WATER-REPELLENT IMPROVEMENT OF GREEN COMPOSITE SHEET SURFACE BY HYDROPHOBIC MODIFIED-SILICA COATING

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ABSTRACT: The phenomenon of hydrophobic surface (contact angle of a water droplet exceeding 90°) has attracted a considerable research interest from academia and industry. Current studies have unveiled the fact that the hydrophobicity of a solid surface is governed by surface free energy and surface roughness. To date, many methods have been proposed for fabricating hydrophobic surfaces. In this paper, a facile, low cost, and time-saving approach for the improvement of water-repellent property of durian peel composite sheet surface is studied. A unique mixture of hydrophobic modified-silica particles and polystyrene was synthesised and applied onto the composite sheet via dip coating method. The hydrophobic property was characterised using scanning electron microscopy (SEM) and water contact angle meter. Results show that a water repellent surface with a contact angle of 143.90° was generated, which is nearly superhydrophobic. This method could be an effective strategy for producing hydrophobic surfaces for promising potential applications in water repellency, self-cleaning, friction reduction, and antifouling.

KEYWORDS: *Hydrophobic Surface; Modified-Silica; Polystyrene; Durian Peel; Composite Sheet*

1.0 INTRODUCTION

Inspired by the natural superhydrophobicity of lotus leaves, the generation of water-repellent surface have attracted much attention from academic researchers and industrial players [1–4]. It is renowned that hydrophobicity can be accomplished by the construction of high surface roughness on the material and low surface energy coating [5–10]. The wetting properties of a solid surface is controlled by the geometric structure and chemical composition of the solid surface [11-12].

Over recent years, thousands of reports have been published regarding the exploration and application on superhydrophobic surface including lubricity [13-14], self-cleaning [15-16], water impermeable textiles [17-18], and antifouling [19-20]. Various techniques have been applied in producing hydrophobic and superhydrophobic surface that mimic the behaviour and chemistry of the lotus leaves, such as sol-gel process [10,21], phase separation [22-23], chemical vapour deposition [11,24], layer-by-layer deposition [25], and dip coating [12,26]. Wu et al. [27] fabricated superhydrophobic textiles and sponges in fluoropolymers by dip coating method, while Mahadik et al. [28] reported that transparent superhyrophobic silica films can be invented on glass substrates by sol-gel procedure. Different approach was taken by Wang et al. [29], where hydrophobic polymer surface was produced through nickel electroless deposition. Meanwhile, Pan et al. [30] invented a superhydrophobic steel surface using poly (methyl methacrylate) (PMMA) and hydrophobic silica nanoparticles (SNs) mixtures through spraying technique. The research was carried out for anti-icing and anticorrosion application.

This paper investigates an extremely simple, low cost, and time-saving technique for making the hydrophobic surface of composite sheet from durian peel by dip coating the mixture solution comprising polystyrene (PS) emulsion and modified silica particles. Even though many studies have been conducted on hydrophobic surface, to the best of our consciousness, there are just a few papers discussing the production of hydrophobic surface on cellulosic materials.

2.0 METHODOLOGY

2.1 Materials

Durian (*Durio zibethinus* Murray) peels were obtained from a local fruit stall in Melaka, Malaysia. Sodium hydroxide pellet (NaOH), tetraethyl orthosilicate (TEOS, 98.0%), ethanol (99.7%), ammonia (28.0%), polystyrene (PS, MW = 100,000), and tetrahydrofuran (THF) were purchased from Polyscientific Enterprise Sdn. Bhd. Octadecyltrichlorosilane (OTS) used for surface hydrophobic modification was purchased from Terra Techno Engineering. Deionised water was used in all experiments. All chemicals were used as received without further purification.

2.2 Preparation of Sample

The surface of fresh durian peels were cleansed and washed with water to take out dirt and unnecessary materials. After dried, the durian peels were chopped to about 2-3 cm in length and 0.3-0.5 cm in thickness. Then, the durian chips were dried in a drying oven at 50 °C for 24 h to get rid of the moisture content [31].

2.3 Pulping Process

After the drying process, about 300 g of the dried durian chips were put in a rotary digester together with NaOH and water for the process of transforming the chips into pulp. The process eliminates the lignin from cellulose. It was carried out at the cooking temperature of 170 °C with 17% of active alkali for 7,200 sec. The ratio of cooking liquor to raw materials was 7:1. After cooked, the unbleached durian peel pulp was washed, screened, and centrifuged.

2.4 Preparation of Silica Particles

Ten millilitres of ammonia, 10 mL of tetraethyl orthosilicate (TEOS), and 0.6 mL of octadecyltrichlorosilane (OTS) were added into 100 mL of absolute ethanol. Then, the mixture was magnetically stirred for 1 h and left for 12 h at room temperature. Lastly, the modified silica particles were collected at 60 °C by drying the system until the solvent was completely evaporated [32].

2.5 Preparation of Hydrophobic Composite Sheet Surface

Approximately 0.2 g of polystyrene (PS) was dissolved in 10 mL of tetrahydrofuran (THF). Next, 0.4 g of modified silica particles were added directly when the polymer completely dissolved and the mixture was dispersed ultrasonically at room temperature for 5 min. After that, the durian peel composite sheet was immersed inside the solution mixture and it was dried at room temperature until the solvent was completely evaporated [32].

2.6 Characterisation

The coated durian shell composite sheets underwent two analyses which were surface morphology and water contact angle. Carl Zeiss Model EVO 50 VP of variable pressure scanning electron microscope (SEM) was used to observe the treated samples' morphology. Water absorption for the samples was examined by the water contact angle (WCA) measurement where 5 μL of deionised water was dropped onto the composite sheet surface at ambient temperature using FECA Contact-Angle Meter.

3.0 RESULTS AND DISCUSSION

3.1 Surface Morphology

This method used silica particles as a mineral pigment in wet-filling, surface filling, and surface coating applications. In this method, the durian peel composite sheet was immersed into the coating formulation of silica particles. The surface morphology of the green composite sheet is shown in Figure 1. It was examined using SEM at 50x and 300x of magnifications. The images show the dispersal of modified silica particles on the composite sheet surface.

Figure 1(b) shows the accumulation of bits of spheres that gathered together having micron or submicron binary structure with the diameter ranging from 0.5 to 20 μm. Water-repellent property was enhanced due to this aggregation of micron or submicron binary structure that increases the roughness of the composite sheet surface. Surface roughness is one of the factors to produce high hydrophobic property [7].

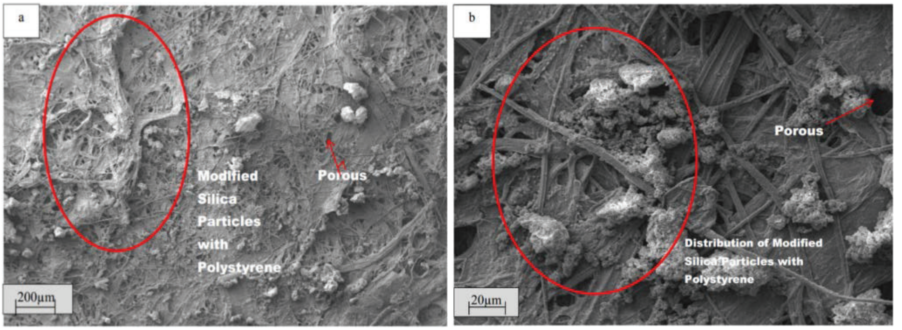

Figure 1: SEM images of modified silica particles on durian peel composite sheet surface at (a) 50x magnification and (b) 300x magnification

3.2 Water Contact Angle

The property of wettability of a solid surface can be determined by measuring the contact angle of liquid on the surface. Figure 2 shows the photograph of a water droplet on the durian peel composite sheet coated with silica/polystyrene composite coating. Understandably, durian peel composite sheet is a hydrophilic material since it was constructed from the natural fibre. However, after the deposition of modified silica particles into polystyrene matrix, the composite coatings on durian peel composite sheet surface exhibited hydrophobicity with the contact angle of 143.90°.

The surface microstructure of silica/polystyrene composite coating was very rough with numerous interspaces or cavities and the air can be easily trapped in these pores. Although the angle did not achieve superhydrophobic angle, but it was highly hydrophobic, which is more than 90° and almost close to 150° [6].

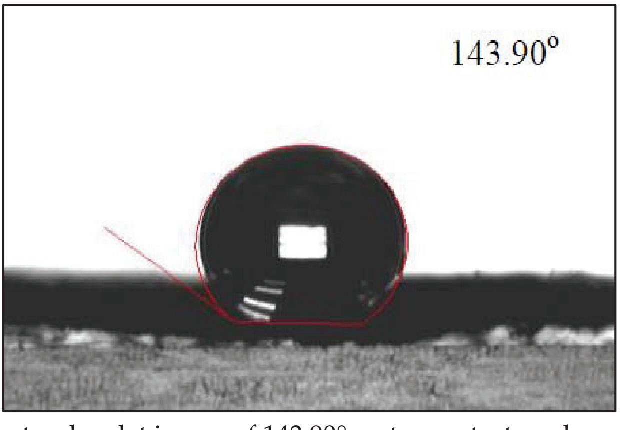

Figure 2: A water droplet image of 143.90° water contact angle on the coated durian peel composite sheet surface

3.3 Effect of Silica Particles and Polystyrene Emulsion

In this method, the silica particles were synthesised using Stober method with tetraethyl orthosilicate (TEOS) and polystyrene (PS). In basic condition, TEOS acts as a silica precursor and undergoes hydrolysis and condensation in the modification stage. The physical blending between hydrophilic inorganic particles such as silica particles and hydrophobic polymers such as polystyrene may accelerate to phase separation or agglomeration of the particles [32]. Thus, it will help in the formation of hydrophobic condition.

Silica particles are one of the most widespread materials being used due to its easy and simple preparation through hydrolysis-condensation reaction of precursor molecules such as TEOS [33]. The surface of modified silica particles produced lower water absorption since it has water contact angle of 143.90° as shown in Figure 2. This occurred due to the modification that made the surface more hydrophobic.

Silica particles have hydroxyl groups on their surface that makes its surface becomes hydrophilic. After modification, the polymer chain in PS replaced the hydroxyl groups on silica particles, producing a more hydrophobic surface [32].

3.4 Effect of Polystyrene Particles

Polystyrene (PS) is a vinyl polymer that has been produced by polymerisation of the monomer styrene. Fundamentally, PS has a good waterproof property and low density which make it suitable to be used in making hydrophobic surface of green composite sheet. These properties will ease the process of making hydrophobic surface through the combination of high surface roughness of modified silica particles and low surface energy of PS.

4.0 CONCLUSION

In conclusion, hydrophobic coating was successfully achieved through the synthesis of modified silica and polystyrene solution with water contact angle of 143.90°. The formulation is also stable and suits to be applied onto other cellulose or wood surfaces. The results proved that the combination effect between the low surface energy and submicron binary structure changed the surface characteristic of durian peel composite sheet from hydrophilic to hydrophobic.

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