

Characterization of Nanocomposite Mixture Polyvinyl Alcohol and Rice Husk Ash

Makmur Sirait^{1*}, Karya Sinulingga¹, Nurdin Siregar¹

¹ Department of Physics, Faculty of Mathematics and Natural Sciences, Universitas Negeri Medan, 20221, Indonesia

* Corresponding author's e-mail: maksir@unimed.ac.id

ABSTRACT

Rice husk ash (RHA) nanoparticles were prepared to be applied as a filler in the formation of Polyvinyl Alcohol (PVA) nanocomposites. The manufacture of rice husk ash nanoparticles involved the Ball Mill method and the coprecipitation method, while the manufacture of Polyvinyl Alcohol nanocomposite membranes and rice husk ash was carried out by the sol-gel method. The results of XRD analysis using Match software indicated that the crystal structure of rice husk ash is monoclinic with a particle size of 16.55 nm. The mechanical test results obtained the largest elastic modulus of 29.28 MPa in 3% rice husk ash mixture, the largest tensile test of 8.83 MPa in 1% rice husk ash mixture, and the largest elongation at break of 82.08% in 4% rice husk ash mixture. The addition of rice husk ash as a filler can improve the mechanical properties of PVA/rice husk ash nanocomposites.

Keywords: nanocomposite, PVA, rice husk ash, filler

INTRODUCTION

Rice husk is one of the agricultural wastes that has not been widely used as a product that has added value (Christopher, et al., 2017). Rice husk ash is a waste obtained from burning rice husk which contains silica. This rice husk is burned at high temperatures to produce pure silica content of 86.9–97.2%. Rice husk ash contains chemical components namely SiO₂, CaO, Na₂O, K₂O, Al₂O₃, and MgO. The higher the combustion temperature, the higher the pure silica content (Abdul, et al., 2020).

Rice husk ash has been used in mortar to improve physical and mechanical properties (Mahmud, et al., 2016) in addition to mixed cement and aggregate recycling (Qiang Su, et al., 2023; Musa, et al., 2022; Srinivasreddy, et al., 2013). Rice husk is mixed with animal dung to produce biogas (Ali, et al., 2016) and rice husk ash is used as an adsorbent for heavy metals and catalysts (Malahayati, 2021; Taha et al., 2014).

Rice husk ash has been synthesized as a nano-catalyst in producing biodiesel (Hazmi, et al.,

2021) and a nano-bifunctional super magnetic catalyst (Balkis, et al., 2020). The addition of rice husk ash composition affects the physical and chemical properties of the compost, whereas the addition of rice husk ash improves the physical and chemical properties of the compost (Hisham and Ramli, 2019).

One type of environmentally friendly polymer the application of which is increasing and being observed is polyvinyl alcohol (PVA). The main functional uses of PVA include filtration, catalysts, membranes, optics, enzyme mobilization, and tissue engineering (Silverio, 2013) when mixed with rice husk ash, it can be employed in ultrafiltration and dye removal (Khairul, et al., 2022; Haq, et al., 2020). PVA has several superior properties, including oxygen resistance, mechanical properties, chemical resistance, film-forming ability, and water solubility (Tian, et al., 2017). PVA is an excellent material as an emulsifier and adhesive for a material because it is colorless, odorless, non-toxic, and has elasticity, flexibility, as well as high oxygen content (Goodship, et al., 2009). PVA is the most widely produced polymer

today, especially for industrial fiber applications, especially in pharmaceutical, biomedical, biochemical applications, due to its biocompatibility, biodegradability, and water-soluble properties (Liu, et al., 2016).

The addition of nanocellulose to PVA films improves mechanical and thermal properties (Qiu, et al., (2012; Ibrahim, et al., 2010). The ability of nanocellulose to improve film characteristics is influenced by the size, diversity, source, nanocellulose content, and plasticizer. Sirait, et al. (2021) synthesized limestone mixed with polyvinyl alcohol to obtain a nanocomposite membrane that has good mechanical properties in a 1% mixture. Polyvinyl alcohol is used as a matrix to be mixed with collagen as a wound dressing to prevent bacterial growth in the wound as well as accelerate wound healing and growth of new tissue (Sudirman, et al., 2020). This study aimed to utilize rice husk ash as a filler to produce nano-composites that can improve their mechanical properties.

MATERIALS AND METHODS

Materials

The main materials used in this study were rice husk and fully hydrolyzed polyvinyl alcohol Sigma Aldrich, 5M HCl, NH_4OH Emsure Merck, and *Aquadest Pro Analysis*.

Methods

Synthesis of rice husk ash nanoparticles

Rice husk was cleaned and dried and then burned using a furnace at 700 °C for 2 hours. Then it was milled with a Ball Mill for 6 hours until smooth to obtain nanoparticle size. After that, the rice husk ash was chemically cleaned with 5M HCl with a 300 rpm magnetic stirrer for 4 hours and stirred with NH_4OH solution until the pH was neutral, then filtered through filter paper and dried in the oven.

Preparation of nanocomposites PVA/RHA

Preparation of polyvinyl alcohol nanocomposite membranes with a mixture of rice husk ash using the sol-gel method. Polyvinyl alcohol was weighed in the amount of 20 g and then dissolved

in 200 ml of distilled water, and rice husk ash was dissolved in distilled water then both were mixed while stirring with a magnetic stirrer on a hot plate and heated at 80 °C to form a gel, then placed into the mold and released until dry. Rice husk ash powder (RHA) mixed with polyvinyl alcohol (PVA) with variations in composition (PVA: RHA) are (100:0)%; (98:2)%; (96:4)%; (94:6)% and (92:8)%. After that it was characterized by XRD test to determine its structure, SEM for morphology. Samples were formed with ASTM D636 type 5 for mechanical testing with the Universal Testing Machine.

RESULTS AND DISCUSSION

The results of the synthesis of rice husk ash using the ball mill and coprecipitation method are shown in Figure 1. Figure 1a) Rice husk ash in a furnace at 700 °C for 2 hours has a reddish color. Figure 1 b) is the result after chemical cleaning in a ball mill and is whitish and finer in color.

The XRD used has a $\text{Cu-K}\alpha$ wavelength of 1.54056 Å with the resulting pattern as shown in Figure 2. The size of the resulting rice husk ash nanoparticles is 16.55 nm with a density of 2.250 g/cm³. The content of rice husk ash is mostly composed of silicate hydrate, silicate oxide, and aluminum phosphate. In Figure 2 it can be seen that the peak of the highest pattern is at 2θ of 19.58°, 20.50°, 21.72°, 36.04°. The resulting crystal structure is monoclinic with crystal planes (dhkl) [0 0 4] and lattice parameters are as follows: a is 25.9190 Å, b is 5.0040 Å, c is 18.5400 Å.

Figure 3 is the morphology of the rice husk ash particles that have been synthesized at 1,500× and 5,500× magnification. From Figure 3(a) it can be seen that the rice husk ash particles are uneven and have lumps, this is also clarified in Figure 3(b) with a magnification of 5,500×. The particle size is still not homogeneous as can be seen from the presence of lumps that form on the surface, where the obtained surface is uneven and has lumps. This is due to imperfect synthesis processes such as in terms of ball mills, and less stable mixing during coprecipitation and heating. This is confirmed by the research of Nurdin, et al (2023).

Mechanical test using Universal Testing Machine with ASTM D638. This tool can determine the tensile test, elongation at break, and Young's



Figure 1. Rice husk ash results (a) furnaces and (b) ball mills

modulus. The results of the mechanical test characterization of the nanocomposite mixture of polyvinyl alcohol and rice husk ash with variations in the composition of rice husk ash (RHA) were 0%, 1%, 2%, 3%, and 4% as shown in the following graph.

Figure 4 shows the relationship between tensile strength and the composition of RHA. The highest tensile strength was found in the addition of 1% composition RHA is 8.83 MPa, while the lowest tensile strength was in the composition of 3% rice husk ash is 7.31 MPa. From the graph above, it can be concluded that the more rice husk ash is added to polyvinyl alcohol, the more the tensile strength tends to increase (Pereira, et al., 2014).

Figure 5 shows the relationship between the elongation at break and the composition of rice husk ash, where the highest elongation at break was 82.02% with the addition of 4% composition RHA. Thus, the addition of rice husk ash to polyvinyl alcohol can increase the flexural properties of nanocomposites, this is also according to the research by Pakravan, et al., 2018. Meanwhile, the lowest elongation at break was 25.47% with the addition of 3% RHA. A decrease in elongation at break can occur due to a lack of homogeneity when the process of mixing rice husk ash with a solution of the polyvinyl alcohol composite or the pore size of the nanocomposite is too high.

Figure 6 shows the relationship between Young's Modulus with the composition of rice

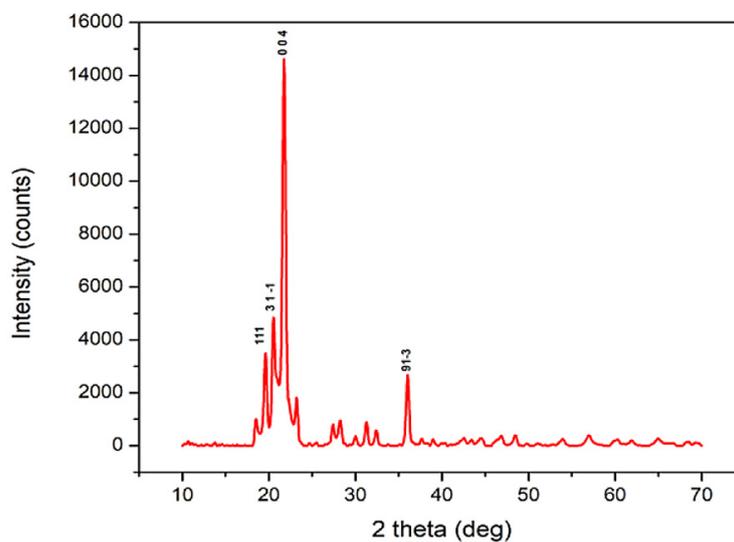


Figure 2. XRD diffractogram rice husk ash

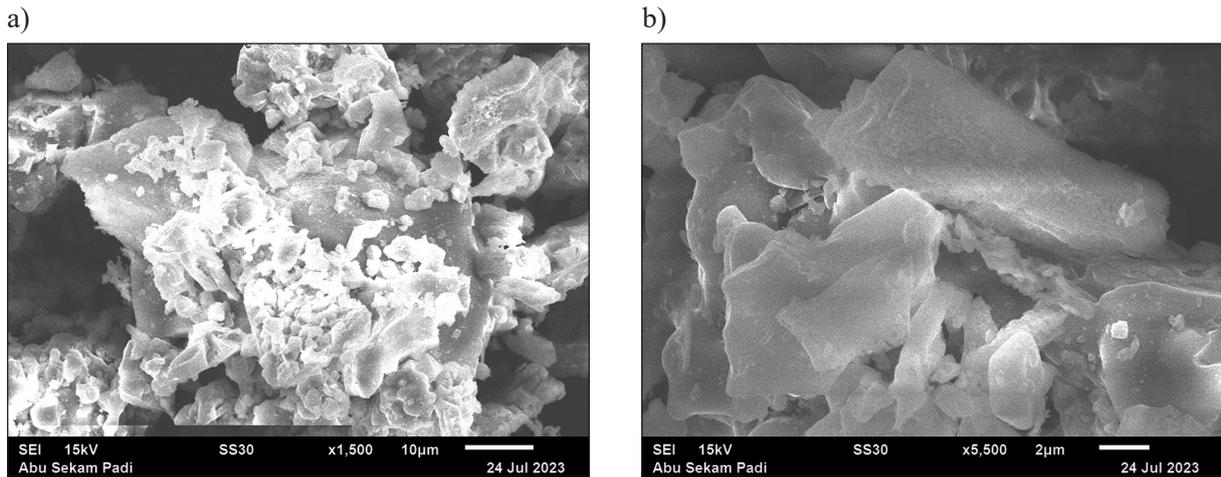


Figure 3. Morphology of rice husk ash with magnification: a) 1,500×; b) 5,500×

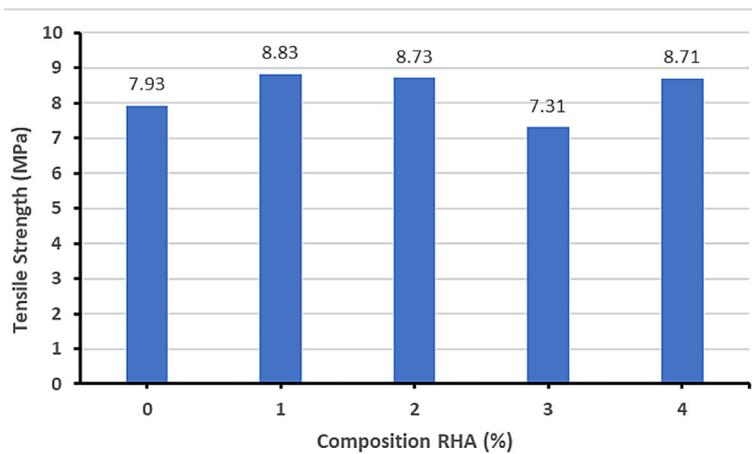


Figure 4. Tensile strength with composition RHA

husk ash, where the highest Young’s Modulus is found in the addition of 3% rice husk ash, which is 29.28 MPa. Meanwhile, the lowest Young’s Modulus was found in the addition of 4% rice husk ash, namely 10.76 MPa. It can be concluded that the addition of rice husk ash to polyvinyl alcohol

can improve its mechanical properties, namely Young’s modulus, where a 3% mixture produces 29.28 MPa higher than without other mixtures and mixtures. From the three figures above it can be seen that if the tensile strength is greater and the elongation at break is small, Young’s Modulus

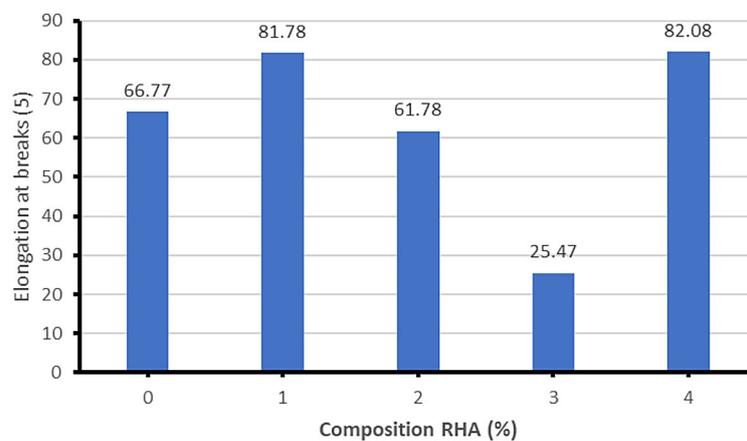


Figure 5. Elongation at break with composition RHA

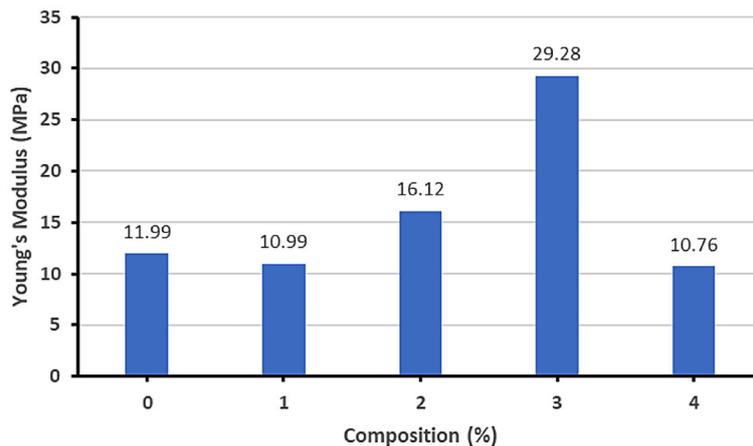


Figure 6. Young's modulus with composition RHA

is greater. In the composition of 3% rice husk ash, it can be seen that the mixture of rice husk ash is also more evenly distributed and the nanocomposite membrane has better features. The addition of rice husk ash to polyvinyl alcohol can improve modulus Young's and tensile strength (Ain, et al., 2020; Roohani, et al., 2008).

CONCLUSIONS

The results of the synthesis of rice husk ash using the ball mill method obtained a size of 16.55 nm, a monoclinic structure with the majority content of silicate hydrate, silicate oxide, and aluminum phosphate. The SEM results indicated that the morphology of the husk ash was still uneven and agglomeration or clumps still occurred. The mechanical test results obtained the largest elastic modulus of 29.28 MPa in 3% rice husk ash mixture, the largest tensile test was 8.83 MPa in 1% rice husk ash mixture and the largest elongation at break was 82.08% in 4% rice husk ash mixture. The addition of rice husk ash affected the tensile strength and Young's modulus, the more rice husk ash, the higher the tensile strength.

Acknowledgements

Thanks are conveyed to the Rector and Head of the Research and Community Service Institute who have funded the 2023 PNB with Number 0137/UN 33.8/KPT/PPT/2023, as well as other related parties who have supported this research until it was completed.

REFERENCES

1. Abdul Wahab, R.A., Mohd Zaid, M.H. Aziz, S.H.A., Amin Matori, K., Fen, Y.W., Yaakob, Y. 2020. Effects of sintering temperature variation on synthesis of glass-ceramic phosphor using rice husk ash as silica source. *Materials*, 13(23), 5413.
2. Ain, H., Putranto, A., Polonia, BSE, Ravi, A. 2020. The effect of rise husk ash on concretemixing quality formula. *Jurnal Gradasi Teknik Sipil*, 4(1), 29–36.
3. Ali, Ghaffar, Bashir, Khalid, M., Hassan, A & Bashir, Hamid, M. 2016. Utilization of rice husk and poultry wastes for renewable energy potential in Pakistan: An economic perspective. *Renewable and Sustainable Energy Reviews*, 61, 25–29.
4. Balkis Hazmi, Umer Rashid, Yun Hin Taufiq-Yap, Mohd Lokman Ibrahim, Imededdine Arbi Nehdi. 2020. Supermagnetic nano-bifunctional catalyst from rice husk: Synthesis, characterization and application for conversion of used cooking oil to biodiesel. *Catalysts*, 10, 225.
5. Christopher Fapohunda., Bolatito Akinbile., Ahmed Shittu. 2017. Structure and properties of mortar and concrete with rice husk ash as partial replacement of ordinary portland cement – A review. *International Journal of Sustainable Built Environment*, 6(2), 675–92.
6. Goodship, Vannessa and Jacobs, D.K., Ogur, E. 2009. Polyvinyl alcohol: materials, processing and applications. *Rapra Review Reports*, 16(12).
7. Haq N.B., Yusra Safa., Sobhy M.Y., Omar H. Shair., Munawar Iqbal., Arif Nazir. 2020. Efficient removal of dyes using carboxymethyl cellulose/ alginate/ polyvinylalcohol/ rice husk composite: adsorption/ desorption, kinetics and recycling studies. *International Journal of Biological Macromolecules*, 150(1), 861–870.
8. Hazmi, B., Rashid, U., Ibrahim, M.L., Nehdi, I.A., Azam, M., Al-Resayes, S.I. 2021. Synthesis and

- characterization of bifunctional magnetic nanocatalyst from rice husk for production of biodiesel. *Environ. Technol. Innov.*, 21, 101296.
9. Hisham, N.E.B. dan Ramli, N.H. 2019. Effect of rice husk ash on the physicochemical properties of compost. *Indonesian Journal of Chemistry*, 19(4), 967–74.
 10. Ibrahim, M.M., Waleed, K.E., Mona, A.N. 2010. Synthesis and characterization of polyvinyl alcohol/nanospherical cellulose particle films. *Carbohydrate Polymers*, 79(3), 694–99.
 11. Khairul Rahmah., Sri Aprilia., Farid Mulana., Zuhra., Syaubari., Sofyana., Lia Meiriza., Amri Amin. 2022. Characteristics of modified polyvinyl alcohol/polyether sulfone membranes grafted silica from rice husk ash for ultrafiltration. *Materials Today: Proceedings*, 63(1), S110–14.
 12. Liu, D., Qibo, B., Yan, L., Yaru, W., Aimin, X., Huafeng, T. 2016. Effect of oxidation degrees of graphene oxide on the structure and properties of poly (vinyl alcohol). *Composite Films Composites*, 40, 645–56.
 13. Mahmud, H.B., Syamsul Bahri, Y.W. Yee., Y.T., Yeap. 2016. Effect of rice husk ash on strength and durability of high strength high-performance concrete. *International Scholarly and Scientific Research & Innovation*, 10(3).
 14. Malahayati., Evi Yufita., Ismail., Mursal., Rinaldi Idroes., Zulkarnain Jalil. 2021. The effect of natural silica from rice husk ash and nickel as a catalyst on the hydrogen storage properties of MgH₂. *Journal of Ecological Engineering*, 22(11), 79–85.
 15. Musa Aminu Alhaji., Pushpendra Kumar Sharma. 2022. Combined effects of rice husk ash and hybrid fibers on the fresh and mechanical properties of recycled aggregate concrete. *Materials Today: Proceedings*, 62, 6695–6700.
 16. Nurdin Bukit, Karya Sinulingga., Abd Hakim S., Makmur Sirait., Bunga Fisikanta Bukit. 2023. Mechanical and thermal properties of HDPE thermoplastic with oil palm boiler ash nano filler. *Journal of Ecological Engineering*, 24(9), 355–363.
 17. Pakravan, H, R., Jamshidi, M., Asgharian Jeddi A.A. 2018. Combination of ground rice husk and polyvinyl alcohol fiber in cementitious composite. *Journal of Environmental Management*, 215, 116–22.
 18. Pereira, A.L.S., Do, N., Morais, J.P.S., Vasconcelos, N.F., Feitosa, J. P. 2014. Improvement of polyvinyl alcohol properties by adding nanocrystalline cellulose isolated from banana pseudostems. *carbohydrate polymers*, 112, 165–72.
 19. Qiang Su., Jinming Xu. 2023. Mechanical properties of concrete containing glass sand and rice husk ash. *Construction and Building Materials*, 393.
 20. Qiu, K., Anil, N., Netravali. 2012. Fabrication and characterization of biodegradable composites based on micro fibrillated cellulose and polyvinyl alcohol. *Composite Science and Technology*, 72, 1588–1594.
 21. Roohani, M., Youssef, H., Naceur, M, B., Ghanbar E., Ali, N.K., and Alain D. 2008. Cellulose whiskers reinforced polyvinyl alcohol copolymers nanocomposites. *European Polymer Journal*, 44, 2489–2498.
 22. Silverio, A.H. 2013. Effect of incorporating cellulose nanocrystals from corncob on the tensile, thermal, and barrier properties of poly (vinyl alcohol) nanocomposites, research article. *Journal of Nanomaterials*. Hindawi Publishing Corporation.
 23. Sirait, M., Sinulingga, K., Siregar, N., Fitri, D., Padang, S.T.W. 2021. Thermal and mechanical properties a membrane of the mixing PVA nanocomposite and limestone hydroxyapatite. *Journal of Physics: Conference Series*, 1811.
 24. Srinivasreddy, Avinash Babu., McCarthy, Timothy J., Lume, Eric. 2013. Effect of rice husk ash on workability and strength of concrete. *Faculty of Engineering and Information Sciences – Papers: Part A*.
 25. Sudirman., Aloma Karo Karo., Sulistioso Giat Sukaryo., Karina Dwi Adistiana., Kiagus Dahlan. 2020. Synthesis of nanofiber from poly vinyl alcohol (PVA)-collagen using electrospinning methods. *J. Kim. Terap. Indonesia*, 21(2), 55–65.
 26. Taha, Mohd F., Shuib, Anis Suhaila, Shaharun., Maizatul S, Borhan., Azry., Dass., Sarat Chandra., Guan., Beh Hoe., Yahya., Noorhana. 2014. Removal of Ni (II), Zn (II), and Pb (II) ions from single metal aqueous solution using rice husk-based activated carbon. Paper resented at the AIP Conference Proceedings.
 27. Tian, H., Jiaan, Y., Varada, R., Aimin, X., Xiaogang, L. 2017. Fabrication and properties of polyvinyl alcohol/starch blend films, effect of composition and humidity. *International Journal of Biological Macromolecules*, 96, 518–523.