Synthesis of Silica from Rice Husk Waste for Hydrophobic Material as an Anti-Water Coating for Eyeglasses

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Abstract: Hydrophobic materials made of silica as a coating for glasses are very attractive. The synthesis of silica (SiO_2) from rice husk ash was successfully carried out using the sol-gel method. The hydrophobic silica composition is made with four variations of silica, consisting of 0.5, 1.0, 1.5, 2.0 g and polydimethylsiloxane (PDMS) keep constant in 20 g. The raw material was first characterized by X-ray Fluorescence (XRF) to investigate element composition. Then, silica powder obtained from rice husk ash was characterized by Fourier Transform Infrared Spectroscopy (FTIR), Particle Size Analyzer (PSA) and Water Contac Angel (WCA) were employed to investigate functional groups material, size of particles and hydrophobicity of the prepared samples for coating glass. The result of contact angel from coating glass exhibited more than 90° and the maximum hydrophobicity properties of polydimethylsiloxane/silica (PDMS/SiO_2) was about 111.40° for the silica composition of 2 g.

Keywords: Hydrophobic, Polydimethylsiloxane, Rice Husk Ash, Silica

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I. INTRODUCTION

The basic surface properties of glass materials are hydrophilic, making it easy for rainwater and dew to stick to the surface. This causes problems for glasses because the sticking water causes fungi to grow on the surface, frosting, sticking dirt, dust, and so on. This reduces the transparency of the glass over time and does not function optimally. Therefore, the ability to resist water (waterproof) on the surface of the glass is crucial and considered, especially in maintaining function and increasing resistance in glasses. One way that is currently possible is to coat using hydrophobic materials.

The advantage of this hydrophobic property is that it is waterproof, always looks clean, reduces fluid on the surface, reduces fluid friction with the surface, and is non-polar property. When applied to glass, it will have a self-cleaning feature [1]. From several studies that have been carried out, one of the most abundant materials in Indonesia is rice husk. Rice husk is a hard layer that covers the kernel, consisting of interlocking lemma and palea, commonly found in rice milling areas. The husk is typically obtained at 20±30%, bran at 8±12%, and milled rice at 50±64% of the initial weight of paddy during the milling process. In addition, the husk has a bulk density of 125 kg/m^3, with a caloric value of 1 kg of rice husk at 3300 kilocalories [2]. In 2018, the rice harvest was around 6.05 million tons of rice with a total of approximately 1.27 million tons of rice husk. Based on these results, the potential for the utilization of rice husks is very large. In terms of economic aspects, the price of rice husk is not as expensive as other materials such as TiO_2, a material for ceramic nano-coating which is more expensive. Silicon dioxide or silica from rice husk has some advantages so it is still comparable with some of its competitors. If the correct washing is carried out, the silica content can reach up to 99% [3]. One of the criteria for good hydrophobic material is a high-water contact angle with its surface (>90°). The contact angle obtained using silica (SiO_2) derived from RHA is 90° to 142.5°, approaching super hydrophobic material [4]. Then, from the experiment carried out by Huang et al., hydrophobics derived from silica have extraordinary properties in the coating of optical devices. In his experiment, it was found that the contact angle obtained using this hydrophobic coating reached 126°. In addition, this coating has very good mechanical properties and a refractive index produced of 1.22-1.42. The transmitted light is also relatively high at 99.30% [5]. Moreover, as a research comparison, hydrophobic coating materials have been carried out using silica from silica sand resulting in a contact angle of 98° [6]. Also according to Priyadi and Wahyuni's research, PDMS or polydimethylsiloxane is a polymer that has hydrophobic properties due to its low surface energy and is commonly used as a coating for a material to increase the hydrophobicity. Additionally, PDMS has a transparent appearance, making it suitable for use as a coating to strengthen the hydrophobic properties of eyeglasses [7]. Based on previous research, this research was carried out to determine the process of synthesis, characterization, and hydrophobic properties of materials derived from natural materials such as rice husk waste and their appli-

TABLE I: XRF element content of the rice husk ash.

Element	Si	K	Ca	Fe	Ti	Mn	Cu	Zn	Sr	Re
wt.%	74.3	9.74	8.01	6.47	0.091	0.921	0.22	0.091	0.063	0.1

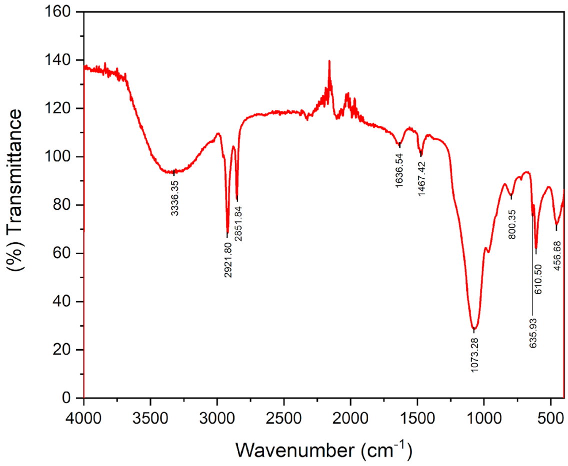

FIG. 1: FTIR spectrum of SiO_2 power from rice husk ash.

cation as a hydrophobic coating in anti-water eyeglasses.

II. METHOD

A. Materials

Rice husk, distilled water, hydrochloric acid (HCl) 37%, sulfuric acid (H2SO4), acetone, sodium hydroxide (NaOH), cetyltrimethylammonium Bromide (CTAB-C19H42BrN), butanol (C4H9OH), polydimethylsiloxane (PDMS), object glass, and digital camera.

B. Experimental Procedure

500 g of rice husk was washed with distilled water. After that, it was sun-dried to evaporate the water content. The rice husk was heated to a temperature of 700°C for 4 hours to obtain RHA using a furnace. The RHA was then purified using a leaching process in a mixture of 10% HCl and 30% H2SO4 in a beaker. The acid leaching sample was stirred and heated to a temperature of 100°C using a hotplate stirrer. The remaining ash was filtered and washed with distilled water until the pH was 7. Then, 3.5 M sodium hydroxide (NaOH) solution was added and heated for 6 hours at a temperature of 100°C. The sample was precipitated for 24 hours to obtain Na2SiO3. Next, 2 wt% cetyltrimethylammonium bromide (CTAB) was dissolved in 50 ml distilled water and 60 ml butanol. The CTAB solution was mixed with sodium silicate with heating at a temperature of 60°C on a hot plate. Then, 0.5 mol/L H2SO4 solution was added gradually until the acidity degree pH value was 4. The SiO_2 gel mixture was heated to a temperature of 100°C until it changed into solid SiO_2 and mashed using a mortar and pestle. Hydrophobic material was made with a mixture of SiO_2 (0.5, 1.0, 1.5, 2.0 g) and PDMS 20 g mixed until homogeneous. Finally, the hydrophobic solution sampel was coated on a glass slide as a media using a fine brush until a glass slide fully thin coated with the hydrophobic solution.

FIG. 2: Product SiO_2 powder from rice husk ash.

C. Characterization

X-ray Fluorescence (XRF) with the type of device is PANalytical type Mini Pal 4 was used to determine the purity of the RHA that has been furnaced. Functional group analysis was determined by Fourier Transform Infrared Spectroscopy (FTIR) with the type of device Shimadzu FTIR-8400S FTIR. The wave used in the analysis using FTIR were in the infrared spectrum with wavenumbers around (400 - 4000) cm^{-1}. The average particle size of the sample was measured using a Particle Size Analyzer (PSA) with the type of device Zetasizer Ultra. The surface condition of the sample coated on a glass slide was viewed using a microscope camera with 1600× magnification to determine its coating morphology, and also measure its water contact angle (WCA) to determine hydrophobicity.

III. RESULTS AND DISCUSSION

A. SiO_2 Analysis based on Rice Husk Ash (RHA)

The element analysis of RHA was carried out using XRF testing, as presented in Table I. The content of Si element, a result of raw material RHA, has relatively high purity obtained at 74.3 wt.%. According to previous research reported range of containing SiO_2 from RHA is between 72.1 to 98.61 wt.%, so this result is good enough because the raw material RHA

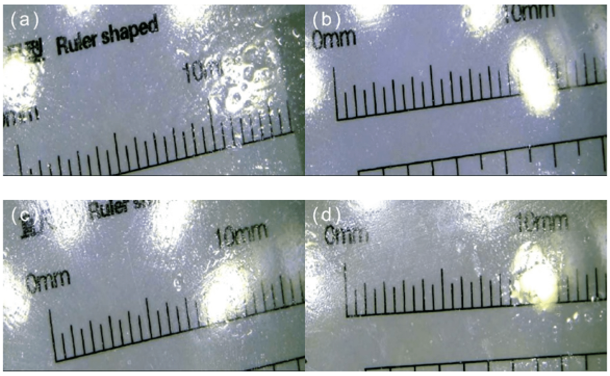

FIG. 3: Glass sample coated by modified SiO_2/PDMS with various SiO_2 (a) 0.5 g, (b) 1 g, (c) 1.5 g, (d) 2 g.

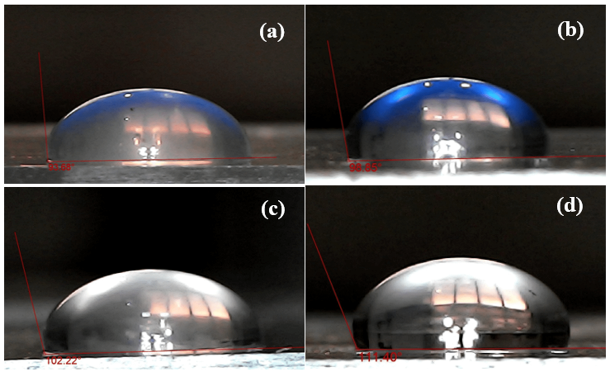

FIG. 4: Water contact angle of glass sample with coating PDMS/SiO_2 (a) 0.5 g (b) 1 g (c) 1.5 g (d) 2 g.

has not been treated to purify [7]. In addition, Linda et al. reported that used sand silica obtained containing element Si of 81.7 wt.%, where the result is nearly comparable [6].

The chemical bonding of SiO_2 synthesized from RHA was carried out using Fourier transform infrared spectroscopy (FTIR) characterization. This chemical bonding has specific characteristics in infrared absorption or transmission, which are identified using absorption or transmission bands. Absorption bands are described in a spectrum that shows the wave value of a specific group and proves the existence of chemical bonds in a particular substance. To clarify FTIR spectrum of the SiO_2 powder sample synthesized from RHA is depicted in Fig. 1. The length of the wavenumber used a range of (400 - 4000) cm^{-1}. Several transmission peaks were detected at wavenumbers 1073.28, 800.35, and 456.68 cm^{-1} that were identified as asymmetric Si-O-Si stretching, symmetric Si-O bending (silanol) and Si-O rocking bond. Similar results have been reported by several researchers who identified functional groups in the range of wave numbers 1064 to 1100 cm^{-1} (asymmetric Si-O-Si stretching), 790 to 800 cm^{-1} (symmetric Si-O bending), and 443 to 466 cm^{-1} (Si-O rocking bond) [9-10]. The bending vibration of the O-H group causes a peak to appear at wavenumber 3336.35 cm^{-1} [9]. Moreover, there are two peaks indicating the asymmetric and symmetric stretching vibrations of -CH2- group function at wavenumber 2921.60, and 2651.84 cm^{-1}. Two similar peaks were reported by Bonakdar et al. at wave numbers 2917 and 2847 cm^{-1}, where the results are close to our result research [11]. Therefore, CTAB functions as a surfactant which is clearly detected in the resulting SiO_2 powder. Based on our research results, it was concluded that SiO_2 from RHA extract by leaching HCl with H2SO4 added CTAB was confirmed to have the same peak as previous studies.

B. Hydrophobic Coating Result

The SiO_2 powder has been obtained from RHA with a sol-gel method that has the characteristics of the above and will be used to filler in modified SiO_2 material. To clarify SiO_2 powder product was depicted in Fig. 2. The modified SiO_2 was carried out by incorporating PDMS compounds as the matrix. Four samples were successfully prepared with SiO_2 mass variations, including 0.5, 1.0, 1.5, and 2.0 g with a constant PDMS mass of 20 g. This research used object-glass as a coating medium for modified SiO_2 samples. A glass sample has been coated with a modified SiO_2 (PDMS/SiO_2) coating depicted in Fig. 3.

The wettability of surface material is a parameter associated with the contact angle, which is the angle between a water droplet and the liquid-gas or solid-liquid interface in the solid, liquid, and gas equilibrium states. Wettability performance heavily depends on surface structure (surface free energy, roughness) and chemistry. The theoretical framework includes four wetting models: Young's model, Wenzel's model, Cassie-Baxter's model, and the Partial Wetting Model [12]. These models greatly assist researchers in analyzing the determination of material hydrophobicity. To evaluate hydrophobicity, water contact angle measurements are conducted to obtain the contact angle of the material surface. These measurements are used to evaluate whether a material is hydrophobic or hydrophilic. Contact angles formed smaller than 90° from the material surface is called hydrophilic, while contact angles formed greater than 90° from the material surface is called hydrophobic [13]. Additionally, based on their hydrophobic properties, it can be categorized into three types based on their contact angles: hydrophobic with a contact angle greater than 90°, overhydrophobic with a contact angle greater than 120°, and superhydrophobic with a contact angle greater than 150°, where is measured from the material surface [14]. The wetting test process is conducted by placing distilled water into the surface of a glass sample coated with a PDMS/SiO_2 layer with varying SiO_2 masses. Subsequently, images of the water droplets on the glass sample are captured using a microscope camera with 1600× magnification. The captured images are then analyzed using Image-J software to measure the contact angle of the material's surface. To further clarify the measurement of the glass sample, the resulting images are depicted in Fig. 4.

The PDMS/SiO_2 coating is a non-polar material. Generally, PDMS is a polymer material with low surface energy and hydrophobic properties [7]. Additionally, in this research, PDMS has functioned as a matrix to strengthen and attach SiO_2 to the glass sample. On the other hand, the variation of SiO_2 mass in this research is expected to increase the surface roughness of the glass sample. The two crucial points in creating a hydrophobic surface are that the surface are low free energy and high roughness [12]. Both parameters significantly affect the hydrophobicity of the surface. The same results are observed in this research, as the contact angle measurements show an increase in contact angle with increasing SiO_2 mass. The contact angle values on the glass samples coated with PDMS/SiO_2 are shown in Table II. Other research has reported that the addition of SiO_2 content to modified SiO_2 material for indium tin oxide (ITO) resulted in changes in the formed contact angle due to surface roughness and low free energy [6]. Therefore, it is evident that the use of PDMS and SiO_2 plays a role in the hydrophobicity of the material in this research.

TABLE II: Water contact angle of a silica coating on glasses.

SiO_2 (g)	PDMS (g)	Contact Angle (°)
0.5		93.68
1.0		98.85
1.5	20	102.22
2.0		111.40

In this research, the highest contact angle value was obtained for the SiO_2 variation of 2 g, which was 111.40°, while the lowest contact angle occurred for the SiO_2 variation of 0.5 g was 93.68°. Overall, the addition of SiO_2 mass increased the contact angle. It is believed that the homogenous particle size of SiO_2 (5 μm) due to the addition of the CTAB compound as an additive to homogenize the particle distribution influenced the formed contact angle. Other research has reported suboptimal contact angle results due to non-homogeneous coating processes and particle sizes (without the addition of a surfactant) [6]. The glass substrate used in this research is polar and has a smooth surface, so when the glass surface interacts with water (which is polar), it results in a hydrophilic interaction. On the other hand, in this research, the glass substrate was coated with PDMS/SiO_2 to modify the smooth surface structure of the glass and create a rough surface with low surface free energy, resulting in a hydrophobic interaction between water and the glass surface [12]. Therefore, the surface roughness and surface free energy of the glass sample coated with PDMS/SiO_2 were considered in this research, based on the selection of materials, composition, and SiO_2 synthesis method.

IV. CONCLUSION

Silica obtained from rice husk ash using the sol-gel method produces a powder that is used as a hydrophobic material. This silica powder has been confirmed by FTIR characterization with three primary peaks at wavenumber 1073.28, 800.35, and 456.68 cm^{-1}. The results of the particle size analyzer (PSA) test showed that SiO_2 with acid leaching HCl+H2SO4 has an average particle size of 5 µm. The water contact angle (WCA) test obtained the best results for the PDMS/SiO_2 (2.0:20 g) composition of 111.40°. The content of silica in PDMS affects the value of the formed contact angle, which becomes larger. Thus, the hydrophobic material made from rice husk ash has the potential to be used as an anti-water coating on eyeglasses.

Acknowledgment

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