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Fabrication of hydrophilic silica coating varnish on pineapple peel fiber based biocomposite

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Abstract: In the last several years, the interest on hydrophobic and hydrophilic solid subtract was increased due to many applications in daily life, agriculture and industry. The continuous effort has been made to fabricate suitable material with more efficient fabrication method. In this research, physical blending method have been used by mixing four components of modifying agent with organic beeswax varnish at different weight percentage. Those modifying agents consists of decamethylcyclopentasiloxane (D5), Silica nanoparticles (R812S), Polydimethylsiloxane (BP-9400) and non-ionic surfactant (Triton X-100) were mixed in the mass ratio of 52:3:34:1. The modified varnish produced was coated on Pineapple peel fiber (PAPF) biocomposite and were characterized using Water Contact Angle (WCA) instrument, Fourier Transform Infrared (FTIR) and Scanning Electron Microscope (SEM). Based on the wettability index analysis; the value of water contact angle was reduced when increasing weight percent of modifying agents from 101.87 to 22.98°. The morphology of the surface was observed to have more silica nanoparticles, along with the increasing concentration of modifying agent. It also supported by FTIR which shows the presence of Si-O peak at 1030.88 cm^{-1}. These results proved that the modifying agent and organic beeswax varnish had successfully produced hydrophilic coated on the PAPF surface.

Keywords: silica nanoparticle, hydrophilic, packaging, varnish, physical blending

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1. Introduction

Wettability is the tendency of fluid to spread on or adhere onto a solid surface of material in present of other immiscible fluid. The angle that made by the fluids into the surface was called water contact angle that was depends on the liquid surface tension/interfacial tension and surface free energy (Enea et al. 2019). The surface roughness also gives an important factor on the wettability value of solid surface (Ji et al. 2019). Basically, there are three important wetting theory to explain the surface wettability; Young Model, Wenzel model and Cassie-Baxter model. Young model is suitable for smooth surface however it has limitation with rough surface of materials. Generally, the surface with static contact angle $\theta<90$ is hydrophilic and $\theta<10$ is super hydrophilic. Based on the Wenzel model, the initial contact angle of the roughen surface will decrease more if the initial contact angle of smooth surface 0<90°C (Young model); the opposite will happen if 0>90° (Senem, Kaya, and Cengiz 2019). Therefore,Vogler indicate that the boundary of hydrophobic to hydrophilic limit is 65° rather than 90° and some experimental results also supports this findings (Li et al. 2017). The two general routes to fabricate hydrophilic or super hydrophilic surface was depend on the appropriate roughness on the material surface or chemical modification of surface to be hydrophilic with contact angle < 65°(Song et al. 2013). As the results, creating the rough surface is important for both surface modifications on super hydrophilic or super hydrophobic.

Hydrophilic and superhydrophilic surfaces are widely used application due to the characteristic of self-cleaning, antifogging (Chen et al. 2013), antireflective properties (Geng, He, and Xu 2012), biomedical devices application (Tarmizi et al. 2018), pharmacological study (Sharif et al. 2018) , bioactive and for water treatment application (Ursino et al. 2019). Many efforts have been done to prepare suitable material and fabrication method such as sol gel method (Senem, Kaya, and Cengiz 2019), layer by layer coating (Geng, He, and Xu 2012), polymerization and anodic treatments (Ali et al. 2014) and various material such as silica nanoparticles, aluminium oxide (Zhang et al. 2018), titania (Saleh, Abdullah, and Yunus 2018), zeolite, have been study to achieve the requirement for hydrophilic or superhydrophilic.

In this current work, we report the fabrication of modified varnish using physical blending method between modifying agent and beeswax varnish. The aim was to study the wettability index of silica nanoparticles coatings on PAPF substrate by various mass ratio of silica nanoparticles. The incorporation of silica on the PAPF with beeswax varnish is verified using Fourier Transform Infrared (FTIR) and Scanning Electron Microscope (SEM) whereas the improvement take places on hydrophilicity was measured using water contact angle measurement.

2. Experimental

2.1 Materials

Beeswaxs varnish (food grade) was purchased by commercial store. Silicon oxide nanopowder (SiO_2) was supplied by US Research Nanomaterials, Inc, USA with average diameter size 20-30nm. Decamethylcyclopentasiloxane oil (D5) (Molecular weight: 370.77) was supplied by J&K Chemical (Shanghai) Co. Ltd., Polydimethylsiloxane (PDMS) was obtained from BOC Sciences, USA with Viscosity 500cSt. Nonionic surfactant (Triton X-100) was brought from Sigma Aldrich and all this chemical was used after received without further purification.

2.1 Functionalization of Beeswax Varnish

Two preparation steps were conducted to fabricate this modified varnish according to previous research with some modification (Chen et al. 2013). First step was related to preparation of the modifying agent prepared by mixing of four components (D5, SiO_2, PDMS and Triton X-100) with the mass ratio (57:3:9:1). The final beeswax varnish was prepared by additional of the modifying agent into commercial beeswax varnish with various mixing ratios, under homogenous stirring. The ratios used were (modifying agent: beeswax varnish percentage): 0:100, 40:60, 60:40 and 100:0. The details preparation protocol was discussed as follows. Firstly, the silica nanoparticles were dispersed into D5, and then PDMS was added into this mixtures, and then prepared mixture was blended homogenous with the beeswax commercial varnish. Finally, newly prepared modified varnish was obtained after non-ionic surfactant was added dropwise using the pipette tip under vigorous stirring for more 10 minutes.

2.2 Application of the modified varnish onto PAPF polymer

The modified varnish was sonicated for 10 minutes using water bath sonication before used. Afterward, this mixture was applied with into Pineapple peel fiber (PAPF) (1cm x 5cm) with equal amount of coated layer using spatula. This procedure was repeated using other ratio of modified varnish. After that, those coated pellets were dried in oven for overnight at 60°C.

2.3 Characterization of modified varnish

The functional group of the commercial varnish and modified varnish was evaluated using Fourier Transform Infrared (FTIR) spectrometer (IRTracer-100, Shimadzu, Japan). The samples were scanned in ATR mode with a 16 scan, resolution 16 cm^{-1} and wave number range from 650 to 4000 cm^{-1}. The force gauge was applied in the range of 100 to 120N. All samples were dried in an oven for 24 hours, put in desiccant before analysis to reduce absorbed moisture. The observation of the morphology was observed using Field Emission Scanning Electron Microscope (FESEM, JEOL JSM-6701F, USA). All the samples were coated with platinum for 60 second before being analyzed. The value of water contact angle was measured using water contact angle instrument (VCA 3000S TM, AST Product Ins. USA). All the sample was analyzed via sessile drop technique using 100µl syringe and 5µl of water was dropped onto sample. All samples were measured five times and averaged values were calculated. The contact angle value was presented as means value (n=5) with a standard deviation.

3. Results and discussion

3.1 Wetting properties of silica nanoparticles coating

According to the theory of Wanzel, surface roughness and void friction gives significantly effect on the wettability of water on surface. When the water contact angle was less than 65° the substance was indicating as hydrophilic and when the surface has water contact angle less than 5°, it is defined as super hydrophilic. Previous work by Geng et al 2012 was using silica nanoparticle (diameter 20nm) point out that higher surface roughness and higher porosity of silica nanoparticles coating was lead to lower water contact angle. This is because of the negatively charge of silica nanoparticles contributed to hydrophilic coating on the poly(methylmethacrylate). In this research, water contact angle test was conducted on the PAPF substrates with contains different weight ratio of modifying agent and beeswax varnish. The value of water contact angle and droplet image on the PAPF substrates was demonstrated in Fig.1. The water contact angle analysis shows that at 0 wt% of modifying agent, the water contact angle value was 101.87°.

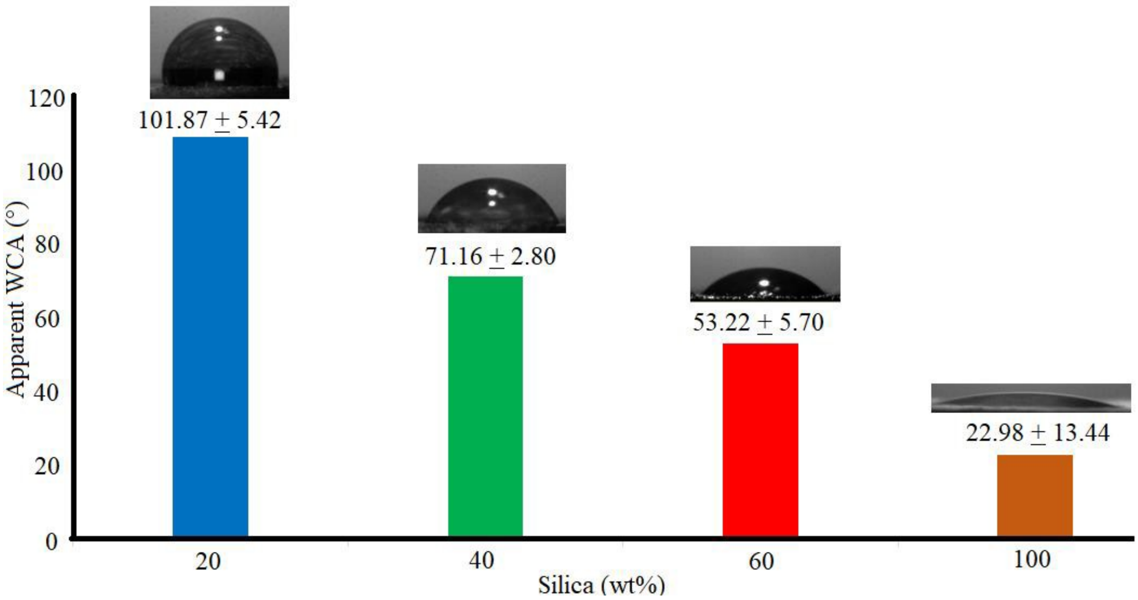

Fig 1 – Visual image of water droplets falling into subtract with the change of wt.% silica content.

By increasing the modifying agent to 40wt%, 60 wt% and 100wt%, decreasing of the water contact angle value can be observed from 71.16°, 53.22° and 22.98° respectively. It was clearly indicating the wettability index was changes form hydrophobic to hydrophilic due to existence of silica nanoparticles in the modifying varnish. Increasing percent of silica nanoparticles, the surface is rougher and the water contact angle on the coating decrease quickly with increase of time from 0 to 60 seconds.

3.2 Surface morphology and functional group of the coating

The surface morphology of the PAPF contains 0 wt% of the modifying agent, coated surface with 40 wt%, 60wt% and 100 wt% modifying agents were shown in Fig.2. Three magnification value was conducted which is 2000, 600 and 300 magnifications. Fig.2(a) until Fig.2(c) is referring to 0 wt% of modifying agent and 100% of beeswax varnish coated on the PAPF substrates. This PAPF morphology shows the flat and smooth surface with less roughness. With increasing concentration of the modifying agent, more and more silica nanoparticles were observed on the PAPF substrates. This hydrophilic silica was uniformly deposited and cover the PAPF surface. Clearly, the coating process on the surface (40 wt% of modifying agent) produced small island and valley indicates the surface is rough shows in Fig.2(d-f). With the increase of the weight percent of the modifying agent, the surface morphology becomes rougher. This finding also quite similar trend with (Senain, Nayan, and Saim 2010), reported that by increase TiO_2 percent after annealing, the surface morphology become rougher due to larger grain size of TiO_2.

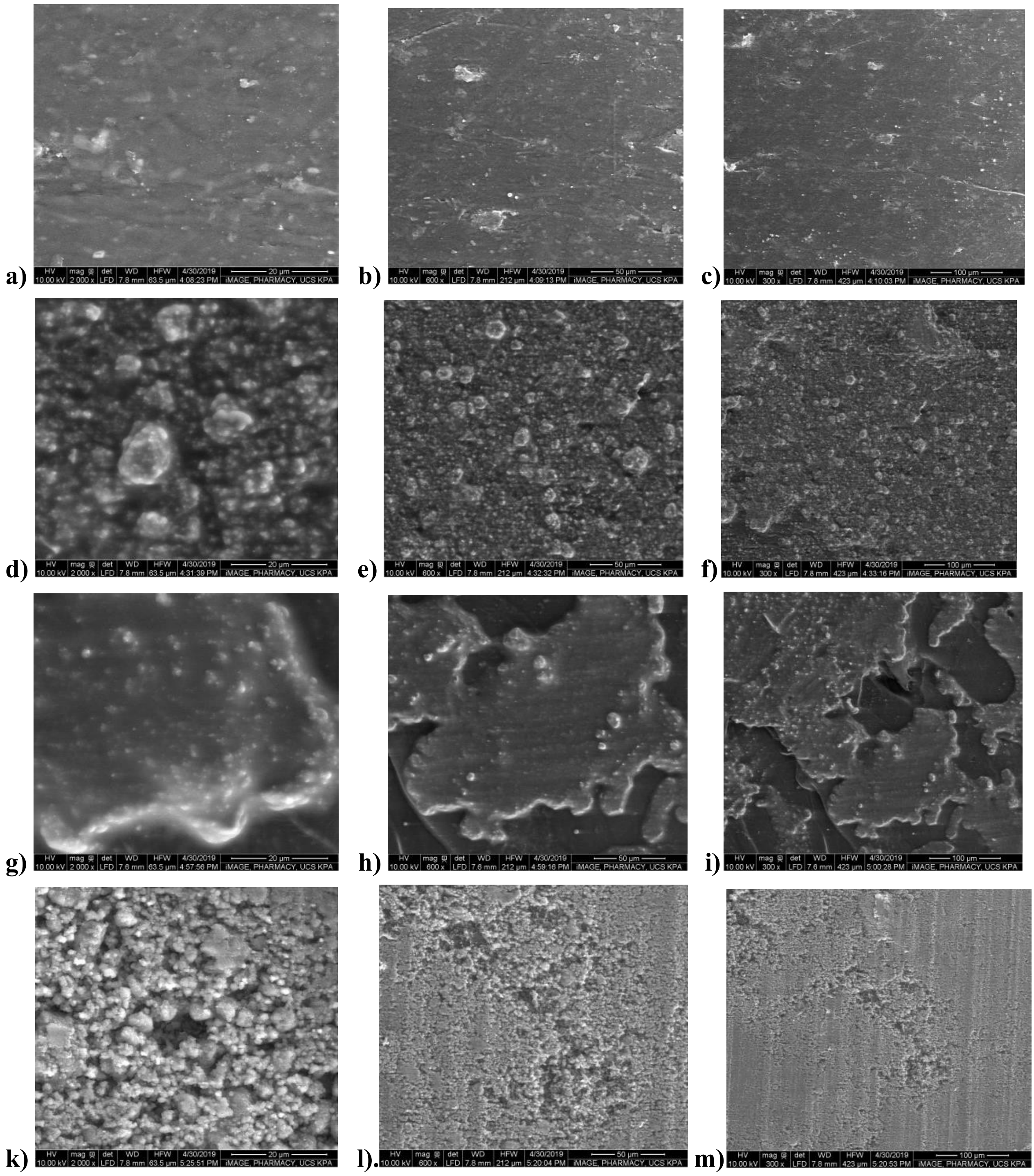

Fig. 2 - (a-c) beeswax coated at different magnification (20µm, 50µm and 100µm); (d-f) 40:60wt% of modifying agent:beeswax varnish at different magnification (20µm, 50µm and 100µm); (g-i) 60:40wt% of modifying agent: beewax varnish at different magnification; (k-m) 100:0 wt% of modifying agent:beeswax varnish at different magnification.

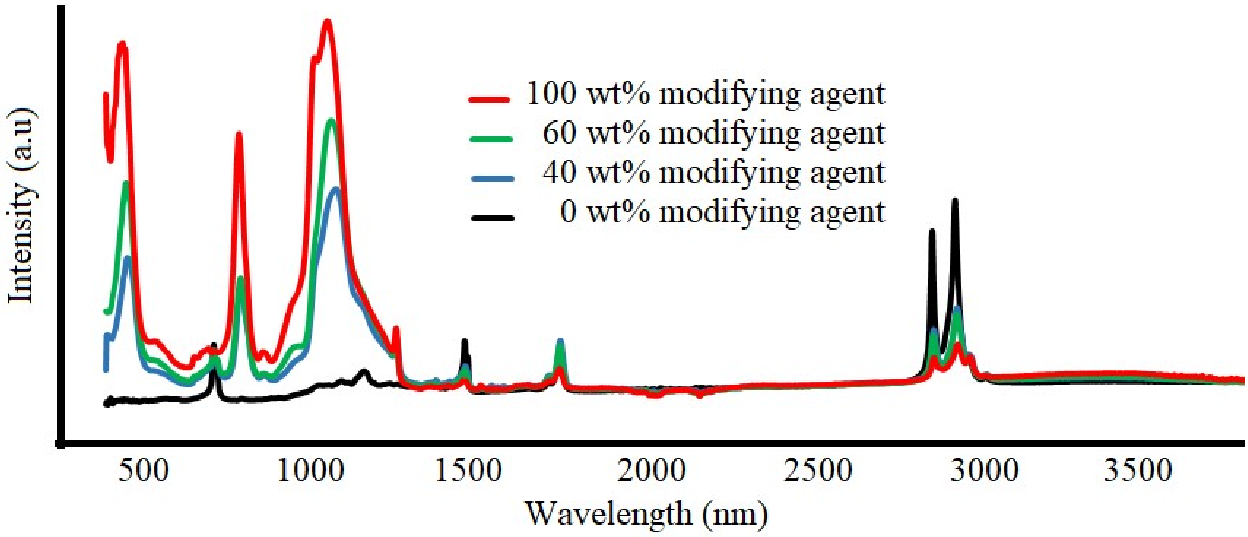

Fig 3 – ATR-FTIR spectra of PAPF, modified varnish comprised of 40 wt% modifying agent, 60wt% modifying agent and 100wt% modifying agent.

The functional group on the coating was analyzed using FTIR instrument via ATR modes and shown in **Fig.3**. The 0 wt% of the modifying agent consists of PAPF bio-composite, where the peak existence at 2916.38 cm^{-1} and 2848.86 cm^{-1} were representing methyl groups stretching vibration and the peak appears at 1462.04 cm^{-1} representing the scissoring and bending for methylene group (-C-H). After coating was applied onto PAPF surface, the strong intensity of C-H peaks become decreases due to the increasing amount of modifying agent on its surface. A very sharp absorption bands suddenly appeared at 1078.21cm^{-1}, and 798.53 cm^{-1} after PAPF subtract was coated with 40wt% modifying agent, assigned to Si-O-Si stretching vibration and the bending mode of the Si-O-Si group. This result also quite similar to the peak appear reported by Geng et al. 2012. By increasing silica weight percent into 60 wt% and 100 wt%, the Si-O stretching was shifted to 1066.64cm^{-1} and 1055.06cm^{-1}. However, the peak for Si-O bending in 60wt% modifying agent is still maintaining its value and was shift to lower wavelength (794.67cm^{-1}) when 100wt% of the modifying agent was applied. This situation shows indicating that the outermost surface is gradually increased with the silica content in the formulation. These results proved that the existence of Si-O on the surface of the materials, which play an important rule to change the hydrophilicity of the coating.

3. Conclusion

In the present study, different weight percent of modifying agent that contains four components were mixed with organic beeswax varnish was successfully produce using physical blending method. These silica nanoparticles coatings with different ratio of beeswax then were coated on the PAPF subtract for observation of hydrophilicity. Based on the wettability index, the value of the water contact angle was reduced with the increasing percentage of modifying agent. From the SEM image, the rougher surface was successfully produced which give important aspect to reduce the water contact angle value. FTIR spectrum revealed increased absorbent peak around 1078.21 cm^{-1} indicate the silica nanoparticles have been distributed well on the surface. The current results were expected to have the potential for the wall of greenhouse, coated for the wall curtain or glass for increase wettability and antifogging.

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References

[1] Ali, N., Duan, X., Jiang, Z.T., Goh, B.M., Lamb, R., Tadich, A., Poinern, G.E.J., Fawcett, D., Chapman, P., and Singh, P. (2014). Surface and interface analysis of poly-hydroxylethylmethacylate-coated anodic aluminium oxide membranes. Applied Surface Science, 289, 560-563.

[2] Chen,W., Wang X., Tao Q., Wang, J., Zheng Z., and Wang, X. (2013). Lotus-like paper/paperboard packaging prepared with nano-modified overprint varnish. Applied Surface Science, 266, 319-325.

[3] Enea, D., Bellardita, M., Scalisi, P., Alaimo, G., and Palmisano, L. (2019). Effects of weathering on the performance of self-cleaning photocatalytic. Cement and Concrete Composites, 96, 77-86.

[4] Geng, Z., He, J., and Xu, L. (2012). Fabrication of superhydrophilic and antireflective silica coatings on poly(methyl methacrylate) substrates. Materials research Bulletin, 47, 1562-1567.

[5] Ji, X., Zhao, D., Dai, C., and Xu, J. (2019). Dropwise condensation heat transfers on super hydrophilic-hydrophobic hybrid surface. International Journal of Heat and Mass Transfer, 132, 52-67.

[6] Li, S., Huang, J., Chen, Z., Chen, G., and Lai, Y. (2017). A review on special wettability textiles theoretical models, fabrication technologies and multifunction applications. Journal of Materials Chemistry A, 5, 31-55.

[7] Saleh, S.S., Abdullah, H.Z., and Muhammad Yunus, M., R. (2018). In vitro apatite deposition on titania film derived from electrochemical treatment on titanium substrate under mixed acid electrolyte. International Journal of Integrated Engineering, 10,102-108.

[8] Senain, I., Nayan, N., and Saim, H. (2010). Structural and electrical properties of TiO2 thin film derivd from sol-gel method using titanium (IV) butoxide. International Journal of Integrated Engineering, 2, 29-36.

[9] Senem, A., Kaya, A., and Cengiz U. (2018). Fabrication and application of super hydrophilic antifog surface by sol-gel method. Progress in Organic Coating, 126, 75-82.

[10] Sharif, F., Yunus, A.M., Saedudin RD., R., Hamid, A.A.A., and Kasim, S. (2018). Molecular docking and dynamics (MD) simulation of 6-gingerol and 6-shogaol against human estrogen receptor alpha (ERα). International Journal of Integrated Engineering, 10,119-127.

[11] Song, M., Liu, Y., Cui, S., Liu, L., and Yang, M. (2013). Fabrication and icing property of super hydrophilic and super hydrophobic aluminium surfaces derived from anodizing aluminium foil in a sodium chloride aqueous solution. Applied Surface Science, 283, 19-24.

[12] Tarmizi, Z.I., Nasef, M.M., Nallapan, M.L., Khairuddin, N.B.A., Ali, R.R. and Ahmad, A. (2018). Preparatin and characterization of hydrophilic polyurethane scaffolds by electrospinning and radiation-induced grafting of 2-hydoxylethyl methacrylate. AIP conference Proceedings. 1985, 030015(1)-030015(9).

[13] Ursino, C., Di Nicolo, E., Gabriele, B., Ciscuoli, A., and Figoli, A. (2019). Development of a novel perflouropolyethe (PFPE) hydrophobic/hydrophilic coated membranes for water treatment. Journal of membrane science, 581, 58-71.

[14] Zhang, D., Wang, G., Zhi, S., Xu, K., Zhu, L., Li, W., and Zheng, Z. (2018). Superhydrophilicity and underwater superoleophobicity TiO2/ Al2O3 composite membrane with ultra-low oil adhesion for highly efficient oil-in water emulsions separaton. Applied Surface Science, 458, 157-165.