MATERIALE PLASTICE
https://revmaterialeplastice.ro
https://doi.org/10.37358/Mat.Plast.1964

Acoustic and Mechanical Properties of Polystyrene Composite Filled with Primary Sludge and Boiler Ash from Pulp Mill

TENGKU RACHMI HIDAYANI[1,2], BASUKI WIRJOSENTONO[3*], DARWIN YUNUS NASUTION[3], TAMRIN TAMRIN[3]

[1]Postgraduate Study Program, Department of Chemistry, Faculty of Mathematics and Natural Science, Universitas Sumatera Utara, Medan, 20155, Indonesia

[2]Department of Agribisnis Kelapa Sawit, Politeknik Teknologi Kimia Industri, Medan, 20288, Indonesia

[3]Department of Chemistry, Faculty of Mathematics and Natural Science, Universitas Sumatera Utara, Medan, 20155, Indonesia

Abstract: *The search for green composite for sound-absorbing material has increased a significant interest. In this study, polystyrene (PS) composite foam filled with primary sludge and boiler ash from pulp mill was produced. The product was characterized using analytical techniques such as FTIR, XRD, SEM, TGA, and DSC, while the mechanical properties and sound-absorbing coefficient were analyzed regarding ASTM D 2240 and ASTM E-1050-98, respectively. The results showed that the sludge has 1β crystalline polymorph cellulose from the pulp. The chemical reaction can be seen by the chemical bond formed during the mixing process. The peak expansion occurred at 1600.44 cm^{-1} indicating the presence of a C=C functional group of maleic anhydride with polymer and filler. SEM images showed that the filler was distributed and dispersed into the polymer matrix and no agglomeration was produced. The sound-absorbing composites had complied ISO 11654:1997 regarding the sound absorption coefficient rating level on materials for rooms with sound absorption classes D and C with the value of α 0.328-0.793.*

Keywords*: boiler ash, noise level, primary sludge, pulp mill, sound-absorbing material*

1. Introduction

Noise pollution is a critical issue, especially for those who live in urban areas. The sources of noise can come from human activities such as road traffics, machinery, electronic devices, industrial and civil entities which influence the environment, health and our comfort. Acoustic comfort has been revealed by physicians due to having several problems for human beings such as hearing loss, cardiovascular diseases, and sleep order, and has become an increasingly important concern for both civil engineering and urban design [1,2]. Hence, it is needed to produce sound-absorbing material which is applied in several applications in building, automobile, aerospace industries, and industrial noise instruments [3].

The most widely used precursor for sound-absorbing materials is polyurethane and polystyrene (PS) foam due to their lightweight and ease of cutting installation [4]. PS is easily amalgamated into concrete to produce lightweight concrete, and it is recyclable. There are two possibilities of using PS in construction materials, (i) the waste of PS can be controlled to prevent environmental pollution, (ii) the new product of PS added resin can be potential construction and insulation materials which are used as partition walls, floorings, ceiling concretes, brick and outer plaster [5].

Sound-absorbing-based composite materials have several advantages: lightweight, fuel-economical, and environmentally friendly. In addition, they convert the impact of kinetic energy to some form of deformation absorbed energy which is higher in energy absorption capacity compared to energy absorbers made from metallic materials, converting the kinetic energy to plastic deformation energy [6]. PS has been mixed with tragacanth as inner plaster or insulation plaster, it is lightweight, energy savings, saving building heating and cooling energy [7]. PS-based particleboard composites were used as fire-resistance, sound-absorbing, and thermal-insulated [8]. In building insulation application, PS and natural gypsum were combined showing that higher pores of PS increased the thermal conductivity, but de-

**email: basuki@usu.ac.id*

decreased the mechanical properties of the composite. PS also decreased the density of the composite and improved the absorption compared to the conventional concrete sample. PS and natural rubber were blended containing bamboo powder as sound damping material. The presence of bamboo increased the sound absorption coefficient (α) of the composites [10].

Furthermore, bio-based building materials become more attractive to be developed due to their being inexpensive, renewable resources, reduction of greenhouse gas emissions, and ecofriendly. One example is hemp concrete which has been widely used in building materials as acoustic dampers [11]. A review from Madushika 2021 stated that sound absorption coefficients of modified fiber structures have a comparable value with traditional materials. Therefore, traditional toxic sound-absorbing materials such as fiberglass and mineral fibers could be replaced using textile fibrous materials by modifying fibers chemically or physically [12]. Cereal straw has been used as a component of insulation material [13]. A good sound absorber material has been successfully produced by layered composite materials based on lignocellulosic waste with low price and minimum impact on the environment, with a medium sound absorption coefficient (α) = 1 [14]. Raw groundnut shell was reported as a sound-absorbing material and it can be used as a promising precursor of sound absorption with a sound-absorbing coefficient at medium and lower frequency [3]. Khalil 2014 studied the physical and mechanical properties of gypsum plaster composites with the addition of waste additives, e.g. unburnt rice husk blast furnace slag, calcium carbonate, or polyvinyl alcohol polymer. The mechanical properties of gypsum composite did not improve with the added wastes; however, it reduced the pollution to a clean environment and low-economical and suitable for desert buildings or similar climatic conditions [15].

From a previous study, we have reported composite panel-based polypropylene waste, coco fiber, and sludge as a filler [16]. However, the sound-absorbing capacity has not been measured. In this present study, foam composite-based-PS was developed as a sound-absorbing material that was mixed with maleic anhydride, polyurethane primary sludge, and boiler ash from pulp industry. Primary sludge and boiler ash are by-products produced from pulp and paper industry. Howevwe, the waste has not been managed well, especially in Indonesia. Hence, it was become a long debate involving the surrounding community. Therefore, this study is expected be a solution for the utilization of existing waste in the factory and to produce a green sound absorber material.

2. Materials and methods

2.1. Materials

The materials used in this study were PS-type general-purpose $(C_8H_8)_n$, polyurethane ($C_{27}H_{36}N_2O_{10}$), xylene $(CH_3)_2C_6H_4$, and maleic anhydride ($C_4H_2O_3$) purchased from Sigma Aldrich. All materials were used without further purification. Primary sludge and boiler ash were collected from Indah Kiat Pulp & Paper Tbk., Riau, Indonesia.

2.2. Preparation of primary sludge and boiler ash

Primary sludge and boiler ash were prepared before using as a filler into PS composite. Firstly, primary sludge was dried under the sun for 7 days and put into a vacuum oven at 100^{o}C for 6 h. The dried primary sludge was blended using a blender and sieved with a 400-mesh sieve. Meanwhile, boiler ash was pulverized to 74 µm in dimension. It was then purified using 2 M HCl, boiler ash:HCl (1:10), and distilled for 2 h. The boiler ash and HCl solution were filtered and washed with distilled water until *p*H neutral. The solid product was calcinated at 600^{o}C for 2 h and allowed to cool at room temperature after the calcination process was finished. Boiler ash was put into a ball mill (Planet PM 200) for 10 h to obtain a smaller particle size. Finally, primary sludge and boiler ash was characterized using FTIR, PSA, and DSC.

2.3. Preparation of composite PS/maleic anhydride primary sludge / boiler ash / polyurethane

The amount of 50g of PPS was put in a three-necked flask and added 100 g xylene, 1% maleic anhydride and 1% benzoyl peroxide. It was reflexed at 100^{o}C for 2 h to produce PS grafed maleic anhydride (PS-g-MA). After that, the amount of primary sludge, boiler ash, and polyurethane was mixed into the reflux and stirred at 100°C for 2h with the composition summarized in Table 1. The compoosite was then molded using a compression press. It was characteized using FTIR, XRD, SEM, TGA mechanical, and sound absorption properties.

Table 1. The composition of PS, maleic anhydride, primary sludge, boiler ash, and polyurethane for PS foam composite

Sample code	PS (%)	PS-g-MA (%)	Primary sludge (%)	Boiler ash (%)	Polyurethane
PS1	1000	0	0	0	0
PS2	75	10	10	5	0
PS3	75	10	5	5	5
PS4	70	10	5	5	10

2.4. Characterization

A particle size analyzer (LA-910, Horiba LA-910, Horiba Ltd, Kyoto, Japan) was used to analyze the particle size distribution of primary sludge and boiler ash which was collected from a by-product of the pulp industry. Fourier-transform infrared (FTIR) spectrometer (Nicolet 380, Thermal Scientific, Boston, USA) was used to investigate the chemical structure of samples in a transmission mode with a resolution of 5 cm^{-1} and 100 scans. The crystallinity of samples was investigated using x-ray diffraction (XRD, Bruker D8, Bruker Optic GmbH, Germany) with λ = 0.154 nm at scanning of 2°/min, a voltage 40 kV, and a current 200 mA. The morphology of PS composite foam was analyzed using scanning electron microscopy (SEM, Hitachi TM3030, JEOL, Ltd., Tokyo, Japan). The instrument was operated at 20 kV with a magnification of 5.000. Thermal analysis of materials was characterized using a thermogravimetric analyzer (DTA/TG Exzztar SII 7300, Hitachi medical system, Tokyo, Japan) and differential calorimetry (DSC) X-DSC7000 (Hitachi medical system, Tokyo, Japan). Both instruments were operated from room temperature to 600°C with a 10°C/min heating rate. ASTM D 2240 was used as a reference to investigate the mechanical properties of the composited with dead load hardness tester, shore A, at room temperature. Finally, the sound absorption of the samples was carried out using an impedance tube according to ASTM E-1050-98.

3. Results and discussions

3.1 Primary sludge and boiler ash analysis

FTIR spectra of primary sludge and boiler ash are presented in Figure 1. It can be seen that the band at 3000-3500 cm^{-1} indicated the OH group in both samples. This area was wider for the sludge than the boiler ash due to the water content in the sludge being higher. In addition, it was seen that the sludge had a band at 2850.31 cm^{-1} that corresponded to H-C-H stretching (alkyl, aliphatic). The OH fiber (water absorption) was seen at 1623.65 cm^{-1}, and the vibrations of HCH and OCH bonds (methyl groups) appeared at 1515.47 cm^{-1}. In the sludge, there was also a peak at 706.52 cm^{-1} attributed to crystalline 1β polymorph cellulose from pulp [17]. In boiler ash aromatic ring, C=O stretching on the ketones, C=C stretching of hemicellulose, and C=C stretching of vinylidene alkenes appeared at 777.60 cm^{-1}, 1621.90 cm^{-1}, 1064.25 cm^{-1}, and 790.10 cm^{-1}, respectively [18].

Thermal analysis of boiler ash and primary sludge was carried out using DSC. The DSC curve for the samples is shown in Figure 2. From Figure 2, boiler ash and primary sludge has a glass transition temperature (T_g) at 110°C and 92°C, respectively. High T_g in boiler ash was probably the material been heated at high temperature and only a small amount of water was produced, as seen in the FTIR data.

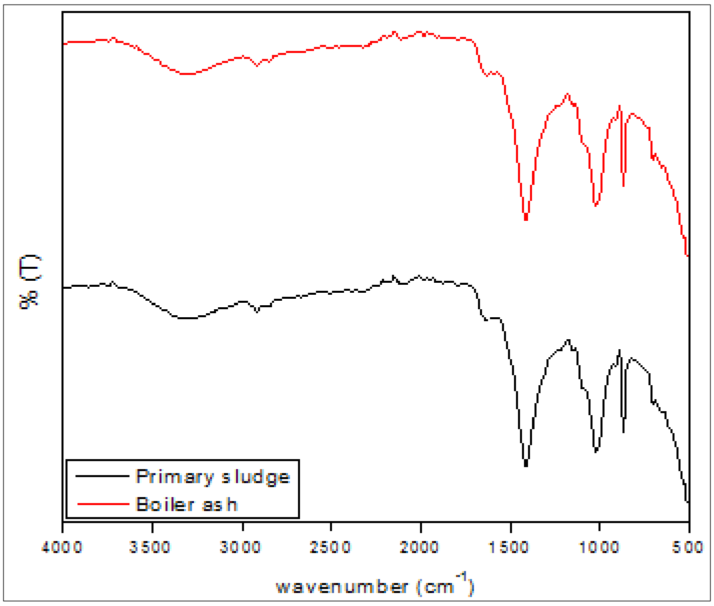

Figure 1. FTIR spectra of primary sludge and boiler ash

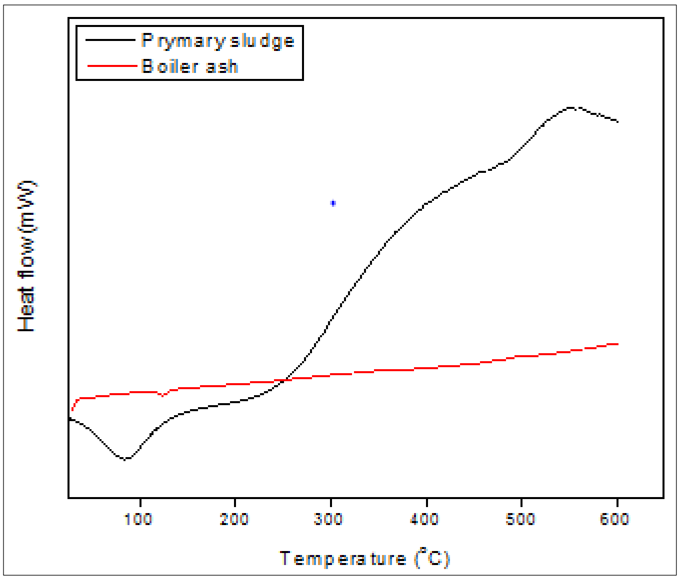

Figure 2. DSC thermogram for primary sludge and boiler ash

3.2. FTIR and x-ray analysis

FTIR spectra and XRD patterns for PS composite were shown in Figures 3 and 4. For PS1, there was a peak in the area of 1492.25 cm^{-1} to indicate the absorption of the (C-H) group as alkyl. For the bond in benzene, there was an absorption 1600.63 cm^{-1} with strong to weak intensity. The peak of 2922.36 cm^{-1} attributed to C-H bonds with benzene, and the band at 3059.46 cm^{-1} corresponded to (Ar-H) bond absorption [19]. Additionally, PS composite foam after the addition of polyurethane showed a decrease in the band 3059.46 cm^{-1}. PS characteristics at 1600.63 cm^{-1} also looked sharper. From the FTIR results, it can be concluded that PS, boiler ash, sludge, and polyurethane have been physically blended ant there was an interaction between hydrogen groups of the filler and polar maleic anhydride group during the mixing process as reported in prevision literature [20].

Regarding Figure 4, PS1 has $2\theta = 15.707^o$ indicating that the polymer was amorphous. For all samples after the presence of maleic anhydride, sludge, and boiler ash, it was observed that there was a new diffraction peak in the $2\theta = 21\text{-}21^o$ to indicate the crystalline region. The diffraction peak becomes higher with the presence of polyurethane as reported by literature [21].

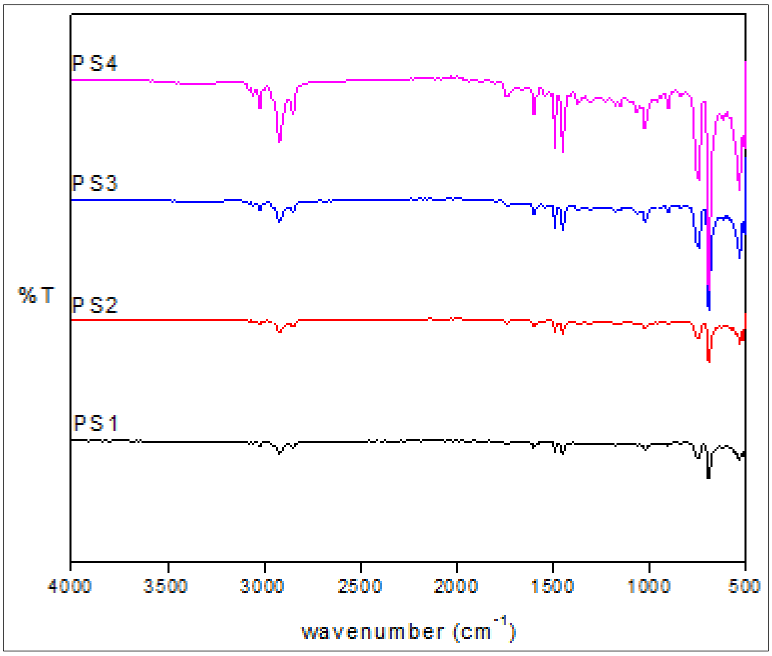

Figure 3. FTIR spectra of PS1, PS2, PS3, and PS4

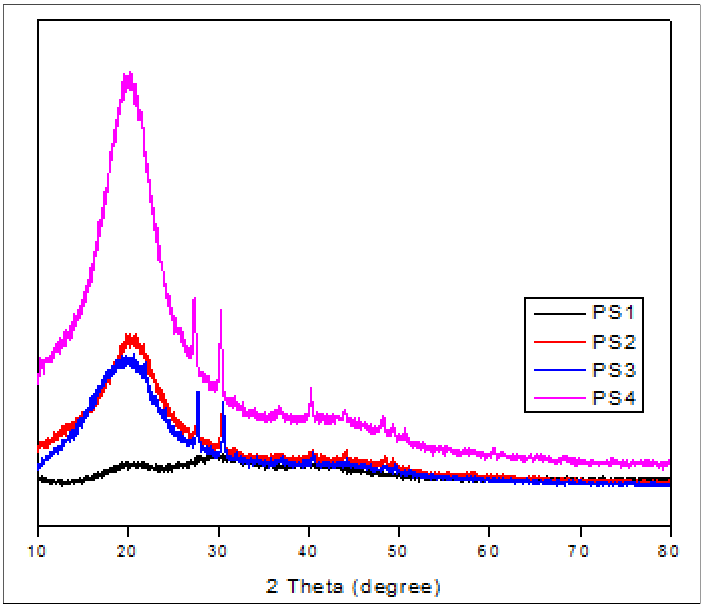

Figure 4. XRD pattern for PS1, PS2, PS3, and PS4

3.3. Morphological characterization

SEM images of PS2, PS3, and PS4 are presented in Figure 5. Before analyzing, the samples were dried in a vacuum oven at 60°C for 5 h to remove moisture from the samples.

From Figure 5, PS with the addition of maleic anhydride, sludge, and boiler ash had several pores with relatively small sizes. However, the fillers were well distributed and dispersed into the polymer matrix and no agglomeration was produced. Morphological analysis for PS3 and PS4 (after the addition of polyurethane) showed a slight difference in surface morphology. PS3 has a rough morphology, while PS4 had a solid surface and no pores produced.

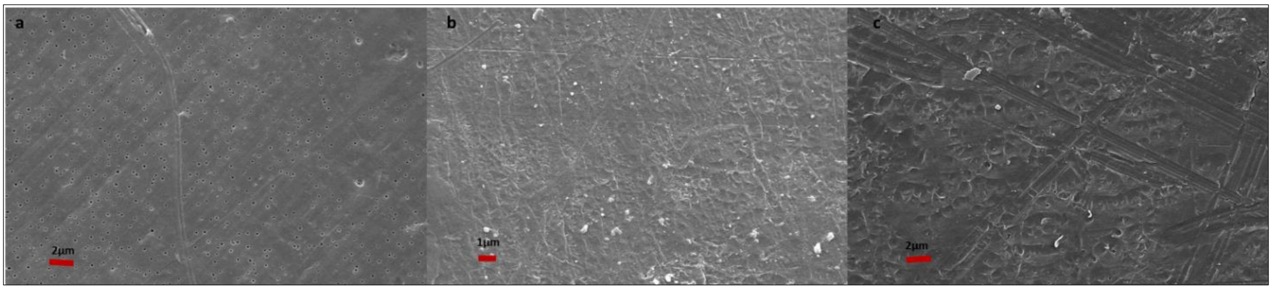

Figure 5. SEM micrograph of: (a) PS2, (b) PS3, (c) and PS4

3.4. Thermal analysis

DSC curve (Figure 6) shows that PS 1 had a T_m 430.79°C. While PS2 and PS4 shifted to 427.13°C and 397.77°C respectively. The shift in the temperature was due to physical interactions between PS, maleic anhydride, sludge, boiler ash, and polyurethane as described in the FTIR data. All samples had three decomposition regions from the TGA analysis (Figure 7). The first decomposition occurred at about 100°C with a mass loss of 2% for PS1 and almost 10% for PS2, PS3, and PS4. The second was occurred at around 200-300°C for PS1, PS2, and PS4, however, PS3 was lower at 131-371°C. At this stage, all samples lost mass by almost 20%. Finally, the decomposition was seen at 349-434°C for PS1 with a residual percent of 0.6%. This decomposition shifted to a higher temperature with a larger percentage of residue for PS2 at 371-442°C and 9.7%, PS3 and PS4 were at 385-437°C with a residual percent of 12.1% and 15%, respectively.

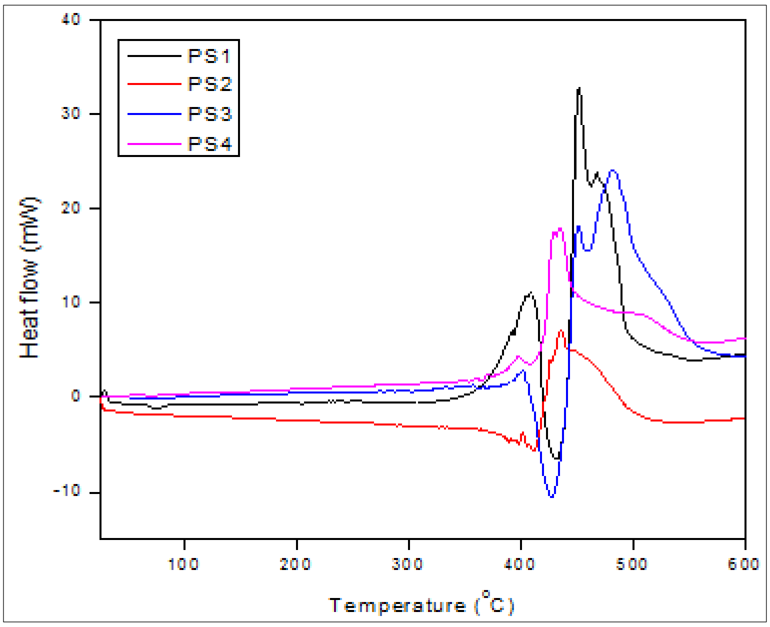

Figure 6. DSC graph of PS1, PS2, PS3, and PS4

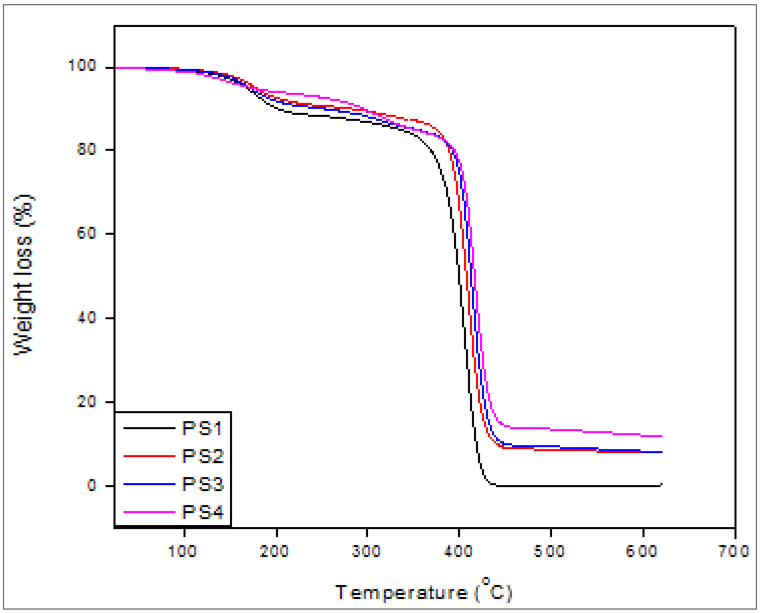

Figure 7. TGA curve of PS1, PS2, PS3, and PS4

3.5. Mechanical properties

The tensile test of PS1, PS2, PS3, and PS4 is presented in Figure 8. In general, the tensile strength for a composite significantly increased with the presence of filler inappropriate percentage and good dispersibility [22]. In this study, PS1 has a tensile 6.526 MPa, after blending with maleic anhydride, sludge and boiler ash (PS2) was 5.670 MPa. A previous study reported that panel added sludge increased the tensile force by 0.61 N/mm^2 compared without sludge 0.59 N/mm^2. This was related to the panel's density value, which was better with the addition of sludge (0.9713 g/cm^3) than without sludge (1.093 g/cm^3). The thickness of the panel also decreased by 66% after the addition of sludge into the wall panel [23]. PS3 and PS4 with the composition of polyurethane 5 and 10 respectively had a tensile 4.499 MPa and 5.765 MPa indicating that higher polyurethane increased the mechanical properties of the composite foam. It is also reported that the higher peak stress, the better energy absorption performance [24]. Furthermore, the increase in the crystallite value for PS composites contributed to the resulting composites' mechanical properties, as shown in the XRD result.

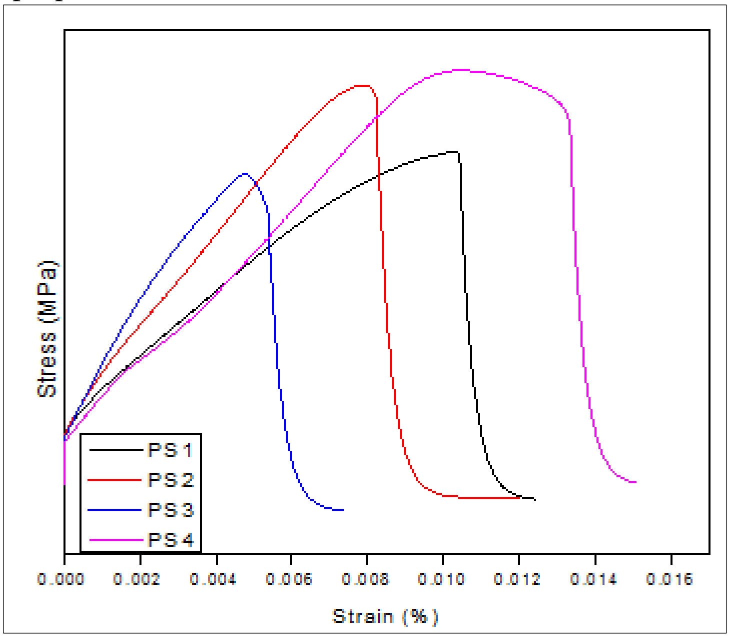

Figure 8. Tensile test of PS1, PS2, PS3, and PS4

MATERIALE PLASTICE
https://revmaterialeplastice.ro
https://doi.org/10.37358/Mat.Plast.1964

3.6. Sound absorption properties

Specimens of sound-absorbing composite foam were prepared with a size of 9.8 cm in diameter and 1 cm in thickness. The sound absorption coefficient of PS composite foam PS before and after adding polyurethane with a frequency range between 200-1200 Hz can be seen in Figure 9 below.

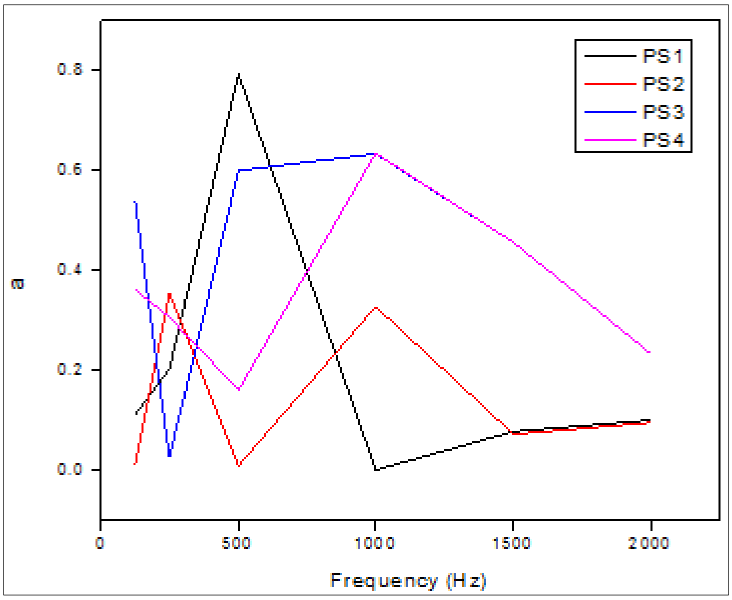

Figure 9. Sound absorption of PS1, PS2, PS3, and PS4

In Figure 9, the absorption coefficient of PS3 and PS4 decreased in the range frequency from 200 Hz to 400 Hz. Meanwhile, PS1 and PS2 increased in the absorption coefficient with the highest value for PS1 0.793. Sound-absorbing composite foam PS3 experienced a significant increase from 250 Hz to 1000 Hz, while PS4 had an increase from 500 Hz to 1000 Hz. The different coefficient value was due to the content of cellulose in which sludge and boiler ash had a function for sound absorption. The presence of polyurethane, sludge, and boiler ash increased the sound absorption of composite foam. Therefore, by-products of pulp industry can be used as a filler for sound insulation material. Furthermore, the sound-absorbing composite foam produced has complied with ISO 11654:1997 standards regarding the sound absorption coefficient level rating on materials for rooms with sound absorption classes D and C with the value of a 0.328-0.793. Meanwhile, a previous study reported that corn cob granules, shredded sunflower stalk, and balls made of sheep wool were qualified as an acoustic barrier according to ISO 10534-1998 [2].

4. Conclusions

Primary sludge and boiler ash from the pulp mill have a particle size distribution in the range 0.1-110 μm in which primary sludge produced still retains its 1β structure. The sound-absorbing composite foam based on PS produced by the reflux reactor method is eligible to be applied as a sound absorber. The chemical reaction between PS, sludge, boiler ash, maleic anhydride, and polyurethane can be seen by the chemical bonds formed during the mixing process as shown in the FTIR results where the peak expansion occurs at a wavenumber of 1600.44 cm^{-1} which indicates the presence of C=C functional group of maleic anhydrides with polymer and filler. The sound-absorbing composite foam produced has complied ISO 11654:1997 standard regarding the rating level for the sound absorption coefficient on materials for rooms with sound absorption classes D and C where the value of α 0.328-0.793.

Acknowledgments: The authors thank the Indonesian Ministry of Industry for research funded from DRPM 2021 PDUPT scheme with contract number 235/UNS.2.3.2/PPM/KP-DRPM/2021.

References

1.ALAM, P., AHMAD, K., AFSAR, S. S., AKHTAR, N., 3D Noise Mapping for Preselected Locations of Urban Area with and without Noise Barriers: A Case Study of Delhi, India, *Noise Mapp*., 7 (1), 2020, 74–83.
2.OANCEA, I., BUJOREANU, C., BUDESCU, M., BENCHEA, M., GRĂDINARU, C. M., Considerations on Sound Absorption Coefficient of Sustainable Concrete with Different Waste Replacements, *J. Clean. Prod*., 203, 2018, 301–312.
3.MALAWADE, U. A., MALGAONKAR, V. V., BADADHE, A. M., JADHAV, M. G., Utilization and Study of Raw Groundnut Shell as a Source of Sound Absorbing Material, J. *Nat. Fibers*., 00 (00), 2021, 1–13.
4.ZHENG, L., AOURAGHE, M. A., ZHANG, K., XU, F., Ultra-Light 3D Fabric Reinforced Composite with Distinct Thermal Insulation and Superior Sound-Absorbing Properties, *J. Phys. Conf. Ser*., 1790 (1), 2021,00.
5.KAYA, A., KAR, F., Properties of Concrete Containing Waste Expanded Polystyrene and Natural Resin, *Constr. Build. Mater*., 105, 2015, 572–578.
6.ISAAC, C. W., EZEKWEM, C., A Review of the Crashworthiness Performance of Energy Absorbing Composite Structure within the Context of Materials, Manufacturing and Maintenance for Sustainability, *Compos. Struct.*, 257, 2021, 113081.
7.BICER, A., KAR, F., Thermal and Mechanical Properties of Gypsum Plaster Mixed with Expanded Polystyrene and Tragacanth, *Therm. Sci. Eng. Prog.*, 1, 2017, 59–65.
8.LU, J., WANG, D., JIANG, P., ZHANG, S., CHEN, Z., BOURBIGOT, S., FONTAINE, G., WEI, M., Design of Fire Resistant, Sound-Absorbing and Thermal-Insulated Expandable Polystyrene Based Lightweight Particleboard Composites, *Constr. Build. Mater.*, (April), 2021, 305
9.BOUZIT, S., MERLI, F., SONEBI, M., BURATTI, C., TAHA, M., Gypsum-Plasters Mixed with Polystyrene Balls for Building Insulation: Experimental Characterization and Energy Performance, *Constr. Build. Mater*., 283, 2021, 122625.
10.SITORUS, R., WIRJOSENTONO, B., TAMRIN., SIREGAR, A. H., NASUTION, D. A., Characteristics of Maleic Anhydride-Modified Polystyrene and Natural Rubber Blends Containing “Talang” Bamboo Powder as Sound Damping Material, *AIP Conf. Proc.*, 2020, 2267.
11.GOURLAY, E., GLÉ, P., MARCEAU, S., FOY, C., MOSCARDELLI, S., Effect of Water Content on the Acoustical and Thermal Properties of Hemp Concretes, *Constr. Build. Mater.*, 139, 2016, 513-523.
12.MADUSHIKA, J. W. A., LANAROLLE, W. D. G., A Review on Novel Approaches to Enhance Sound Absorbing Performance Using Textile Fibers, *J. Text. Inst*., 0 (0), 2021, 1–8.
13.BELAYACHI, N., HOXHA, D., SLAIMIA, M., Impact of Accelerated Climatic Aging on the Behavior of Gypsum Plaster-Straw Material for Building Thermal Insulation, *Constr. Build. Mater.*, 125, 2016, 912-918.
14.DURBACA, I., SPOREA, N., VASILE, O., Assessment of the Acoustic Absorption Characteristics of Layered Composite Structures Obtained from Plates with Lignocellulosic Coatings (I), *Mater. Plast.*, 57 (2), 2020 8-14.
15.KHALIL, A. A., TAWFIK, A., HEGAZY, A. A., EL-SHAHAT, M. F., Effect of Some Waste Additives on the Physical and Mechanical Properties of Gypsum Plaster Composites, *Constr. Build. Mater.*, 68, 2014, 580-586.
16.HIDAYATI, T. R., WIRJOSENTONO, B., YUNUS, D., TAMRIN., PELITA, E., RAHMAD, D., MIRANDAULIA, M., RUSMAR, I., The Effect of Sludge from Waste Paper Industry Additon as a Filler into Composite Panel Based on Polypropylene Plastic Waste and Cocofiber, *AIP Conf. Proc.*, 2342, 2021, 090003.

17.RISNASARI, I., FEBRIANTO, F., WISTARA, N. J., SADIYO, S., NIKMATIN, S., TERAMOTO, Y., LEE, S. H., JANG, J. H., HIDAYAT, W., KIM, N. H., Characterization of Cellulose Nanocrystal with Cellulose II Polymorph from Primary Sludge and Its Application to PVA Nanocomposites, *Wood Sci. Technol*., 52 (2), 2018, 555–565.
18.REZA, M. S., AFROZE, S., BAKAR, M. S., SAIDUR, R., ASLFATTAHI, N., TAWEEKUN, J., AZAD, A. K., Biochar Characterization of Invasive Pennisetum Purpureum Grass: Effect of Pyrolysis Temperature, *Biochar.*, May. 2020,
19.HUAN, S., BAI, L., LIU, G., CHENG, W., HAN, G., Electrospun Nanofibrous Composites of Polystyrene and Cellulose Nanocrystals: Manufacture and Characterization, *RSC Adv.*, 5 (63), 2015, 215.
20.QIAO, X., ZHANG, Y., ZHANG, Y., Maleic Anhydride Grafted Polypropylene as a Coupling Agent for Polypropylene Composites Filled with Ink-Eliminated Waste Paper Sludge Flour, *J. Appl. Polym. Sci.*, 91 (4), 2004, 2320-2325.
21.WANG, C., CHUNFENG, M., CHANGDAO, M., WEI, L., Tailor-made Zwitterionic Polyuretahne Coatings: Microstructure, Mechanical Property and their Antimicrobial Performance, *RSC Adv*., 7, 2017, 27522.
22.MAHDI, E., DEAN, A., The Effect of Filler Content on the Tensile Behavior of Polypropylene/ Cotton Fiber and Poly(Vinyl Chloride)/Cotton Fiber Composites, *Materials (Basel).*, 13 (3), 2020,
23.HIDAYANI, T. R., HARMILENI., MIRNANDAULIA, M., YUNIARTI, T., SILVANY, R., Comparison the Using of Polypropylene and Low-Density Polyethylene Plastic Waste as a Matrix in Making Biodegradable Plastics and Starch from Empty Fruit Bunch of Palm Oil as a Filler, *In IOP Conference Series: Earth and Environmental Science*., Vol. 765, 2021, 1-6.
24.CHEN, Y., YUAN, F., SU, Q., YU, C., ZHANG, K., LUO, P. P., HU, D., GUO, Y., A Novel Sound Absorbing Material Comprising Discarded Luffa Scraps and Polyester Fibers, *J. Clean. Prod*., 245 (October), 2020, 118917

Manuscript received: 10.01.2022