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Synthesis and study of microcapsules with beeswax core and phenol-formaldehyde shell by Taguchi method †

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Abstract: Phenol-formaldehyde shelled phase change material microcapsules (MPCM) were fabricated and their processing parameters were analyzed with Taguchi method. Core to shell ratio, surfactant concentration and speed of mixing are the parameters which were optimized in five levels. The optimized values for surfactant concentration, core to shell ratio and agitation speed were 3%, 1:1 and 800 rpm. The obtained microcapsules were spherical in shape. The melting enthalpy of MPCM synthesized with optimized processing parameters was 148.93 J/g in 35-62 ^{0}C. The obtained temperature range of phase transition temperature can be used for storing different food articles such as chocolate and hot served foods.

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

1. Introduction

Beeswax has been used as phase change material (PCM) in various applications such as building applications, solar energy storage applications owing to its phase change temperature around 60^{0}C. Beeswax used in waterborne coating for preparing hydrophobic thermoregulating coating [1]. Dispersion of Beeswax/ perfluorinated copolymer (used as encapsulant) / silica nanoparticle made with homogenizer to give 350 nm particle size. The obtained material had phase transition enthalpy of 84 J/g at 61^{0}C. Addition of thermally conductive nanomaterials in beeswax composite improves heat dissipation characteristics. Beeswax mixed with copper, aluminium and graphite nanoparticle to increase conductivity [2]. The prepared composite filled in 55 mm capsules and placed in heat storage tank. Graphite/ beeswax composite outperform with less time for charging and high discharging time as compared to other nanocomposite. Bentonite clay integrated with beeswax and mixed with concrete [3]. Heat absorption of composite with PCM increased by 6.67%, but compression strength reduced. Modified carbon nanotubes (CNTs) (5%) incorporated in beeswax by vacuum impregnation [4]. Incorporation of CNTs increased thermal conductivity. The composite had reduced melting enthalpy of 115.5 J/g at 60^{0}C. Black beeswax incorporated in prototype roof model made from MDF sheet and covered with EPS foam [5]. Increase in temperature in night by 3.6 ^{0}C was observed. Simulation study projected 67% energy saving. Beeswax stored in container with PV panel [6]. It reduced heat waste from panel and increased voltage of generation

The microencapsulation method is an exhaustively used technique for PCM shape-stabilization. This technique has been used in broad spectrum of applications such as building, medical, electronics, food, etc. Polymeric encapsulation is characterized with high toughness and good heat transfer property due to large surface area of capsules. In this paper, phenol-formaldehyde shelled PCM microcapsules are fabricated and their properties are studied.

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2. Materials and Methods

Phenol, formaldehyde purchased from SD Fine chemicals private limited. Beeswax was procured from SRL, Mumbai. Sodium dodecyl sulfate (SDS) and polyvinyl alcohol (PVA) bought from loba chemie. Resorcinol, ammonium chloride, xylene purchased from research lab pvt. Ltd. Deionized water (DI) used in all the experimental work.

Beeswax/phenol-formaldehyde core/shell particles were prepared by suspension polymerization technique. 5 wt.% aqueous solutions of PVA prepared in DI water with varying amount of SDS surfactant. The solution mixed by magnetic stirring at 500 rpm until PVA dissolution. Under agitation 2.1 g phenol and of 0.5g ammonium chloride were dissolved in PVA solution for 30 minutes. The pH of PVA solution adjusted to 7-8 using ammonia solution. Different amount of PCM was added to 10 ml of xylene in beaker and subjected to agitation for 5 minutes with magnetic stirrer at 60 ^{0}C. Emulsion was allowed to form by adding PVA solution to PCM solution under ultrasolnication for 30 minutes. 3.35 g of 37 wt.% aqueous solution of formaldehyde and ultrasonicated solution was added in another heated container placed inside heater. Solution was slowly added to container and maintained at 65 ^{0}C under stirring at 500 rpm for next 2 hours. Then, 5 wt.% of HCl was added to maintain pH at about 3-4 and 0.5 g of resorcinol was added. Reaction was continued at same temperature for next 2.5 hours. Microcapsules were recovered by filtration under vacuum. The microcapsules rinsed with water and washed with xylene and dried for 24 hours.

Accurately weighed sample crushed and stirred in xylene for 1 hour at 70 ^{0}C under magnetic stirring. The microcapsules rinsed with water and washed with xylene and dried for 24 hours. The core content is the percentage of microcapsule weight difference before and after the treatment. Core content was calculated taking average of 3 readings. Olympus BX41 optical microscope was used to measure size of microcapsule. The size of microcapsule was calculated taking mean of 100 readings from Image J software. Differential scanning calorimeter (Shimadzu DSC-60) was used to determine enthalpy and phase transition temperature of microcapsules.

3. Results

Core to shell ratio, surfactant concentration and speed of mixing are the parameters which need to optimize. This can be done thoroughly with Taguchi method. Three parameters were varied in five levels. The core to shell ratio was varied as 0.5:1, 1:1, 1.5:1, 2:1 and 2.5:1. The surfactant concentration varied as 1, 2, 3, 4 and 5%. Agitation speed varied as 400, 600, 800, 1000 and 1200 rpm. The batches studied are shown in table 1. Varying all the parameters require to fabricate 5^3 (125) batches. But with the help of Taguchi orthogonal array, the optimized parameters can be obtained with only 25 batches. Increasing core content will increase thermal energy storage property. Thus, larger the better analysis was chosen.

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

Run [Nos.]	Surfactant concentration [g]	Core-to-shell ratio [in moles]	Agitation speed [rpm]	Core content [wt%]				SNR [dB]
				R1	R2	R3	Average	
1	1	0.5:1	400	59.4	63	61.2	61.2	35.73503
2	1	01:01	600	77.4	68.4	70.2	72	37.14665
3	1	1.5:1	800	67.5	72	71.1	70.2	36.92674
4	1	02:01	1000	52.2	48.6	50.4	50.4	34.04861

5	1	2.5:1	1200	14.4	27	18	19.8	25.9333
6	2	0.5:1	400	75.6	75.6	79.2	76.8	37.70722
7	2	01:01	600	73.8	77.4	81	77.4	37.77482
8	2	1.5:1	800	68.4	68.4	63	66.6	36.46948
9	2	02:01	1000	52.2	57.6	52.2	54	34.64788
10	2	2.5:1	1200	73.8	77.4	81	77.4	37.77482
11	3	0.5:1	400	66.6	72	82.8	73.8	37.36113
12	3	01:01	600	69	81	84	78	37.84189
13	3	1.5:1	800	57	73.5	72	67.5	36.58608
14	3	02:01	1000	67.5	76.5	76.5	73.5	37.32575
15	3	2.5:1	1200	70.5	67.5	73.5	70.5	36.96378
16	4	0.5:1	400	70.5	72.45	73.5	72.15	37.16473
17	4	01:01	600	79.2	79.2	73.8	77.4	37.77482
18	4	1.5:1	800	62.4	66	58.8	62.4	35.90369
19	4	02:01	1000	61.2	58.8	60	60	35.56303
20	4	2.5:1	1200	66	69.6	73.2	69.6	36.85218
21	5	0.5:1	400	48	45	51	48	33.62482
22	5	01:01	600	57	66	61.5	61.5	35.7775
23	5	1.5:1	800	78	74.4	78	76.8	37.70722
24	5	02:01	1000	64.8	66	60	63.6	36.06914
25	5	2.5:1	1200	60	52.5	67.5	60	35.56303

The effect of parameter values on core content can be studied with main effects plots of SN ratio which is shown in figure 1.

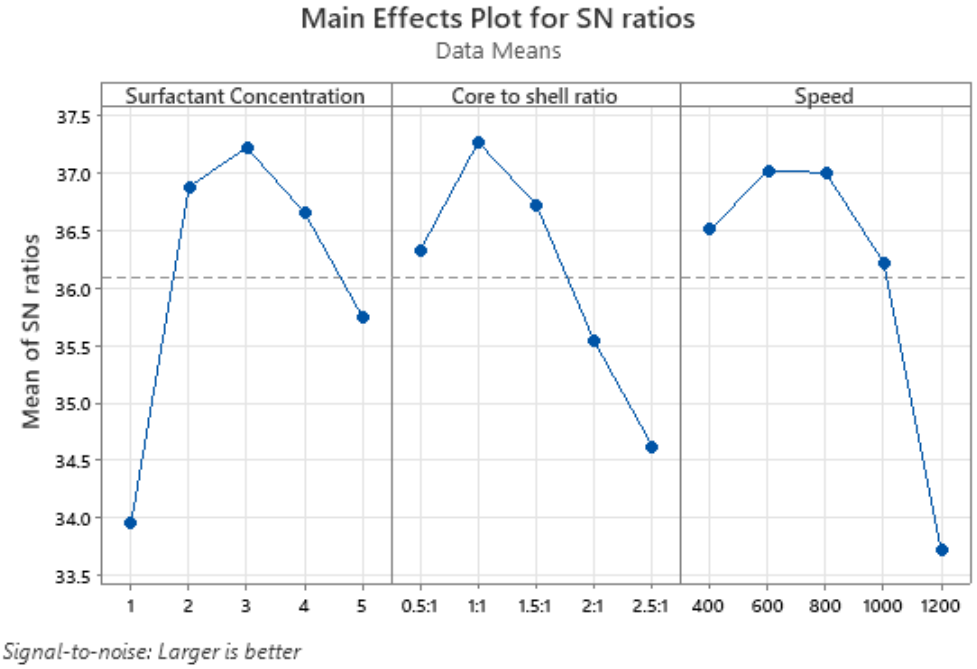

Figure 1. Main effects plots of SN ratio

Increasing surfactant concentration gave finer emulsion with better dispersion. As the surfactant concentration increases above 3 wt.%, core content reduces. This is the reason for decrease in SNR value. Increase in core content was observed for core to shell ratio

1:1 and 0.5:1. Further increase in the ratio reduced shell thickness. The ruptured thin shell can show low core content. Increase in speed up to 800 rpm help in formation of core/shell morphology, Increasing the speed above this may rupture the shell. So, the optimized value for surfactant concentration, core to shell ratio and agitation speed are 3%, 1:1 and 800 rpm.

The size of MPCM with optimized parameters was calculated taking mean of 100 readings in image J software. Thus, the size obtained was 62.61 μm. The optical micrograph was shown in Figure 2. The obtained microcapsules were spherical in shape. The suggested parameters of reaction give small sized microcapsules which can be easily used in coating applications with smaller thickness coatings.

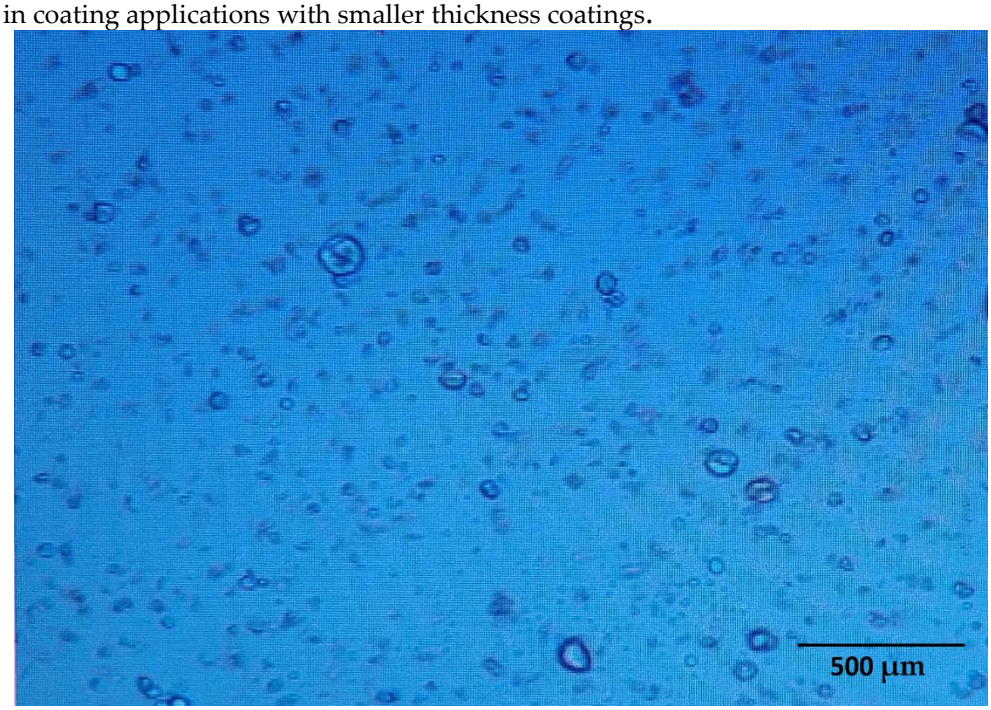

Figure 2. Optical micrograph of optimized batch

The melting enthalpy of MPCM was 148.93J/g in the range of 35-62 ^{0}C. The melting thermogram can be seen in figure 3. The obtained temperature range of phase transition temperature can be used for storing different food articles such as chocolate and hot served foods, building materials and solar energy storing materials. The two peaks of phase transition allow for storing the heat for larger temperature range.

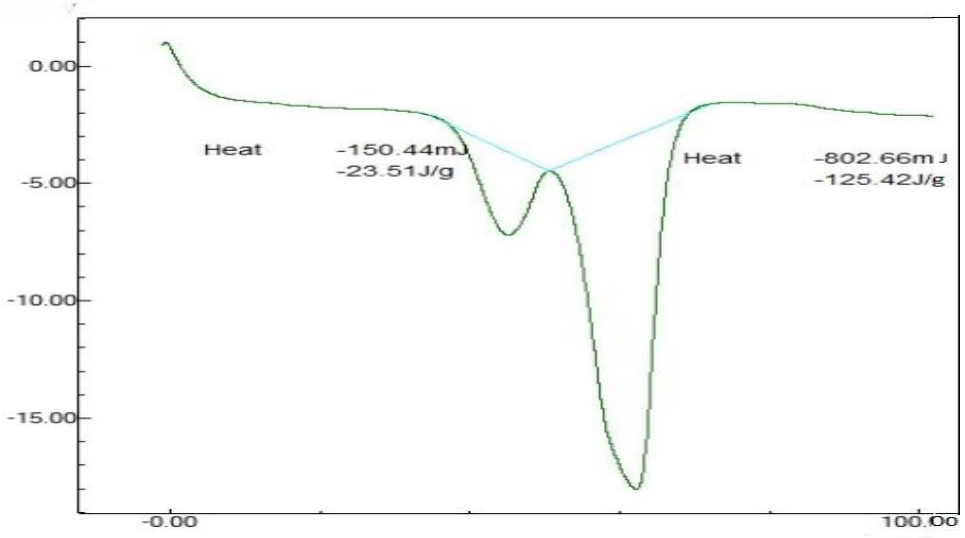

Figure 3. DSC thermogram of optimized batch

4. Discussion

The effect of surfactant concentration, core to shell ratio and agitation speed on core content of MPCM was studied. The optimized value for surfactant concentration, core to shell ratio and agitation speed were 3%, 1:1 and 800 rpm. The structure of MPCM is spherical and size in micrometers allow its use for myriad applications. The melting enthalpy and temperature range of phase transition are suitable for thermal energy storing applications.

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