A Facile Coating Method for Superhydrophobic Magnetic Composite Sheet from Biodegradable Durian Peel for Electromagnetic Wave Absorbance Application

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Keywords: Superhydrophobic, Durian peel, Stearic acid

Abstract. Most of the electromagnetic (EM) wave absorbers are commonly made from polymer-based materials. A large number of polymers are resistant to the environmental degradation and are thus responsible for the buildup of polymeric solid waste materials. These solid wastes cause acute environmental problems and remain undegraded for quite a long time. In a view of the awareness and concern for the problems created by the polymeric solid wastes, new biodegradable cellulose composite with low cost and nontoxic materials, have been designed and developed. However, the properties of natural fibers that tends to absorb water, thus limiting their application. In this study, precipitated calcium carbonate (PCC) was added with stearic acid (SA) in order to generate a hydrophobic coating formulation. PCC works as filler and SA act as surface hydrophobic modification agent. Polymer latex was then added to the coating compound as the binder. The composite surface morphology was inspected using scanning electron microscope (SEM). Results show that durian peel composite sheet had successfully achieved a superhydrophobic surface with a water contact angle of 154.85° which exceed 150°.

Introduction

The quick-tempered growth of telecommunication application in industrial, medical and also military field has led to the high frequency of electromagnetic (EM) wave [1]. This high frequency generated electromagnetic interference (EMI) which may cause disruption in those applications. Therefore, EM wave absorber with the capability of absorbing these unwanted EM signals is invented.

Presently most of the EM wave absorbers are made from polymers due to the light weight and flexibility. However, they are environmental degradation resistance and the accumulation of their solid wastes may cause serious environmental problems. Due to that, a new EM wave absorber from natural fiber is designed and produced in this research with the goal of overcoming the problem. Furthermore, the selection of natural fiber as the absorber material is due to its sustainable ability, low cost and renewability.

Due to the great prominence in research and real-life applications including the prospective in industrial applications, superhydrophobic surfaces with a water contact angle higher than 150° have attracted increasing interest [2-4]. The wettability characteristic of a solid surface is generally driven by the chemical composition and the geometrical structure of the surface [5-7]. In fabricating superhydrophobic surface, two techniques are adopted: (1) forming more rough structure on the
hydrophobic material surface [8-10]; (2) modifying the rough surface to lower the surface energy [9,10]. There were a lot of approaches to achieve superhydrophobic surface, such as multi-layer deposition [11,12], sol–gel process [13-15], chemical vapor deposition [16] and solution-immersion [17].

Although the selection of natural fiber as an alternative to polymer is perfect in terms of availability, less cost and renewability, however, it is very hygroscopic. It tends to have high water and moisture absorptions because of the presence of hydrophilic groups, mainly hydroxyl, carboxyl or sulfonic groups [18]. The high hygroscopicity may cause the magnetic composite sheet to absorb water vapor easily from the environment and loss its valuable mechanical properties. This is the reason the coating method by using the hydrophobic stearic acid is applied in this study. This method was selected based on the factors of simplicity, non-toxicity and cost efficiency.

Durian peel is a new material used in this study, selected for its properties such as hard shell texture and high pore volumes [19]. The higher the pore distribution in the fiber, the higher the degree of loading of the magnetic particles should be and the better the quality of the magnetic sheet.

Experimental Method

Sample Preparation. Durian peels were cleaned up and repeatedly washed with tap water to remove grime and unnecessary materials on the peel’s surface. Then, they were cut to about 2 to 3 cm in length and about 0.4 cm in thickness. Next, the durian chips were dried at 50 °C for 24 hours in drying oven to remove the moisture content.

Pulping. After the drying process, dried durian peel underwent the process of pulping to remove lignin from cellulose. The selected pulping process was soda pulping process and it was conducted by using rotary digester machine. The use of sodium hydroxide (NaOH) in making wood pulp is called as soda pulping process. The process was carried out with 17 % concentration of NaOH. About 300 g (oven-dried) of durian chips was placed into the digester. The ratio of durian chips to cooking liquor was 1:7. The cooking temperature was set at 170 °C for the period of 7200 sec. Once cooked, the unbleached durian peel pulp was washed, screened and centrifuged.

Preparation of Magnetic Composite Sheet using Lumen Loading Method. The magnetic composite sheets were produced through the lumen loading method. 15 g dry weight of unbleached durian shell pulp was disintegrated in 1250 ml of distilled water containing 0.1 g L⁻¹ aluminium sulphate for 1800 sec in a mechanical stirrer at the rotor speed of 1000 rpm. Magnetic pigment, iron (II, III) oxide Aldrich 637106 nanopowder with particle size less than 50 nm at the weight of 30 g was mixed in 250 ml distilled water which contained 0.1 g L⁻¹ aluminium sulphate. Next, both suspensions were mixed and mechanically stirred for 1800-3600 sec at the rotor speed of 1000 rpm. This stage is called as impregnation stage, where particles are forced to embed in the lumen fibers. Later, the suspension was slowly stirred to 400 rpm for 14,400 sec when polyethyleneimine (PEI, 2 %, w/w polymer on pulp) was added in the mixture. PEI was added as retention aid to retain the magnetic particles inside the lumen. The excess pigment that remained on surface of fiber and suspension were removed by washing with tap water in a self-designed fiber classifier containing a filter screen (45 µm) for period range of 1800-3600 sec. The clean pulp then proceeded to paper making process [20].

Fabrication of Superhydrophobic Surface of Composite Sheet. 40 g of PCC (Albacar® HO) and 1.238 g of SA (Peter Greven Asia Sdn. Bhd.) were added into 90 ml of distilled water. Next, the suspension were stirred at 75 °C for 1800 sec to allow fatty acid to form a thin layer of water insoluble calcium salt on the PCC surface. The suspension was then mixed with 9.16 g of polymer latex (Acronal NX 4787 X) after cooling down to room temperature and was stirred again for 1800 sec with agitator. Next, a roller was used for applying the coating suspension onto the surface of the durian magnetic composite sheet. The coated paper was air-dried for 3600 sec before immersing into a container of 0.0015 mol/L potassium stearate (PS) suspension for 180 sec. The immersion
process in the PS suspension is known as treatment stage to further improve the water resistance of the coated paper. The coated paper was then rinsed with distilled water at 75 °C to remove the excess stearate salt present on the paper surface. The coated paper was air-dried again for another 3600 sec prior to paper testing [2].

**Characterization of Coated Magnetic Composite Sheet.** The Carl Zeiss Model 1450VP variable pressure scanning electron microscope (SEM) was used in characterizing the morphology of the composite sheet surface. The water contact angle (WCA) of the samples was examined using FECA Contact-Angle Meter. The mechanical properties; tensile strength and tear strength were measured using a Büchel-Van Der Korput horizontal tensile tester Elmendorf tear tester, respectively. The test was in accordance to TAPPI standard method T 949 om-01 and TAPPI Standard T414 om-88.

**Result and Discussion**

**Surface Morphology of Coated Magnetic Composite Sheet.** This method involved several dipping process stages in order to form a superhydrophobic surface. The coating formulation in the technique applied in this study contains both fatty acid modified PCC and polymer binder. Since the acid group in a stearic acid molecule forms an insoluble calcium salt and leaves a hydrophobic tail oriented to the air, therefore the fatty acid modified PCC is hydrophobic. The main function of stearic acid (SA) in coating formulation is to reduce high surface energy and decrease the agglomeration of particles such as precipitated calcium carbonate (PCC).

At the same time, PCC is widely used as fillers in this method that act as adhesives and sealants when react with SA. By knowing the general properties of each material that are used, the formation of the superhydrophobic surface can be performed effectively. To improve both WCA and water resistance, the surface coated paper was treated further by dipping in potassium stearate aqueous solution [21].

The surface morphology of coated durian peel magnetic composite sheet is given in Fig. 1(a) and 1(b). It shows the distribution of PCC particles on the coated composite sheet. The surface roughness can be seen from SEM image and this contributes to the superhydrophobicity properties of the magnetic composite sheet [7].

![Fig. 1 SEM images of surface coating using stearic acid at (a) low magnification, and (b) high magnification.](image)

**Water Contact Angle of Magnetic Composite Sheet.** The water contact angle measurement is one of the important analyses in measuring superhydrophobic properties. Generally, material surface is classified as hydrophilic if the contact angle is less than 90°. When the contact angle higher than 90°, the surface is called hydrophobic, and when the contact angle is greater than 150°, the surface is called superhydrophobic [2].
Fig. 2(a) shows the image of a water droplet on the coated magnetic composite sheet surface with a water contact angle of 154.85°, which indicates that the resulting coated surface achieves superhydrophobicity. As compared to the uncoated magnetic composite sheet in Fig. 2(b), no water contact angle was produced. The untreated magnetic composite sheet has zero water contact angle due to the hydrophilic properties of natural fiber [18].

![Fig. 2(a) Image of a water droplet that formed WCA of 154.85° on the coated magnetic composite sheet surface, (b) Image of water droplet being fully absorbed onto the surface of uncoated or untreated magnetic composite sheet.](image)

**Summary**

A superhydrophobic surface of magnetic composite sheet with a water contact angle 154.85° was successfully fabricated by simple coating method in PCC, stearic acid and polymer latex. Dipping the coated composite sheet into potassium stearate (PS) solution resulted in sheet with a higher resistance to water penetration.

**Acknowledgements**

Authors would like to thank Universiti Teknikal Malaysia Melaka (UTeM) and Ministry of Higher Education, Malaysia for supporting this research under Fundamental Research Grant Scheme (FRGS): FRGS/2013/FKP/TK06/02/2/F00157. The Faculty of Manufacturing Engineering, Universiti Teknikal Malaysia Melaka is gratefully acknowledged for providing the facilities.

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