TECHNICAL NOTE: MELT DISPERSION TECHNIQUE FOR PREPARING PARAFFIN WAX MICROSPHERES FOR CELLULOSE ENCAPSULATION

Yuanfeng Pan
Research Scholar
State Key Laboratory of Pulp and Paper Engineering
South China University of Technology
Guangzhou 510640, China
E-mail: sunshaolong328@126.com

Shaolong Sun
PhD Candidate
Beijing Key Laboratory of Lignocellulosic Chemistry
Beijing Forestry University
Beijing, China
E-mail: sunshaolong328@126.com

Qingzheng Cheng
Research Associate
E-mail: qzc0007@auburn.edu

Yusuf Celikbag
PhD Candidate
E-mail: yzc0003@auburn.edu

*Brian K. Via**†*
Associate Professor and Director
Forest Product Development Center
Auburn University
Auburn, AL
E-mail: bkv0003@auburn.edu

Xiaoying Wang
Associate Professor
E-mail: xyw@scut.edu.cn

*Runcang Sun***
Professor
State Key Laboratory of Pulp and Paper Engineering
South China University of Technology
Guangzhou 510640, China
E-mail: rcsun3@bjfu.edu.cn

(Received February 2014)

* Corresponding author
† SWST member

Wood and Fiber Science, 46(4), 2014, pp. 594-599

Abstract. A practical and convenient approach for making paraffin wax microspheres with a melt dispersion technique was reported in this study. Surfactants were melted in water by water bath and then added to a flask after the wax was completely melted with stirring. Paraffin wax microspheres were generated by cooling. The obtained microspheres exhibited uniform diameters in the range of 10-60 μm observed with a scanning electrical microscope and were mainly dependent on the surfactant ratio. Encapsulated microcrystalline cellulose particles with the previously mentioned conditions were also generated and demonstrated the possibility of encapsulating microcrystalline cellulose with some acceptable agglomeration, although some encapsulated individually. Encapsulation of cellulose could be beneficial if agglomeration could be minimized and the encapsulated microcapsules could be dispersed during blending for wood composites manufacture.

Keywords: Wax microsphere, microcapsule, melt dispersion, cooling phase separate, microcrystalline cellulose.

INTRODUCTION

Much research has been performed in the medical field of the encapsulating technique, which is a simple and effective method to distribute drugs in a time-released fashion. The benefits of encapsulation include protection against oxygen, heat, and light (Uddin et al 2001; Cahill et al 2002), controlled evaporation and release (Solomon et al 2012), coverage of unpleasant odors (Schrooyen et al 2001), and improvement of material appearance. Other fields include the food, feed, chemical, drug, makeup, materials industries (Sánchez-Silva et al 2012), etc. Optimization of shell material is an important research area because the final microscopic size and surface or interface area needs to be well controlled for final product performance (Umer et al 2011).

Waxes are common materials that are currently used in the forest composites industry for additional moisture resistance and consequently acceptable panel dimensional stability. Other significant advantages across a broader array of industries include superior reproducibility without special instrumentation, lower production costs, good long-term stability under storage conditions, and environmental friendliness (Kamble et al 2004). For wood composite applications, there is also the benefit of good flow properties and uniformity after preheating to melt temperature. Many reports focus on their applications, for example on preparation of microcapsules for self-healing curing agent of thermoset resins (Rule et al 2005), decreased rate of water flow in material capillaries (Lesar and Humar 2011), and significant increases in the dimensional stability under wet conditions (Kurt et al 2008). In addition, wax additives were incorporated into aqueous wood preservatives to decrease cracking and improve the appearance of treated wood when exposed outdoors (Evans 2009).

The potential to use wax microspheres in wood composites has not been explored. A potential application of wax microspheres could be the encapsulation of cellulose for composite applications. Recent investigations have found cellulose agglomeration during dispersion resulting in limited improvements or even decreases in particleboard mechanical properties, which was perhaps caused by spring-back shearing forces at the cellulose–adhesive–wood interface after pressing (Atta-Obeng et al 2012). Conversely, when cellulose was properly dispersed and tested in a neat phenol–formaldehyde matrix, significant improvements in polymer mechanical properties occurred (Atta-Obeng et al 2013). This suggests that significant benefits for the wood composite industries may be possible, but dispersion issues in the manufacturing process will have to be overcome.

Hence, to investigate the effect of process parameters on the size and properties of spheres as well as to further investigate the encapsulation of cellulose for wood-based composites, we chose paraffin wax. A practical and convenient approach for making microspheres by melt dispersion was reported in this study. This strategy is based on the melting of wax and combining with water to form an emulsion with stirring. Solidified spheres were then obtained by cooling

in cold water or an ice bath, and the spheres were observed under an optical and scanning electrical microscope (SEM). Finally, the procedure was validated in the presence of microcrystalline cellulose (MCC) in which cellulose particles were encapsulated. The shape and size of the encapsulations were controlled by dialing in the proper ratio of surfactants.

MATERIALS AND METHODS

Paraffin wax (purchased from Gulf Oil Corporation, Houston, TX) microspheres were produced by a modified microcapsule synthesis method (Herbig et al 1965) consisting of melting condensation and dispersion in water. Typically, the temperature of the thermostat water bath was first adjusted to 70°C, which was 5-10°C higher than the melting point of wax. About 100 mL of solution containing water and two surfactants (Span 60 and Tween 40, purchased from VWR Chemical Co., Radnor, PA) were then placed in a common flat flask and stirred with a magnetic stirrer. The ratio of Span and Tween was marked as S:T = 7:3 for 0.35 g Span and 0.15 g Tween. Two ratios of Span and Tween (8:2 and 7:3) were chosen because they were the best ratios to make large and small wax spheres. Subsequently, the dispersion was emulsified and stirred at 1000 rpm for 5 min after 10.0 g of paraffin wax was added to the flask and melted completely in a 70°C water bath. To solidify paraffin wax spheres, the next step consisted of adding cold water or placing the flask into an ice bath while maintaining mechanical stirring. Finally, the paraffin wax microspheres were collected by filtration with No. 2 filter paper and washed three times with water or ethanol.

The MCC (purchased from VWR Chemical Co., Radnor, PA) had an average diameter of 90 μm. The paraffin wax microcapsules were obtained by mixing the MCC with paraffin wax and surfactants while maintaining the temperature at 70°C coupled with continuous mechanical stirring. A mixture containing 10.0 g paraffin wax and 0.5 g surfactants with different ratios was placed in a beaker and then stirred at 1000 rpm for 5 min after the mixture was melted completely at 70°C. Then cellulose coating was performed by adding 40 g MCC to the mixture, maintaining the stirring at 1000 rpm, and keeping the temperature at 70°C for 10 min. Finally, the temperature was decreased to room temperature and the mixture was cooled with stirring at 300 rpm. All final products were observed by an optical microscope (BX 53 system; Olympus, Tokyo, Japan) and SEM (EVO 50VP; ZEISS, Jena, Germany).

RESULTS AND DISCUSSION

Figure 1 shows SEM images at different magnification and the different ratios of Span and Tween surfactants, 8:2 and 7:3, respectively. Other ratios were explored, but these ratios yielded the greatest differences in wax particles size. The images show that sphere size was dependent on surfactant ratio. The diameter of the obtained microspheres could be tuned from an average of 10-60 μm by altering the surfactant ratio. This size range could be useful for many material applications, especially for wood-related composites manufacture and other wood-based products. One example is adding in liquid resins for spraying to increase composite water resistance (Wang and Xing 2010). For example, a ratio of 8:2 was necessary for larger spheres (Fig 1a-b), whereas a ratio of 7:3 yielded smaller particles (Fig 1c-d). With increasing S:T ratios, final spheres were larger in size and wider in size distribution. The microsphere sizes were 10-60 μm, but most of them were about 20-30 μm at the S:T ratio of 8:2 (Fig 1a-b). Although the sizes ranged from 10 to 34 μm, most sizes were approximately 10-20 μm at the ratio of 7:3 (Fig 1c-d). One possible reason for the bigger size might be that the emulsion droplets agglomerated into larger drops. The images demonstrated that the spheres were fairly standard, exhibited a smooth surface, and lacked agglomeration. Also, for a simple and versatile method, there was minimum difference in sphere size and appearance for a specific S:T ratio treatment. These results indicated that it is possible to control the melt dispersion technique to manufacture different-sized paraffin wax microspheres.

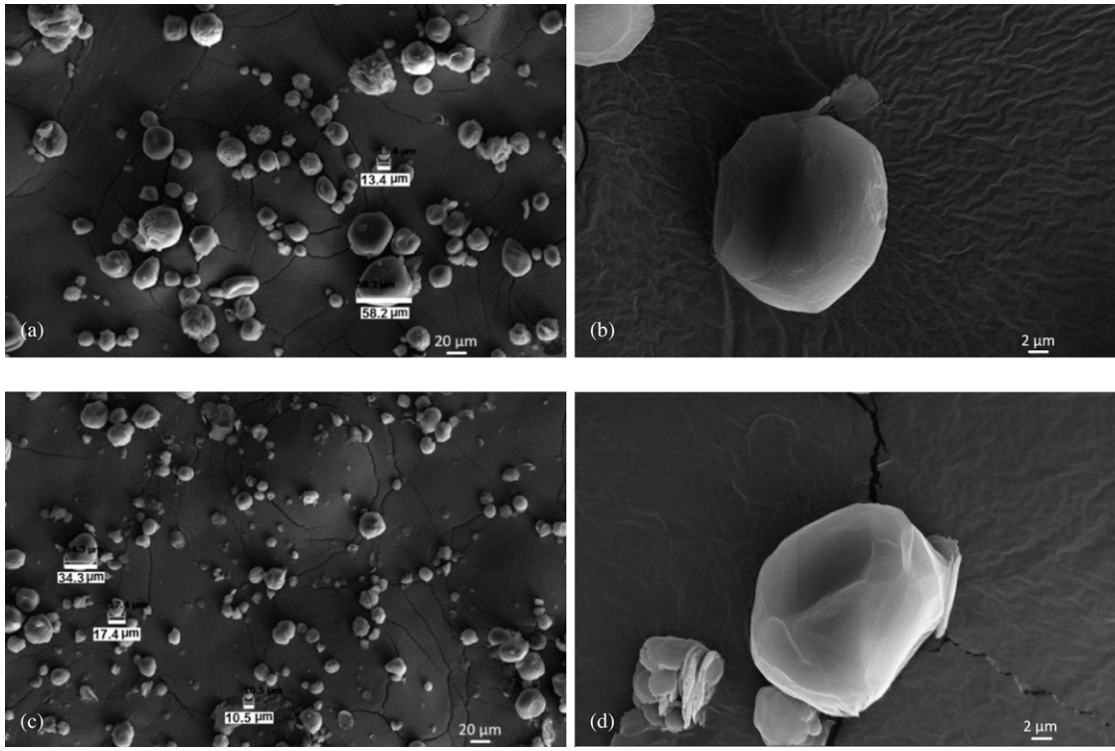

Figure 1. Scanning electron microscope images for paraffin wax microspheres with different surfactant ratios: (a-b) 8:2 and (c-d) 7:3.

Figure 2 demonstrates that the encapsulating method was successful and yielded large quantities of encapsulated cellulose particles with little change in length. These insulated particles could be useful for many hydrophobic adhesive and coating applications. There were some cellulose agglomerations within a particle, but this appeared minimal and is probably acceptable

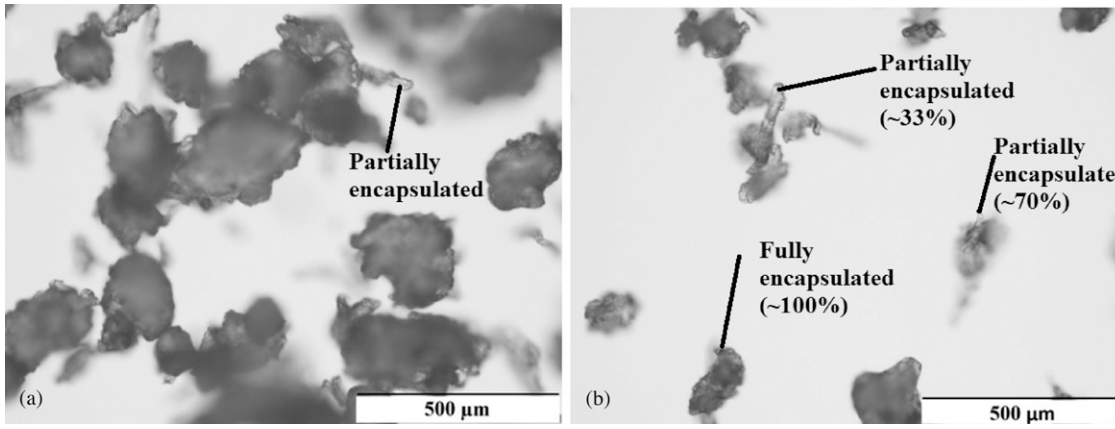

Figure 2. Optical microscope images of encapsulated cellulose particles by wax and surfactants with different ratios: (a) 8:2 and (b) 7:3.

for many wood product applications. For the surfactant ratio of 8:2, almost all MCC particles were completely coated (Fig 2a). Conversely, at 7:3, some MCC particles were not totally encapsulated, whereas others were well encapsulated. Also, the wax-covered percentage of MCC particles can be estimated from the images (Fig 2b). These images suggest that the parameters used during the synthesis of smaller paraffin wax microspheres were also suitable for the enclosure of isolated cellulose particles.

CONCLUSIONS

This study examined a cost-effective technique for making paraffin wax microspheres using the melt dispersion technique and modifying the surface properties of MCC by an encapsulated approach and with the help of surfactants. SEM and optical images demonstrated that this method yielded large quantities and high uniformity of microspheres with diameters ranging from 10 to 60 μm depending on the different surfactant ratios. The present method for producing microspheres does not require special or professional apparatus and equipment. This study demonstrated that a target encapsulation morphology can be achieved through precise control of surfactant ratio. For example, to completely encapsulate MCC particles with big sphere sizes, a Span and Tween surfactant ratio of 8:2 was required, whereas partly encapsulated MCC particles were achieved at a Span and Tween ratio of 7:3. This process has many advantages, eg natural and nontoxic, environmentally friendly, changeable size for different applications, and easy process. The approach appears especially suitable for those industrial applications for which large quantities of paraffin wax microspheres or microcapsules with wax as shell materials are required.

ACKNOWLEDGMENTS

This work was in alignment with 5-yr goals set forth in Hatch-Project No. ALA-031-1-09020 by B. K. Via. This work was also financially supported by the China Scholarship Council. This work will be further leveraged by a recent Alabama Agricultural Experiment Station (AAES) Equipment Grant in which equipment was awarded to spray dry microcapsules. Regions Bank is also acknowledged for their financial support. This project was in alignment with their goal of value-added product development from trees.

REFERENCES

Atta-Obeng E, Via BK, Fasina O (2012) Effect of microcrystalline cellulose, species, and particle size on mechanical and physical properties of particleboard. Wood Fiber Sci 44(2):227-235.

Atta-Obeng E, Via B, Fasina O, Auad M, Jiang W (2013) Cellulose reinforcement of phenol formaldehyde: Characterization and chemometric elucidation. International Journal of Composite Materials 3(3):61-68.

Cahill SM, Upton ME, McLoughlin AJ (2002) Bioencapsulation technology in meat preservation. Pages 239-266 *in* A Durieux and J Simon, eds. Applied microbiology. Springer, Houten, The Netherlands.

Evans PDW-HRRB (2009) Wax and oil emulsion additives: How effective are they at improving the performance of preservative-treated wood? Forest Prod J 59(1/2):66-71.

Herbig AJ, Franklin HJ (1965) Process for making capsules. US Patent 3,161,602.

Kamble R, Maheshwari M, Paradkar A, Kadam S (2004) Melt solidification technique: Incorporation of higher wax content in ibuprofen beads. AAPS Pharm Sci 5(4):75-83.

Kurt R, Krause A, Militz H, Mai C (2008) Hydroxymethylated resorcinol (HMR) priming agent for improved bondability of wax-treated wood. Holz Roh Werkst 66(5):333-338.

Lesar B, Humar M (2011) Use of wax emulsions for improvement of wood durability and sorption properties. European Journal of Wood and Wood Products 69(2):231-238.

Rule JD, Brown EN, Sottos NR, White SR, Moore JS (2005) Wax-protected catalyst microspheres for efficient self-healing materials. Adv Mater 17(2):205-208.

Sánchez-Silva L, Gutiérrez N, Sánchez P, Romero A, Valverde JL (2012) Smart microcapsules containing nonpolar chemical compounds and carbon nanofibers. Chem Eng J 181-182:813-822.

Schrooyen PMM, Meer Rvd, Kruif CGD (2001) Microencapsulation: Its application in nutrition. Proc Nutr Soc 60(4):475-479.

Solomon B, Sahle FF, Gebre-Mariam T, Asres K, Neubert RHH (2012) Microencapsulation of citronella oil for mosquito-repellent application: Formulation and in vitro permeation studies. Eur J Pharm Biopharm 80(1):61-66.

Uddin MS, Hawlader MNA, Zhu HJ (2001) Microencapsulation of ascorbic acid: Effect of process variables on product characteristics. J Microencapsul 18(2):199-209.

Umer H, Nigam H, Tamboli AM, Nainar MSM (2011) Microencapsulation: Process, techniques and applications. International Journal of Research in Pharmaceutical and Biomedical Sciences 2(2):474-481.

Wang S, Xing X (2010) Wood adhesives containing reinforced additives for structural engineering products. US Patent Application, No. 2010/0285295, filled in 2008.