A green approach of superhydrophobic surface fabrication on recycled high-density polyethylene using sodium chloride

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Abstract. In this work, the water-dissolved surface modifier method was introduced to recycled high-density polyethylene (rHDPE) matrix to fabricate green superhydrophobic surfaces. Surface cavities on rHDPE are formed by sodium chloride particles which can be readily rinsed off and reused. Water contact angle, self-cleaning properties, and surface morphology were characterized. By creating porosity onto the rHDPE matrix, the surface exhibits an excellent self-cleaning property with a water contact angle larger than 150°. Surface morphology reveals the porosity and roughness of the surface. In this fabricating process, no chemicals are used while rHDPE is selected for the purpose. Based on the findings, it is proven that the superhydrophobic surface can be fabricated with a simple yet green approach.

1 Introduction

Surfaces with a low sliding angle of less than 10° and a high contact angle of more than 150° with water derived from the unique structure of the surface and its chemical composition have acquired a high interest in the industry due to their different applications [1-2]. These surface forms are called superhydrophobic surfaces. Numerous superhydrophobic surfaces exist, such as the traditional example of lotus leaves, butterfly wings, and natural leaves [2]. Since the water droplets falling on the surfaces can roll off easily without the assistance of

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any tools used for cleaning, the characteristics of these surfaces are critical for self-cleaning [3].

In general, super-hydrophobic surfaces are prepared in two steps. The method begins with the development of a rough surface and the treatment of surfaces with other substances capable of producing a molecule with low surface energy or treating it with materials able to produce superhydrophobic elements [4-5]. The manufacturing approach has been made simple and optimized with the introduction of the recent mechanism for research studies and the basic principle [6]. During several types of research that have been developed, the environmentally friendly manufacturing process has been accomplished. A better efficiency, such as mechanical stability and superhydrophobic properties is also shown by the prepared manufacturing process. The achievement of superhydrophobic requires, according to the classical theory of surface wettability, to fulfill many aspects of the necessity of rough structure and low surface energy that can restrict the reaction between the structure superhydrophobic surface and surface water [7].

This paper focuses on the fabrication of superhydrophobic surfaces on recycled high-density polyethylene using sodium chloride. The water contact angle, self-cleaning properties and surface morphology will be analyzed and discussed.

2 Experimental Method

2.1 Materials

The recycled high-density polyethylenes (rHDPE) were supplied by Lotte Chemical Titan (M) Sdn. Bhd. is used as the matrix meanwhile fine sodium chloride (NaCl) was purchased from Adabi Consumer Industries Sdn. Bhd. is used as a surface modifier.

2.2 Methods

The NaCl particles were hand-ground for 30 minutes, 60 minutes, 90 minutes and 120 minutes, respectively. In the compression moulding process, the hot press was used for 5 samples with the Technopress 50HC-β model of the compression moulding machine. The empty mold is preheated for 5 minutes under the temperature of 180 °C. Then, the weighted sample of rHDPE was placed into the mould. The mold and the samples were hot-pressed with a temperature of 180 °C in 10 minutes.

For the compression of the rHDPE sheet and the filler which is sodium chloride, the mold is preheated for 5 minutes with a temperature of 180 °C. Then, the weighted sodium chloride which is 50% of the weight of rHDPE sheet has been placed inside the mold while the rHDPE sheet was placed on top of the salt. After that, rHDPE sheet and the filler were hot-pressed for 10 minutes under the temperature of 180 °C in 10 minutes. Lastly, the mold is cooled for 5 minutes to allowing it to solidify before being taken out as a finished sample.
Fig 1. (a) rHDPE is pressed under a pressure of 20 N/cm³, (b) placing of rHDPE and sodium chloride before compression, (c) molding with heat and pressure

2.2.1 Preparation of superhydrophobic sample

In preparing the superhydrophobic sample, a sheet of rHDPE that has been pressed with sodium chloride was taken into ultrasonic cleaning with a frequency of 53 Hz for 30 minutes. The purpose of ultrasonic cleaning is to leach the sodium chloride from the rHDPE sheet.

![Fig 1](image)

Fig 2. Schematic of sample in the ultrasonic cleaning

2.3 Characterization and Testing

2.3.1 Element and microstructural analysis

Energy Dispersive X-Ray analysis has been used to analyze the composition available on the sample after the leaching process. For microstructural analysis, scanning electron microscopy was used to study the surface morphology of the sample. A thin layer of platinum coating has been used to create a conductive layer on the sample’s surface which can exhibit the charge. 1000x magnifications were used on the SEM with the accelerating voltage of 20 kV. The morphology of the sample has been discussed.

2.3.2 Contact angle measurement

The contact angle analysis has been done by dropping a drop of water onto the sample surface. An image has been taken using the camera and ImageJ software has been used to
measure the contact angle of water and the surface sample. The contact angle measurement was done at least 8 times for each sample.

2.3.3 Self-cleaning ability

Charcoal powder was used as a contaminant to demonstrate the self-cleaning ability after the fabrication with salt. A layer of charcoal powder was spread on the surface and inclined with a continuous drop of water was poured onto the charcoal powder spread on the surface.

3 Results and discussion

3.1 Element and microstructural analysis

Figure 1 (a) and (b) shows the mapping of elements found on the surface of the rHDPE sample after the filler has been leach from the sample using Energy Dispersive X-Ray analysis (EDX). The percentage is shown in Figure 1 (c) indicates that the sample element which is Na and Cl found on the sample is less than 1% which is also negligible to state that the element of the sample has been successfully removed from the sample surface.

Fig 3. a) SEM micrograph under 5000x magnification b) Mapping of element available on sample and c) mass percent of element found on the sample surface
Micrographs were taken under magnification of 1000x as shown in Figure 2. This figure shows that samples with filler of 120 minutes hand-ground contained high porous features at the surface when compared with filler of 30 minutes hand-ground. This indicates that the higher the porous feature available on the surface of the material, the higher the superhydrophobicity of the sample.

![SEM micrographs](image)

**Fig 4.** SEM micrograph under 1000x magnification of a) 30 minutes, b) 60 minutes, c) 90 minutes and d) 120 minutes

The reason why the porous feature in 120 minutes sample is the highest when compared with other samples is because the filler is the finest among others. The process in the making of the sample surface which is the hot press of the sample with the filler gives the sample this characteristic which depends on the particle size of the filler. The finer the filler size can give the sample with higher porosity characteristics.

The fine size of filler can also help to produce a fine size of pores on the sample’s surface allowing air to trap on the surface. Thus, the superhydrophobic characteristics of the sample are high. Therefore, the finer filler size could have a better result when compared with the course size of the filler as finer filler size can enhance the superhydrophobicity of the sample with the contact of water.

### 3.2 Contact angle measurement

Figure 3 shows the measured water contact angle between the water and the sample's surface. Some of the samples have high water repellency, with superhydrophobic properties of over 150°. Sample with finer filler size tends to reach the characteristics of the superhydrophobic surface compared with sample with coarser filler size.
3.3 Self-cleaning ability

The solution that has been used in the self-cleaning test is methylene blue. Methylene blue can impart a blue colour to the surface of the sample that can be resulting in a clear sign of residue available on the sample. Other than that, a mixture of 3.5% NaCl solution also has been used to mimic the condition when seawater has passed through the surface. In this research, the methylene blue and the NaCl solution have shown a similar behavior as the normal water which is can separate the charcoal powder from the surface of the recycled HDPE and the solution also flow past the surface of the sample. Figure 4 a), shows the behavior of the solution on the surface.
The results of the self-cleaning tests with distilled water, methylene blue, and 3.5% NaCl solution all showed that the surface of recycled HDPE managed to keep its superhydrophobic features, or self-cleaning properties as shown in Figure 4 b).

4 Conclusion

In this study, rHDPE sheet was successfully fabricated by using NaCl as the filler to create a rough surface for the superhydrophobic surface. The NaCl has been ground to a smaller size by grinding within 30, 60, 90 and 120 minutes, respectively. The water contact angle, self-cleaning properties and surface morphology show that 120 minutes of hand-ground NaCl particles have the optimum performances and can be a green approach in fabricating superhydrophobic surfaces which brings the self-cleaning ability to the modified rHDPE surface.

References

1. A. Rahim Siddiqui, W. Li, F. Wang, J. Ou, and A. Amirfazli, Appl. Surf. Sci. 542 (2020)
2. Q. Wen and Z. Guo, Chem. Lett. 45, 10 (2016)
3. Y. T. Lin and J. H. Chou, Int. J. Plast. Technol. 20, 1(2016)
4. S. Heinonen, E. Huttunen-Saarivirta, J.-P. Nikkanen, M. Raulio, O. Priha, J. Laakso, E. Storgärds, and E. Levänen, Colloids Surfaces A Physicochem. Eng. Asp. 453, 1 (2014)
5. X. Zhang, M. Järn, J. Peltonen, V. Pore, T. Vuorinen, E. Levänen, and T. Määttylä, J. Eur. Ceram. Soc. 28, 11 (2008)
6. C. Zhang, M. Kalulu, S. Sun, P. Jiang, X. Zhou, Y. Wei, and Y. Jiang, Y., Colloids Surfaces A Physicochem. Eng. Asp. 570 (2019)
7. S. Xu, Q. Wang, N. Wang, and X. Zheng, J. Mater. Sci. 54, 19 (2019)