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Synthesis of Microcapsules with Beeswax Core and Phenol-Formaldehyde Shell by Taguchi Method †

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Abstract: The Taguchi method was used to evaluate the effect of process parameters in the microencapsulation process of beeswax with resorcinol modified phenol-formaldehyde shell. An orthogonal array of 5^3 was constructed to study the effect of process parameters' core to shell ratio, surfactant concentration and agitation speed on control parameter core content. The amount of core content is directly proportional to the heat storing capacity of microcapsules. Surfactant concentration, core to shell ratio, and agitation speed were optimized at 3%, 1:1, and 800 rpm, respectively. The microcapsules synthesized with optimized process parameters values possessed spherical morphology and heat transition enthalpy 148.93 J/g within temperature range 35–62 °C.

Keywords: phase change material; thermal energy storage; beeswax; latent heat

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1. Introduction

Beeswax is a naturally available phase change material (PCM) with peak phase transition enthalpy in the temperature range 60–68 °C and can be used in variety of thermal energy storage applications. Beeswax is studied for its service years and thermal stability [1]. Beeswax can work for 37 years if there were three transitions per day. Beeswax is thermally stable up to 250 °C. It is suitable for using in poultry incubators. Beeswax incorporation in cement and thermal diffusion of composite have been tested [2]. The incorporated beeswax had a melting enthalpy of 242.81 J/g at 62.95 °C. Composite was sandwiched between a hot plate and cold-water circulating assembly. Beeswax has been used as coarse aggregate and rice husk used as fine aggregate in a concrete mixture [3]. Increase in the incorporation percentage of rice husk reduced the compressive strength of concrete. For 5%, 10% and 15% rice husk incorporation, compressive strength was 10.28 MPa, 6.71 MPa and 4.33 MPa. Thermal conductivity of composite was obtained as 0.022 W/m°C and thermal diffusivity of composite was calculated as 2.89×10^{-6} m^2/s. Beeswax was used in a parabolic solar concentrator [4]. It reduced fluctuation in the efficiency of the concentrator at the time of low and high solar radiation. Beeswax melt has been blended with copper oxide, zinc oxide and soot particles [5]. Nanoparticle incorporation increases thermal conductivity but reduces latent heat capacity and transition temperature. Soot particle/ beeswax composite showed the highest conductivity, at 2.89 W/mK. Modified CNT (5%) was incorporated in beeswax by vacuum impregnation [6] which increased thermal conductivity. The composite had reduced melting enthalpy of 115.5 J/g at 60 °C. Beeswax was encapsulated in an AgBr shell by precipitation of AgBr at the beeswax micelle wall [7]. Microencapsulated PCM (MPCM) was thermally stable up to 220 °C. The obtained beeswax microcapsules had the highest melting enthalpy of 99 J/g, at 58 °C.

Taguchi is a method of determining the effect of various process parameters on control parameters. It helps in obtaining correlation between variance of input parameters on

control parameters with lesser formulations. Microencapsulation is most widely used technique for form-stabilizing PCM with multiple process parameters. The studied literature for beeswax form-stabilization in thermal energy storing applications has not evaluated process parameters with the Taguchi method though it can contribute to deep understanding of process. In this study, the effects of process parameters are studied in microencapsulation of beeswax with phenol-formaldehyde shell using Taguchi method for determining optimized values of process parameters.

2. Methods

Phenol and formaldehyde were supplied by SD Fine chemicals Pvt. Ltd., Mumbai, India. SRL Mumbai supplied the beeswax. Polyvinyl alcohol (PVA) was supplied by Loba chemie, Mumbai. Resorcinol, xylene, and ammonium chloride were obtained from Research Lab Pvt. Ltd, Mumbai, India. DI water was utilized throughout experiments.

Microcapsules prepared by suspension polymerization with PVA surfactant. 2.1 g phenol and 0.5 g ammonium chloride were dissolved in PVA-water solution for 30 min and adjusted to pH 7. Beeswax-xylene solution was ultrasonicated at 60 °C for 5 min. This solution was added to 3.35 g of formaldehyde solution and heated to 65 °C under stirring at 500 rpm for 2 h. At pH 3, 0.5 g of resorcinol was added. Reaction was continued for 2.5 h. Obtained microcapsules were vacuum filtered, washed and dried. Core content was obtained by determining change in weight percentage comparing crushed microcapsules dissolved in xylene and dried microcapsules. Core content of microcapsules was calculated in triplets for different levels of parameters and the average value was taken for calculation which is mentioned in Table 1. For sufficient data, the better approach was to construct 5^3 orthogonal array. The microcapsules' morphology was studied with an optical microscope (Olympus BX41) and scanning electron microscope (SEM). Thermal energy storage characteristics were determined with a differential scanning calorimeter (Shimadzu DSC-60).

Table 1. Taguchi L_{25} orthogonal array with signal to noise ratio (SNR).

Run [No]	Surfactant Concentration [g]	Core/Shell Ratio [Moles]	Agitation Speed [rpm]	Core Content [wt%]	SNR [dB]
1	1	0.5:1	400	61.2	35.75
2	1	01:01	600	72	37.14
3	1	1.5:1	800	70.2	36.92
4	1	02:01	1000	50.4	34.04
5	1	2.5:1	1200	19.8	25.93
6	2	0.5:1	400	76.8	37.70
7	2	01:01	600	77.4	37.77
8	2	1.5:1	800	66.6	36.46
9	2	02:01	1000	54	34.64
10	2	2.5:1	1200	77.4	37.77
11	3	0.5:1	400	73.8	37.36
12	3	01:01	600	78	37.84
13	3	1.5:1	800	67.5	36.58
14	3	02:01	1000	73.5	37.32
15	3	2.5:1	1200	70.5	36.96
16	4	0.5:1	400	72.15	37.16
17	4	01:01	600	77.4	37.77
18	4	1.5:1	800	62.4	35.90
19	4	02:01	1000	60	35.56
20	4	2.5:1	1200	69.6	36.85
21	5	0.5:1	400	48	33.62
22	5	01:01	600	61.5	35.77
23	5	1.5:1	800	76.8	37.70
24	5	02:01	1000	63.6	36.06
25	5	2.5:1	1200	60	35.56

3. Results and Discussion

Microencapsulation is a process of engulfing core material with shell material. In the studied polymeric microencapsulation process, dispersed microscaled oleophobic beeswax

grains were coated with resorcinol modified phenol-formaldehyde polymer with an in-situ polymerization process which is depicted in Figure 1. Higher reactivity of resorcinol with formaldehyde helps in faster shell formation.

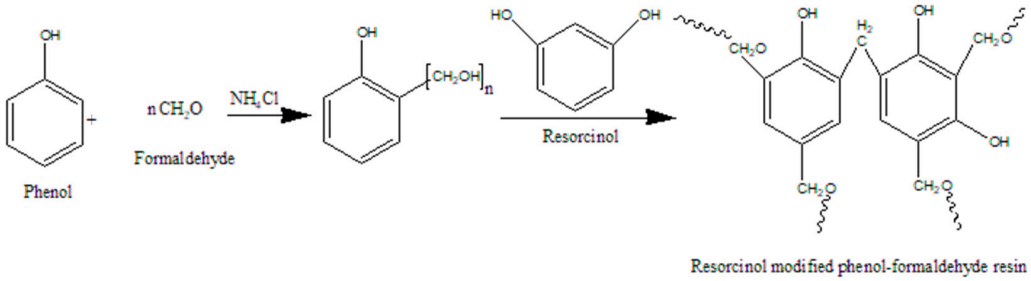

Figure 1. Polymerization reaction.

The amount of core content is directly proportional to the heat storing capacity of microcapsules. The factors that must be optimized include the core to shell ratio, surfactant concentration, and mixing speed. This will be accomplished using the Taguchi technique. Thus, a Taguchi array was constructed with three process parameters and one control parameter. Three factors were changed at five different levels. The core-to-shell ratios were changed as follows: 0.5:1, 1:1, 1.5:1, 2:1, and 2.5:1. Surfactant concentrations were between 1%, 2%, 4%, and 5%. The agitation speed was changed between 400, 600, 800, 1000, and 1200 rpm. Table 1 shows the batches that were investigated. Signal to noise (SN) ratio was calculated with larger the better option in minitab software, in order to maximize control parameter. Changing all of the parameters necessitates the production of 5^3 (125) batches. The optimal parameter values, however, can be achieved in only 25 batches using the Taguchi orthogonal array.

The change in values of process parameters on SN ratio can be understood with Figure 2. The horizontal axis represents values of each process parameter tested in the formulations and the vertical axis represents the SN ratio. The SN ratio showed a steep increase with increase in surfactant concentration up to 3 wt% followed by decrease. Increasing the concentration of surfactant above 3 wt% level resulted in a finer emulsion with lower core content. In the case of core to shell ratio, 1:1 and 0.5:1 ratios in the graph resulted in an increase in core content and thus increase in SN ratio. Reduction in SN ratio above 0.5:1 level is from decrease in the shell thickness of microcapsules. A ruptured thin shell may have a low core content. The figure depicts speed to SN ratio correlation in the last section. Increase in speed up to 800 rpm aids in the development of core/shell morphology; however, beyond this speed, the shell may rupture. This relation can be clearly observed with SN values. Considering the effect of SN ratio and in order to maximize core content, the optimum values for surfactant concentration, core to shell ratio, and agitation speed were selected as 3%, 1:1, and 800 rpm respectively.

A batch was synthesized with optimum parameter values and characterized for its thermophysical properties. In image J application, the size of the MPCM with optimum process parameter values was determined by taking the average of 100 measurements. The average size was 62.61 µm. Figure 3a depicts an optical micrograph of microcapsules. Figure 3b is a SEM image of a single microsphere with a diameter of 70.33 µm. With image analysis, successful microcapsule formation can be postulated. The microcapsules that were created were spherical in form. The reaction conditions proposed yield tiny sized microcapsules that may be easily utilized in coating applications with thinner coatings.

In the temperature range of 35–62 °C, the melting enthalpy of MPCM was 148.93 J/g. Figure 4 depicts the melting thermogram. The temperature range found for the phase transition temperature may be utilized to store various food items such as hot served cuisines, building materials, and solar energy storage materials. The two peaks of the phase transition allow heat to be stored across a wider temperature range.

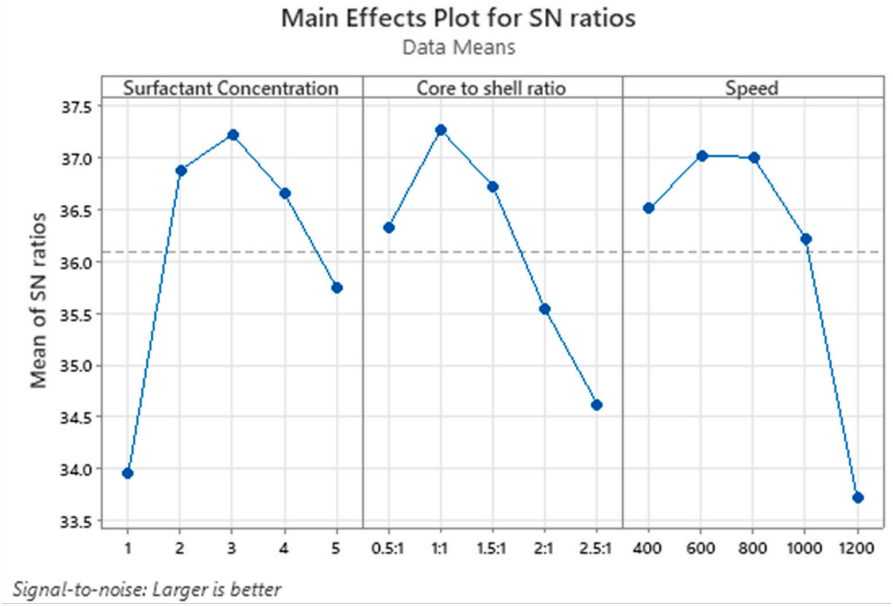

Figure 2. Main effects plots of the SN ratio.

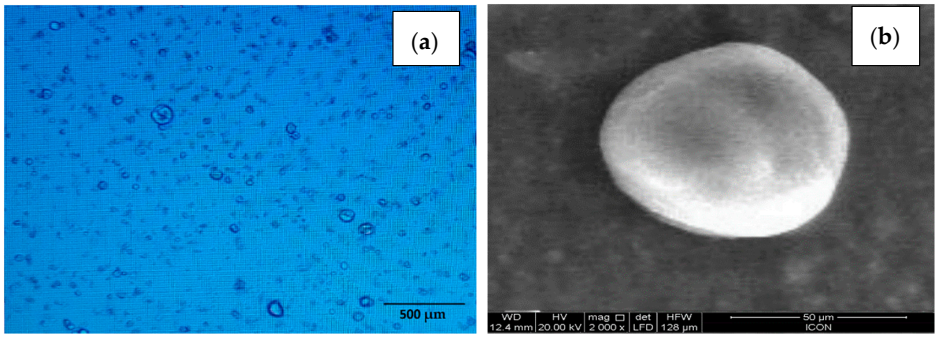

Figure 3. Image analysis with (**a**) Optical micrograph (**b**) SEM micrograph.

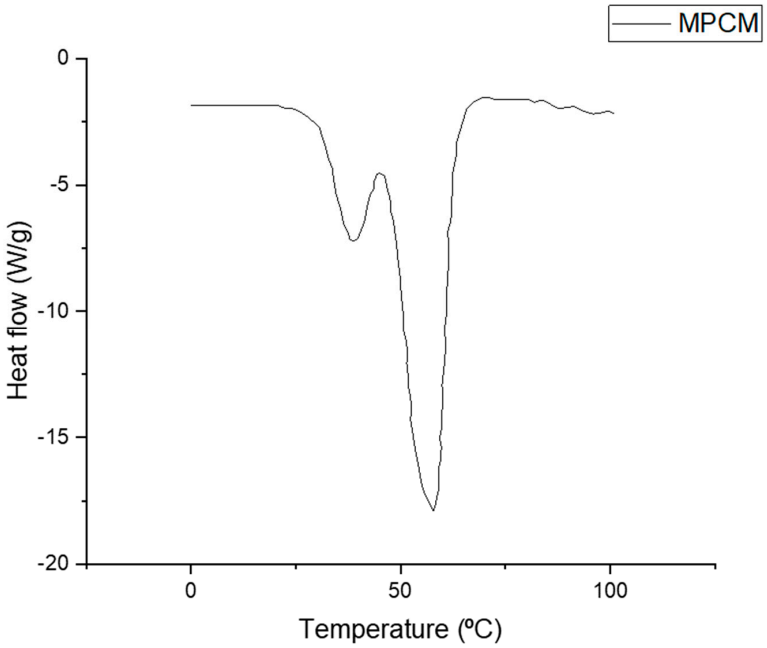

Figure 4. DSC thermogram of an optimized batch.

4. Conclusions

The impact of surfactant concentration, core to shell ratio, and agitation speed on MPCM core content was investigated. The optimal values for surfactant concentration, core to shell ratio, and agitation speed were 3%, 1:1, and 800 rpm, respectively. MPCM's spherical shape and micrometer size allow it to be used for a wide range of applications. The melting enthalpy and temperature range of the phase transition are appropriate for various thermal energy storage applications.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/ecsoc-25-11671/s1.

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References

1. Kabir, M.S.; Yola, I.A. Beeswax as a low temperature phase change material for thermal storage. *FUDMA J. Sci.* **2020**, *4*, 764–769.
2. Hakim, I.I.; Putra, N.; Agustin, P.D. Measurement of PCM-concrete composites thermal properties for energy conservation in building material. *AIP Conf. Proc.* **2020**, *2255*, 030066. [CrossRef]
3. Thaib, R. Experimental Study of Beeswax / Rice Husk Ash Phase Changes Material as Energy Storage in Concrete. *Eur. J. Eng. Technol. Res.* **2021**, *6*, 34–37. [CrossRef]
4. Ibne, F.; Imam, A.; Rafiqul, M.; Beg, A.; Rahman, S. Performance Analysis of a Natural Phase Change Material-Based Solar Collector with Compound Parabolic Concentrator at Different Flow Rates. *Appl. Model. Simul.* **2020**, *4*, 237–245.
5. Andoshe, D.M.; Essay, B.; Ahemd, O.; Merga, A.; Tesefaye, D.; Gezahegn, N.; Miressa, T.; Meri, T. Tuning Thermal Properties of Beeswax Incorporating ZnO, CuO and Soot Particles. *Ethiop. J. Sci. Sustain. Dev.* **2020**, *7*, 79. [CrossRef]
6. Putra, N.; Rawi, S.; Amin, M.; Kusrini, E.; Kosasih, E.A.; Indra Mahlia, T.M. Preparation of beeswax/multi-walled carbon nanotubes as novel shape-stable nanocomposite phase-change material for thermal energy storage. *J. Energy Storage* **2019**, *21*, 32–39. [CrossRef]
7. Yuan, H.; Bai, H.; Chi, M.; Zhang, X.; Zhang, J.; Zhang, Z.; Yang, L. A novel encapsulation method for phase change materials with a AgBr shell as a thermal energy storage material. *Energies* **2019**, *12*, 717. [CrossRef]