

# Supplementary information

## Development of Novel Microencapsulated Hybrid Latent/Chemical Heat Storage Material

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Thermal energy storage, Phase change material, Thermochemical material,  
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## 1. Visual observation of the effect of CaO impregnation on CaO/Ca(OH)<sub>2</sub> expansion

For examining the effect of supporting CaO on MEPCM, CaO "just" mixed MEPCM, in which MEPCM and CaO particles (99.9%, Kanto Chemical Co., INC.) were mixed at a weight ratio of 4:1, was prepared as a comparative sample. Each sample is named CaO20M/MEPCM (CaO20mass% mixed MEPCM) and CaO20S/MEPCM (CaO20mass% supported MEPCM). Then, both samples were heated under the same condition of former analysis by TG-DTA respectively, and the difference was investigated.

**Figure S1** shows SEM images, EDS mappings and photographs before and after TG measurement of each sample. From the results of SEM and EDS, MEPCM and CaO were completely independent in the CaO20M/MEPCM, while CaO were coated on the surface of MEPCM in the CaO20S/MEPCM. Also, CaO coating did not peel off before and after TG. From the results, it was found that the impregnation method played an important role in bonding the shell of MEPCM and CaO.

From the photographs, it was shown visually that CaO on the MEPCM surface could suppress the volume expansion after TG measurement more than normal CaO. In this experiment, CaO reacts with water vapor and changes to Ca(OH)<sub>2</sub> after TG. Since the respective densities are 3.34 g cm<sup>-3</sup> for CaO and 2.21 g cm<sup>-3</sup> for Ca(OH)<sub>2</sub>, it is obvious that volume expansion occurs after TG. However, the amount of volume expansion is completely different between CaO20S/MEPCM and CaO20M/MEPCM. The reason of this result is considered that the crystal structure between CaO and  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> has a great influence. XRD results show that calcium aluminate exists at the interface between CaO and alumina. As described in discussion part, from the results of past studies<sup>43-45</sup>, it is mentioned that the volume expansion caused by the reaction of CaO/Ca(OH)<sub>2</sub> is suppressed by forming a

network of this calcium aluminate as an impurity layer and covering the CaO particles.

Therefore, it is considered that CaO supporting on the MEPCM by impregnation method has the advantage of suppressing the volume expansion, and it leads to preventing from thermal conductivity decreasing.

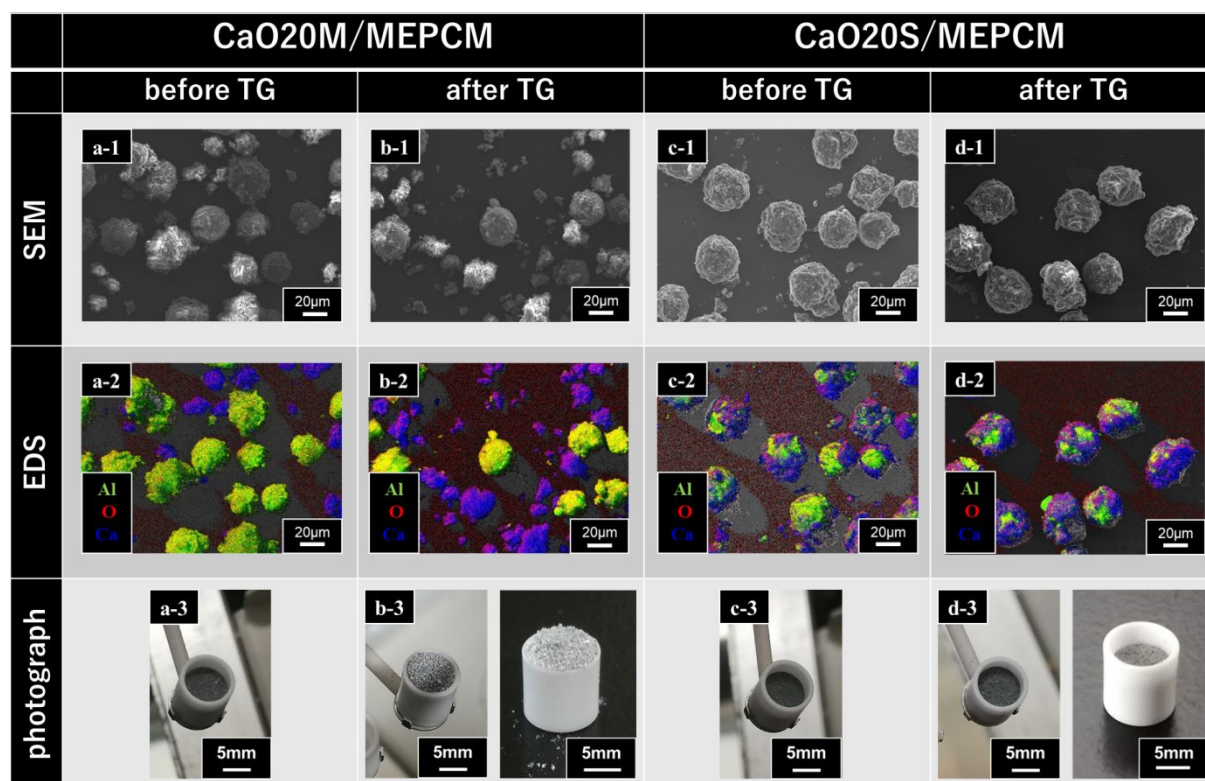


Figure S1. SEM, EDS and photograph of CaO20M/MEPCM and CaO20S/MEPCM before and after TG-DTA measurement.

## 2. Heat storage capacity after 25 cycles test

In the main part, the result of five cycle durability test shows stable heat storage and release behavior of CaO30/MEPCM. However, when actually using CaO30/MEPCM as a heat storage material, further stability should be shown. Therefore, 25 cycles durability test was additionally carried out.

However, the thermal analysis equipment (TG-DTA) used in the text was found to be defective, so it was not possible to conduct repeated tests under the same experimental conditions. Therefore, 25 cycles durability test was conducted using an alternative apparatus. Then, each sample after 1, 5, 25 cycles was checked by SEM and EDS mapping.

**Figure S2** shows the illustration of apparatus used for the 25 cycles test of CaO30/MEPCM. CaO30/MEPCM was inserted into a quartz tube, and the tube was heated and cooled from 300 °C to 700 °C by Infrared (IR) furnace at a rate of 10 K/min.

The flowing gas is a mixed gas of nitrogen and steam, and the steam pressure is 31.2 kPa. The gas was obtained by flowing nitrogen into distilled water in a flask that was continuously heated at 70 °C. The gas flow rate was 50 ml/min.

**Figure S3** shows SEM images and EDS mappings of CaO30/MEPCM before and after cycles test. Note that the surface of the sample before the test is CaO, and that the surface of the sample after cycling is Ca(OH)<sub>2</sub>. Focusing on the SEM images of the samples after each cycles, it was found that all of the samples were spherical with a diameter of 30 ~ 40 μm and maintained almost the same shape as the sample before the test. Furthermore, no breakage of the shell or leakage of the core was observed in the SEM image of the surface. From the results of EDS mapping, mapping of Ca element can be confirmed on surface in all samples, and it can be concluded that Ca coating was kept during cycles test.

In summary, CaO30/MEPCM was found to show the great durability in 25 cycles repetition test with infrared furnace.

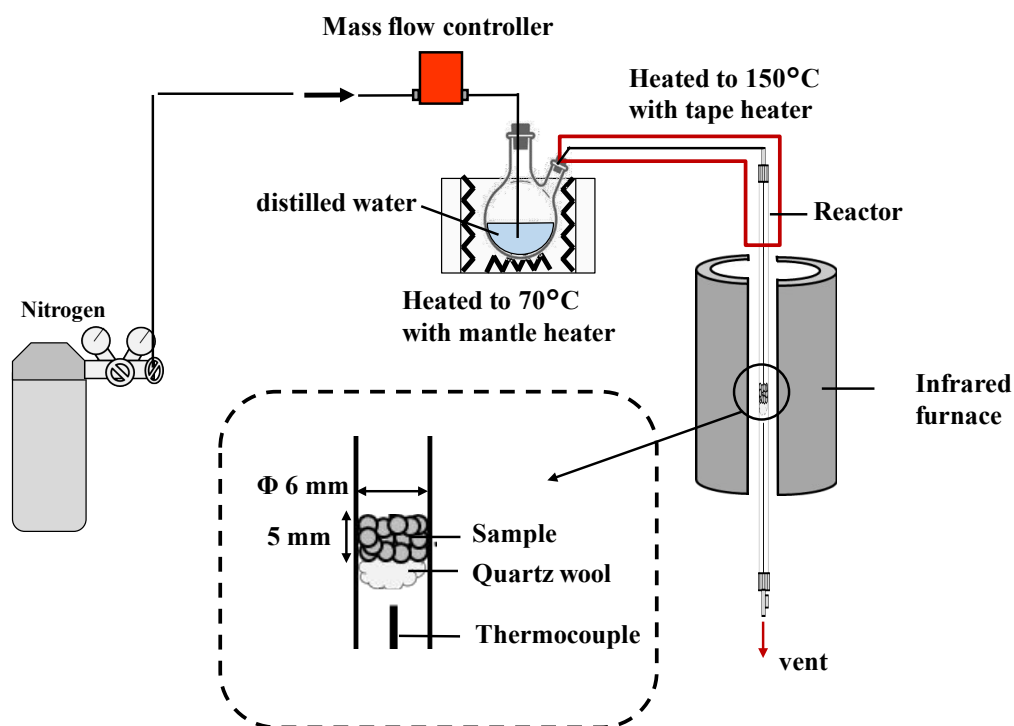


Figure S2. The illustration of apparatus used for 25 cycle durability test of CaO30/MEPCM under steam/N<sub>2</sub> condition.

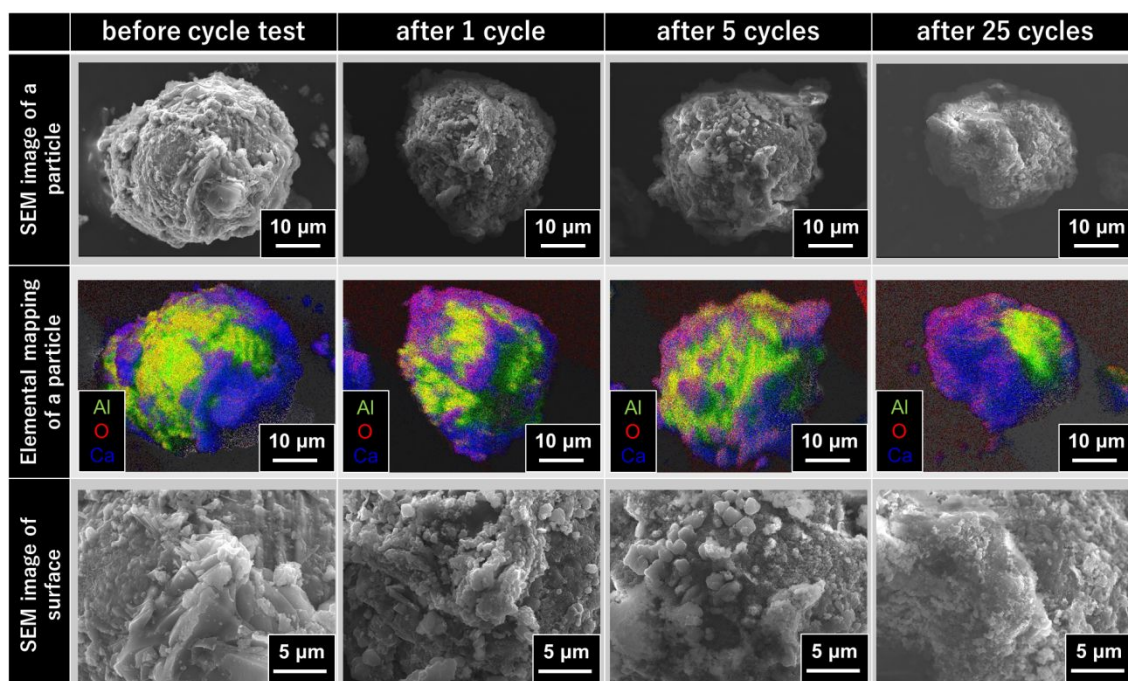


Figure S3. SEM images and EDS mappings of CaO30/MEPCM before and after cycle test.