rough capsule surface and the presence of internal silica nanoparticles. The increase of added TEOS volume from 1 to 3 mL improved the thermal stability of the shell, despite having minimal effect on shell thickness.

RSS nanocapsules had excellent thermal properties, as demonstrated by TGA and DSC. The thermal cycling stability of the Mg(NO\(_3\))\(_2\)·6H\(_2\)O was profoundly increased upon encapsulation. The bulk salt hydrates are stable for fewer than 20 melt/freeze cycles. In contrast, salt hydrates within RSS capsules are stable over many cycles (>500 cycles), due to hermetic sealing within the silica shell resulting in stable 112.8 J·g\(^{-1}\) phase transition enthalpy after 100 to 500+ cycles. No structural changes that occurred in the bulk phase were detected in the nanoencapsulated Mg(NO\(_3\))\(_2\)·6H\(_2\)O by Raman spectroscopy.

Future work in the field of functional nanocontainers should focus on the development of both multifunctionality and scale-up of production. A microfluidic technology approach will enable the formation of monodisperse nanocapsules with multifunctional core and shell materials, as well as waste reduction through optimal synthesis conditions. The obtained core–shell particles could be arranged in various energy-saving materials by additive manufacturing technology (3D printing). The use of silica as the shell material should also lead to the fabrication of other more functional oxide shell materials such as TiO\(_2\) and ZrO\(_2\). Developing cutting-edge energy nanomaterials will help to lead a revolution in low-energy applications and to reduce the enormous carbon footprint of the thermal energy sector.

**EXPERIMENTAL SECTION**

**Materials.** LUDOX TMA (34 wt % aqueous dispersion), tetraethyl orthosilicate, hydrochloric acid (HCl, 37%), magnesium nitrate hexahydrate (Mg(NO\(_3\))\(_2\)·6H\(_2\)O, 99%), and sodium dodecyl sulfate (SDS) were purchased from Sigma-Aldrich UK. Cyclohexane (99%) and toluene (99%) were received from ReAgent and used as a solvent and for washing. Octadecyltrimethoxysilane (OTMS, 90% purity), aqueous ammonia (32%), and reagent-grade ethanol were purchased from Fisher Scientific UK. All materials were used as received with no further purification. Milli-Q water was used throughout.

**Synthesis of Hydrophobic SiO\(_2\).** LUDOX TMA colloidal silica was hydrophobically modified by the addition of alkylsilane groups to the silica surface, from the method of Schoth et al.\(^4\) LUDOX TMA 34 wt % solution in water (50 mL) was mixed thoroughly with reagent-grade ethanol (50 mL) and SDS (50 mg). The pH was set to 9.5 with NH\(_3\) and OTMS was added (0.02 mol, 8.49 mL). The mixture was stirred overnight to equilibrate before refluxing at 80 °C for 2 h. As the surface reaction of OTMS occurred, the mixture became a sludge due to the hydrophobic SiO\(_2\) particles precipitating out of the aqueous solution. The precipitation confirmed the reaction was complete. The product (OTMS-LUDOX) was washed with ethanol and was separated by centrifugation (16128 rcf, 5 min) before being dried at 120 °C.

**Synthesis of Silica–Silica Capsules.** A 5 wt % solution of OTMS-LUDOX in cyclohexane (7.5 g overall) was stirred overnight, followed by bath sonication to ensure the particles were fully dispersed. The aqueous phase (1 g overall) containing 50 wt % Mg(NO\(_3\))\(_2\)·6H\(_2\)O was added and shaken to create an initial macroemulsion. The mixture was then ultrasonicated using a QSonica Q700-220 (10 min, 10 on 5 off pulse regime, 1/2 in. tip) with ice cooling to create a Pickering emulsion. TEOS was added, immediately followed by HCl. The sample was allowed to stir overnight in a closed vial to complete the formation of the silica shell. Samples were washed with toluene (10 mL) and removed from solution by centrifugation (1792 rcf, 2 min). Products were left to dry in a fume hood at room temperature, yielding a white powder. The product could be redispersed by bath sonication in organic solvents.

**Characterization.** SEM images were taken using a JEOL JSM-7001F. Samples were prepared by taking 10 μL of the capsule solution straight after reaction and diluting it in 2 mL of toluene. The droplet (20 μL) of the diluted sample was added to a glass coverslip attached to an SEM stub with carbon tape. They were dried under ambient conditions. Samples were coated with chromium for 45 s before measurement.

TEM micrographs were registered with JEOL 2100+ LaB\(_6\) TEM operated at 200 kV. TEM samples were prepared by diluting 10 μL of capsule suspension straight after reaction in 4 mL of toluene. One drop of the diluted sample was added to a copper-coated TEM grid and allowed to dry under ambient conditions. Size distribution for SEM and TEM images was analyzed using ImageJ software (https://imagej.nih.gov/ij/download.html).

Thermogravimetric analysis was performed with a TA Instruments Q6000. Measurements were taken from room temperature to 800 °C with a ramp of 10 °C min\(^{-1}\) under a nitrogen atmosphere.

DSC measurements were taken using a DSC6, PerkinElmer, USA, to determine the latent heat storage properties and thermal cycling stability of the encapsulated PCM. DSC was measured between 40 and 120 °C for up to 100 cycles under a nitrogen atmosphere with a 5 °C/min ramp. DSC pans were not sealed to view the effect of the encapsulation on the stability on crystallohydrate content during thermal cycling.

Thermal conductivity of the capsule shell was performed by laser flash thermal conductivity method employing a Nd:Cr/GGG glass laser (BLS400, Basel Lasertech) operating at a wavelength \(λ = 1.064 \mu\text{m}\). The pulse energy was adjusted to keep the sample temperature increase below 5 °C. The transient temperature was measured with a HgCdTe infrared detector (HCT-80, Infrared Associated, Inc.). The sample was held in a horizontal position in the chamber.

Raman measurements were taken using a HORIBA XploRA Plus Raman microscope with a 532 nm wavelength laser. Dry powder samples were used for analysis.

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Thermal images were taken using a FLIR T660 high-performance infrared camera (https://www.flir.co.uk/products/t660/, accessed 04/06/2020) from FLIR Systems Inc. Mg(NO₃)₂·6H₂O and RSS capsule powders were placed on glass slides and heated in an air atmosphere for 6 h at 120 °C.

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ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsnano.0c03706.

Additional information for TGA, capsule size distribution graphs for all samples, size distribution of the shell thickness and DSC analysis of the first, second and fifth heat uptake/release cycles for the RSS3 sample (PDF)

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Author Contributions

M.G. and J.S. conceived and performed most of the experiments, interpreted the data, and wrote the manuscript. M.B. performed the electron microscopy measurements. E.S., A.N., and V.V. characterized heat storage properties and thermal conductivity of the capsule shell. A.N. revised the manuscript. D.S. supervised the completion of the project, corrected the final draft of the paper, and submitted it.

Notes

The authors declare no competing financial interest.
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