International Journal of Nanoelectronics and Materials
IJNeaM
ISSN 1985-5761 | E-ISSN 2232-1535

Influence of Sodium Chloride on Recycled HDPE in Superhydrophobic Surface Development

Thor Keat Seng[a], Muhammad Salihin Zakaria[a,b,*], Razif Muhammed Nordin[c,d], Khairul Anwar Abdul Halim[a,b], Nur Farhana Hayazi[e] and Lokman Hakim Ibrahim[f]

[a]*Faculty of Chemical Engineering & Technology, Universiti Malaysia Perlis (UniMAP), Arau 02600, Perlis, Malaysia*
[b]*Biomedical and Nanotechnology Research Group, Center of Excellence Geopolymer and Green Technology (CEGeoGTech), Universiti Malaysia Perlis (UniMAP), Arau 02600, Perlis, Malaysia*
[c]*Department of Chemistry, Faculty of Applied Sciences, Universiti Teknologi MARA, Perlis Branch Arau Campus, 02600 Arau, Perlis, Malaysia*
[d]*Green and Functional Polymer Research Group, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia*
[e]*SIG Surface Technology, Universiti Malaysia Perlis (UniMAP), Arau 02600, Perlis, Malaysia*
[f]*Advanced Polymer Research Group, Center of Excellence Geopolymer and Green Technology (CEGeoGTech), Universiti Malaysia Perlis (UniMAP), Arau 02600, Perlis, Malaysia*
**Corresponding author. Tel.: +604–9798154; fax: +604-9798751; e-mail: salihin@unimap.edu.my*

ABSTRACT

Inspired by lotus leaf, superhydrophobic surfaces have emerged rapidly in recent years. It is due to its excellence ability in self-cleaning and corrosion protection, to name a few. In this research, a green superhydrophobic surface was constructed on the surface of recycled High-Density Polyethylene (rHDPE) by utilizing Sodium chloride (NaCl) as a water-soluble surface modifying agent. The treatment process is simple,and cost-efficient as it only used rHDPE and NaCl as the main materials with additional chemicals and at the same time leave no impact on the environment. Variation of NaCl grinding period influenced the value of CA by which 240 minutes grinded NaCl displayed highest CA of 161.4°. This may be attributed by the increase of surface roughness as the grinding time increase. Furthermore, the surface morphology, topography, and self-cleaning ability of superhydrophobic surafce were analyzed to further understand the surface characteristics. The anti-sticking and self-cleaning properties were successfully displayed by the superhydrophobic surface thus, showing its potential as an eco-friendly material.

Keywords: *Grinding, Sodium chloride, Recycled-HDPE, Surface modifier, Superhydrophobic*

1. INTRODUCTION

The wettability of solid surfaces with liquid is widespread in everyday life, and managing it is crucial to many practical applications. Wettability depends on two factors. One is a chemical composition of the liquid, and the other is geometrical. Both factors, respectively refer to the effect of surface tension of liquid as high surface tension resulted in poor wettability while surface structure, particularly high surface roughness will decrease the adhesion between water and surface. [1,2]. The water droplet Contact Angle (CA) on a surface indicates wettability [3]. A superhydrophobic surface repels water droplets. A superhydrophobic surface may be defined as a surface that demonstrated CA of more than 150° and sliding angle of less than 10°. Good hydrophobicity makes water droplets slide off easily from the surface. [4].

Two requirements are needed for superhydrophobic surfaces, first, rough the surface. Hierarchical micron-to-nanoscale roughness is preferred [5], as the roughness increases the material's surface area, the hydrophobicity will geometrically increase (Wenzel model) [6]. The second criterion is the low surface angle. If these two constraints are satisfied, a water droplet on the surface will be in the Cassie-Baxter wetting state, in which resulting in poor total contact between a liquid and solid and micro- or nanosized gaps occur [5,6].

This study utilizes recycled plastic to generate the superhydrophobic surface instead of raw material to preserve natural resources and energy. Recycled materials have previously been purified and processed. Therefore, second-time production is cleaner and less energy-intensive. Plastic waste is wasteful and creates environmental problems. Thus, initiatives to recycle plastics.

A literature evidence by [7] was one of the early research study that demonstrated the effect of sodium chloride (NaCl) on superhydrophobicity and longetivity of polystyrene. The result revealed the decreased in NaCl concentration which in turn increased the value of CA up to 160±1°. Thus, by considering the research gap in their study enabled this research that focuses on a green approach to develop superhydrophobic surface on recycled High-Density Polyethylene (rHDPE) by employing NaCl as a water-soluble surface modifier that was hand-grinded at variation of time. The CA and self-cleaning properties of the surface as well as surface morphology, and topography will all be assessed.

2. EXPERIMENT

2.1. Materials

The rHDPE pellets were reprocessed HDPE granules that passes through several processes in order to be turned into pellets; from Lotte Chemical Titan (M) Sdn. Bhd. is utilized as the matrix while NaCl from Sigma-Aldrich® is employed as the chemical surface modifier in this research.

2.2. Procreation

The rHDPE pellets were hot pressed using a compression molding machine (GT-7014-H, Go-tech) to produce the 2 mm rHDPE sheet. NaCl particle was hand-grinded manually for 150 minutes, 180 minutes, 210 minutes, and 240 minutes, respectively. The sample of NaCl is equally disseminated over the surface of the rHDPE sheet in the mould after the grinding is fully prepared. Then, before being transferred to the hot press, two sheets of Overhead Projector Film (OHP) film are inserted in between the molds. NaCl particles, which are 50% of the weight of the rHDPE sheet, were evenly distributed throughout the surface of the rHDPE sheet and were hot pressed onto the surface of the rHDPE sheet. The prepared sample was leached away by using an ultrasonic bath with distilled water as a medium. The parameters for hot press and NaCl leaching are reported in Tables 1 and 2.

3. CHARACTERIZATION AND TESTING

3.1. Water Contact Angle

The water CA was evaluated using the sessile drop technique to assess the superhydrophobicity of the NaCl-modified rHDPE surface. The camera captured the angle from the study and the captured images were further evaluated by using ImageJ software. For each sample, the CA was measured ten times in various regions.

3.2. Microstructural Analysis

By using a sputter coater (SPT-20, Coxem Ion Coater), the rHDPE sample was coated with platinum and Scanning Electron Microscopy (SEM) (TJSM-6460LA, JEOL) analysis was conducted to evaluate the microstructure and porosity of the sample's surface after modification. For element composition analysis, the SEM is aided by Energy-dispersive X-ray spectroscopy (EDX) (TJSM-6460LA, JEOL). The element composition of the sample was also determined using EDX.

3.3. Surface Profiling

The topography of the sample's surface is studied using a 3D Nano profiler (Pemtron, Hawk 3D Wt-250). Aside from SEM analysis, surface morphology can be supplied via a 3D nano profiler, which can provide 3D morphology over a much broader area than an atomic force microscope.

3.4. Self-Cleaning Ability

Charcoal powder was selected as a contaminant to exhibit self-cleaning on a modified rHDPE surface. A water dropper was used to apply a methylene blue solution (1 ppm), a 3.5% NaCl solution, and distilled water to the surface full of contaminants. The self-cleaning ability test was observed with and without titling.

Table 1 The parameter of compression molding

Materials	Description	Parameter
rHDPE sheet	Temperature	180°C
	Pressure	15 MPa
	Duration (Preheat-hot press-cooling)	5-10-5 (minutes)
NaCl particles	Temperature	180°C
	Pressure	15 MPa
	Duration (Preheat-hot press-cooling)	5-10-5 (minutes)

Table 2 The parameter of ultrasonic cleaner

Description	Parameter
Frequency	53 Hz
Temperature	60°C
Duration	30 minutes

4. RESULTS AND DISCUSSSION

4.1. Water Contact Angle

Figure 1 displays the water contact for different durations of grinding time of the surface modifier. In Figure 1, when the NaCl grinding time was increased, the shape of the water droplet was perfectly spherical. Figure 1 also illustrates the average water CA of a water droplet from a surface-modified rHDPE surface with varying grinding durations of NaCl particles. According to Figure 1, increasing the grinding time will increase the water CA. Consequently, 240 minutes of hand-grinded NaCl has the highest water CA among the four samples, while 150 minutes has the lowest water CA. Hence, the effect of grinding duration on wetting property correlates well with the predicted results, indicating that increasing the grinding time will increase the water CA [6].

4.2. Scanning Electron Microscopy

Figure 2 illustrates the SEM image of a control rHDPE sheet. Figures 3 (a)-(d) portray the SEM image for the surface of the rHDPE sheet, which had been modified using NaCl with different grinding duration. The surface micro or nanostructure is important in determining wetting properties. Note that microscale hierarchical surfaces with low surface energy generate superhydrophobicity [7]. From Figures 2 and 3, the control sample's micrographs are smooth since it has not been surface-modified. However, the four surface-modified rHDPE sheets revealed micro and nanoscale porosity and protuberances. Thus, all surface-modified samples increased in surface roughness, enhancing superhydrophobicity.

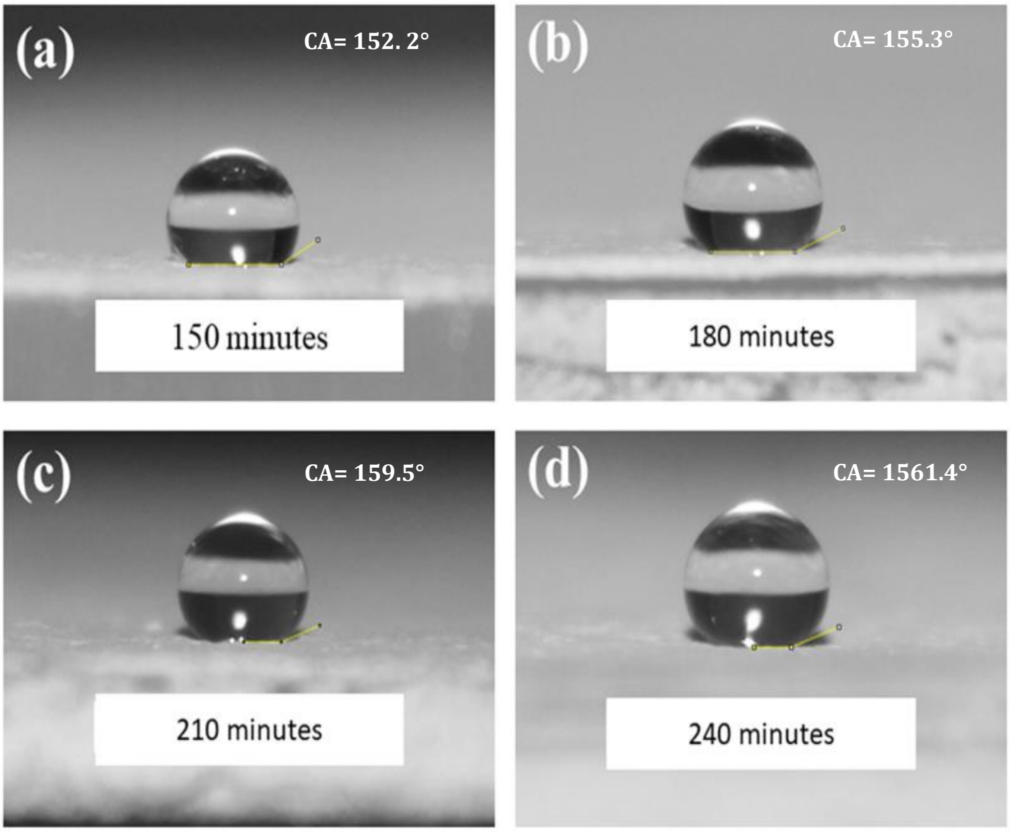


Figure 1. Water contact angle for (a) 150 minutes grinding time, (b) 180 minutes grinding time, (c) 210 minutes grinding time, and (d) 240 minutes grinding time.

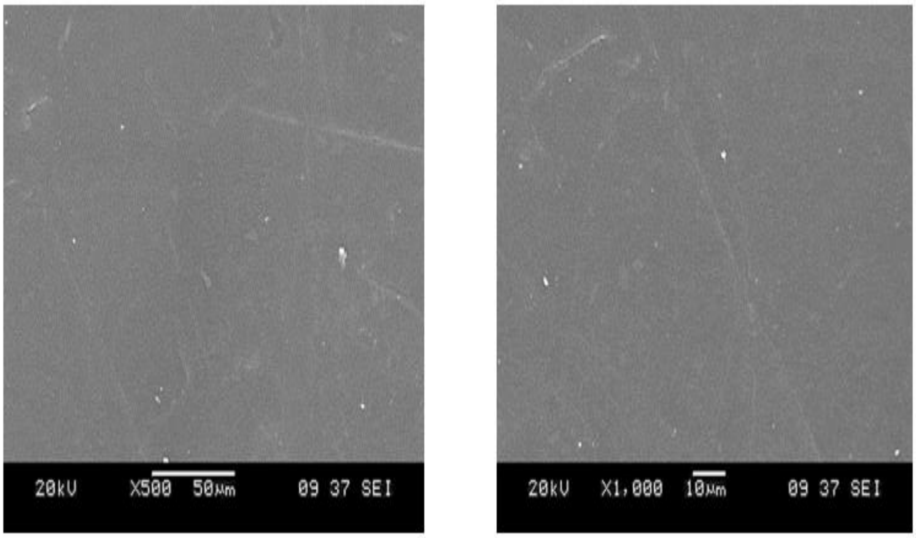


Figure 2. SEM image for control rHDPE sheet under magnification (a) 500x, and (b) 1000x.

As observed in Figures 3 (a)-(d), the 240-minute sample had greater porosity than the 210-minute sample. The 240-minute sample had the most porosity and few visible pores due the 240-minute hand-grinded NaCl having the smallest particles. Longer grinding time created more micro and nanoporous structures, enabling air to trap on the surface and resulting in a high water CA and high surface roughness. Therefore, the smaller the particle size, the more micro and nanoporous the surface sample becomes, enhancing the stability of the superhydrophobic surface [8].

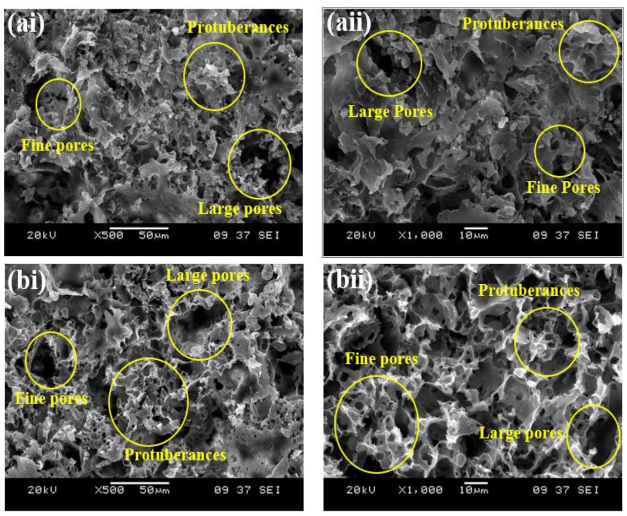
4.3. Energy Dispersive X-ray

Figure 4 (b) and (c) display EDX analysis of the surface-modified sample after leaching by an ultrasonic cleaner. The percentage illustrated in Figure 4 (c) indicates that virtually all of the Na (0.97%) and Cl (2.62%) were effectively leached from the sample, with less than 4%, which can be said to be negligible remaining on the surface of the rHDPE sample, demonstrating that the rHDPE samples were properly modified [9].

4.4. 3D Nano Profiler

Observing topography improves surface morphology. Figure 5 portrays surface topography for 240-minutes hand-grinded NaCl particles as a surface modification on rHDPE plastic sheet over large scanning areas (559.44μm x 361.12μm). Overall surfaces are rough due to microporous feature structures (small and big pores) and protuberances that form multiscale surface roughness for modified surfaces, as illustrated in Figure 5.

Consequently, models based on quantified measurements for wettability and surface roughness might be developed, yielding insights that could improve NaCl hand-grinding. As displayed in Figure 6 below, the graph for the surface roughness and waviness of the surface modified rHDPE. The average roughness (Ra) parameters and root mean square roughness (Rq) differ significantly. As can be observed in Figure 6 (a), the Ra of the sample was 27902.70 nm, whereas the Rq was 31782.05 nm. On the other hand, the average waviness (Wa) and root mean square waviness (Wq) parameters, as displayed in Figure 6 (b), were 43.98 nm and 55.40 nm, respectively. As a result, the modified sample's surface had multiscale properties, with the wavy patterns indicating microstructures and the roughness factor signifying nanostructures.



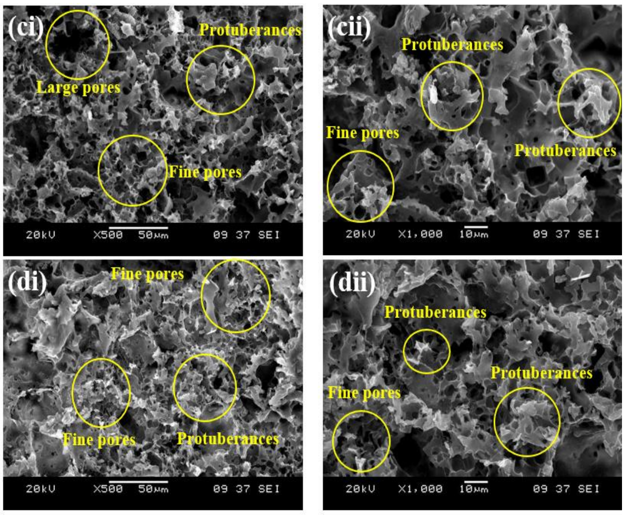


Figure 3. SEM image for surface modified rHDPE sheet under magnification (ai) 500x & 150 minutes ground, (aii) 1000x & 150 minutes ground, (bi) 500x & 180 minutes ground, (bii) 1000x & 180 minutes ground, (ci) 500x & 210 minutes ground, (cii) 1000x & 210 minutes ground, (di) 500x & 240 minutes ground, and (dii) 1000x & 240 minutes ground.

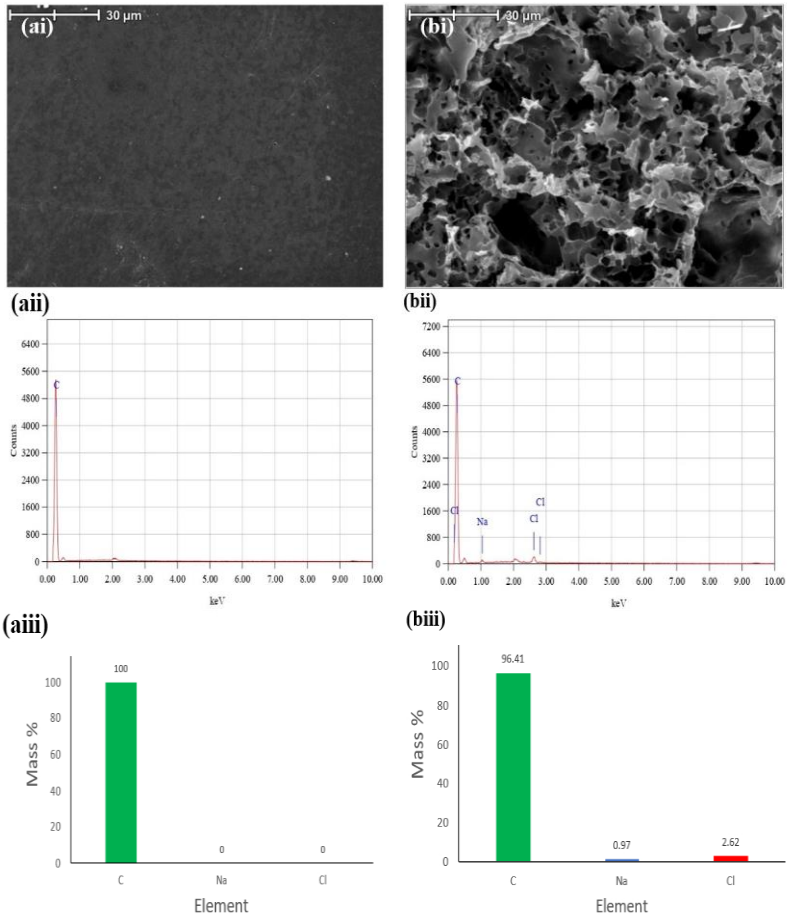


Figure 4. (ai) SEM micrograph for control, (aii) EDX spectra for the control sample, (aiii) mass % of an element discovered on the control sample surface, (bi) SEM micrograph for surface modified sample, (bii) EDX spectra for the surface modified sample and, (biii) mass % of the element discovered on the modified sample surface.

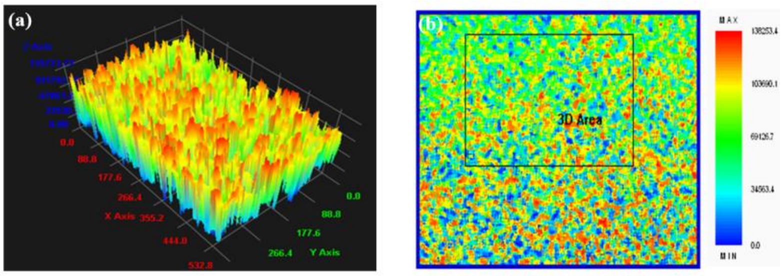


Figure 5. 3D nano profiler imaging for (a) 3D view of 240 minutes hand-ground of rHDPE sheet and (b) 2D view of 240 minutes hand-ground of rHDPE sheet.

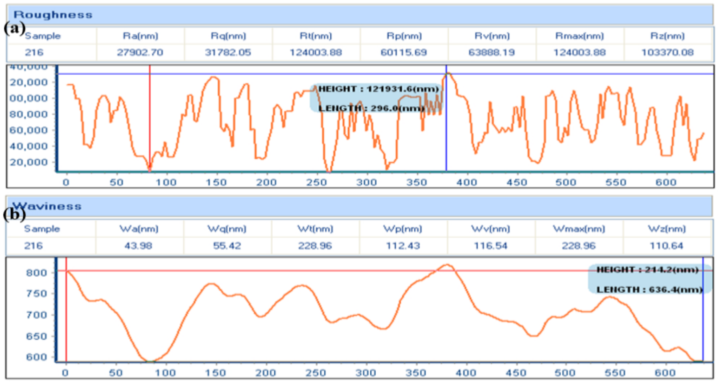


Figure 6. Surface roughness (a) and waviness (b) based on the black line showcased in the 2D image.

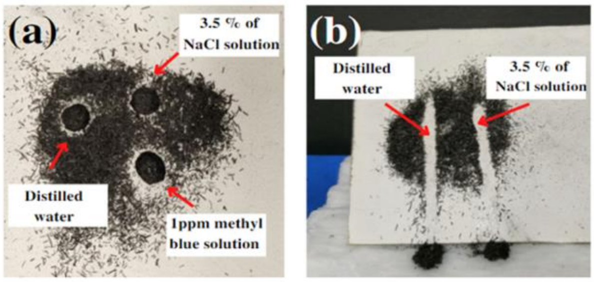


Figure 7. Self-cleaning ability test for (a) various type of liquid and (b) when the sample tilted.

4.5. Self-Cleaning Ability

The NaCl-modified rHDPE sheet displayed a self-cleaning ability as the charcoal powder, imitating dust, was easily washed away by water droplets when the sample is tilted. As illustrated in Figure 7 (a), the self-cleaning ability of a superhydrophobic modified rHDPE sample after collecting charcoal powder imitates dust. Each drop of methylene blue solution, 3.5% NaCl solution, and distilled water was deposited on the surface of superhydrophobic rHDPE sheet. Moreover, water droplets can completely remove charcoal powder from a contaminated surface. The methylene blue and 3.5% NaCl solutions behaved similarly to distilled water, as displayed in Figure 7 (a).

In Figure 7 (b), water droplets flowed when the sample was tilted 45°. After water droplets were dropped on the sample surface, they rolled off, clearing the charcoal powder along the way, and their surrounding region was clean. Note that no charcoal particles may remain on the water droplet's path. This indicates the self-cleaning ability of samples, as the sample may be quickly flushed by water without any residual particles, comparable to the lotus leaves' impact [10].

5. CONCLUSION

In this study, NaCl was proven to be a suitable surface modifier for performing surface modification on the rHDPE surface. In addition, micro and nanopores structures can be produced on the rHDPE surface using a green approach technique by modifying the surface microstructure. Hence, the smaller the porosity formed on the rHDPE surface, the greater the superhydrophobicity of the surface, resulting in a larger water CA and allowing water to roll off easily.

ACKNOWLEDGMENTS

The authors would like to express their gratitude and acknowledge the support from the Fundamental Research Grant Scheme (FRGS) under grant number FRGS/1/2020/TK0/UNIMAP/03/24 from the Ministry of Higher Education Malaysia.

REFERENCES

[1] Szewczyk, P. K., Ura, D. P., Metwally, S., Knapczyk-Korczak, J., Gajek, M., Marzec, M. M., Bernasik, A., Stachewicz, U. (2018). Roughness and fiber fraction dominated wetting of electrospun fiber-based porous meshes. Polymers. Volume: 11 Issue: 1.

[2] He, B., Yang, S., Qin, Z., Wen, B., Zhang, C. (2017). The roles of wettability and surface tension in droplet formation during inkjet printing. Scientific Reports. Volume: 1 Issue: 11841.

[3] Yuan, Y., Lee, T. R., "Contact angle and wetting properties,". in Surface Sciences Techniques, G. Bracco, B. Holst, Eds. Heidelberg, Berlin: Springer, (2013) pp. 3-34.Ren, J. X., Zhu, J. L., Shi, S. C., Yin, M. Q., Huang, H. D., Li, Z. M., Carbohydate Polymers. vol 296, (2022).

[4] Liu, Y., Choi, C. H. (2013). Condensation-induced wetting state and contact angle hysteresis on superhydrophobic lotus leaves. Colloid and Polymer Science. Volume: 291 Issue: 2. 437-445.

[5] Seo, K., Kim, M. (2014). Candle-based process for creating a stable superhydrophobic surface. Carbon. Volume: 68. 583-596.

[6] Zhao, X., Hu, T., Zhang, J. (2018). Superhydrophobic coatings with high repellency to daily consumed liquid foods based on food grade waxes. Journal of Colloid and Interface Science. Volume: 515. 255-263.

[7] Ochanda, F. O., Samaha, M. A., Tafreshi, H. V., Tepper, G. C., Gad-el-Hak, M. (2011). Salinity effects on the degree of hydrophobicity and longetivity for superhydrophobic fibrous coatings. Journal of Applied Polymer Science. Volume: 124 Issue: 6. 5021-5026.

[8] Ji, S., Ramadhianti, P. A., Nguyen, T. B., Kim, W. D., Lim, H. (2013). Simple fabrication approach for superhydrophobic and superoleophobic Al surface. Microelectronic Engineering. Volume: 111. 404-408.

[9] Sutar, R. S., Nagappan, S., Bhosale, A. K., Sadasivuni, K. K., Park, K. H., Ha, C. S., Latthe, S. S. (2021).

[10] Superhydrophobic Al2O3–Polymer Composite Coating for Self-Cleaning Applications. Coatings. Volume: 11 Issue: 10.

[11] Nagappan, S., Park, J. J., Park, S. S., Lee, W. K., Ha, C. S. (2013). Bio-inspired, multi-purpose and instant superhydrophobic–superoleophilic lotus leaf powder hybrid micro–nanocomposites for selective oil spill capture. Journal of Materials Chemistry A. Volume: 1 Issue: 23. 6761-6769.

[12] Yap, Y. W., Zakaria, M. S., Nordin, R. M., Halim, K. A. A., Lim, B. Y., & Abdurrahman, M. (2021). The potential of using sucrose particles for self-cleaning surface fabrication on recycled high-density polyethylene. Journal of Physics: Conference Series. Volume: 2080 Issue 1. 1-8.

[13] Gong, X., & He, S. (2020). Highly durable superhydrophobic polydimethylsiloxane/silica nanocomposite surfaces with good self-cleaning ability. ACS Omega. Volume: 5 Issue: 8. 4100-4108.