SYNTHESIS OF POROUS CARBON MATERIAL PROPYLENE MEDICAL MASK AS ADSORBENT HEAVY METAL IRON (Fe) IN WATER EX-TIN MINING BANGKA BELITUNG

Addela Amelia, Faizah Alivia Putri, Widia, Della Adelia, Alkasiyah, Verry Andre Fabiani*
Department of Chemistry, Faculty of Science and Engineering, University of Bangka Belitung, Jalan Kampus Terpadu Universitas Bangka Belitung, Bangka 33172, Bangka Belitung Province, Indonesia
*Email: verry-andre@ubb.ac.id

Received 19 October 2023
Accepted 25 April 2024

Abstract

Bangka Belitung Islands Province faces severe environmental challenges, exacerbated by illegal tin mining activities leading to heightened pollution levels, particularly in stagnant water within former tin mining holes, reaching depths of up to 40 meters. The presence of iron metal (Fe) in this water poses a health risk, potentially damaging intestinal walls and compromising lung function upon ingestion. This study presents a novel approach to address this issue by synthesizing porous carbon material from propylene waste sourced from medical masks. The process involves initial sulfonation for sterilization and pore size enhancement, followed by activation using KOH and carbonization at 750 °C. Characterization methods validate the successful synthesis, including FTIR highlighting polypropylene groups at 822 cm^{-1}, XRD indicating graphite carbon with high crystallinity, and SEM exhibiting rod shapes and cavities at 5,000× magnification. The adsorption test demonstrates outstanding performance, with porous carbon exhibiting a 100% adsorption efficiency in purifying water, eliminating odors, and reducing iron (Fe) levels. This innovative method effectively reduces iron levels in stagnant water environments, providing a sustainable solution to environmental pollution, particularly in Bangka Belitung.

Keywords: *adsorbent, iron (Fe), polypropylene, stagnant water, sulfonation*

Introduction

The Province of Bangka Belitung Islands faces significant environmental challenges, primarily stemming from the proliferation of illegal tin mining activities. The surge in these activities has led to alarming levels of environmental pollution. According to data from IKPHLD (2021), 12,607 ex-tin mining lakes are spread across the region. This number is on the rise, underscoring the urgent need for proper management of mining activities. The environmental degradation in Bangka Belitung is predominantly attributed to elevated concentrations of heavy metal pollutants in these lakes, causing harm to the surrounding ecosystem. Meyzilia's research in 2018 revealed that iron (Fe) was the predominant heavy metal, with excessive doses posing risks such as damage to the intestinal walls and lung alveoli (Murraya *et al*., 2018).

The term "air kolong" in Bangka refers to stagnant water in former tin mining holes created through hydraulic mining, reaching depths of up to 40 meters (Yusuf, 2011). Effectively addressing water resources from these abandoned mines involves mitigating or neutralizing the metal content. Prior studies have identified porous activated carbon adsorbent materials as effective due to their extensive surface area, high porosity, and substantial adsorption capacity. The abundance of raw materials for porous

carbon, such as polymer waste, presents significant research opportunities.

Medical mask waste, a prevalent form of polymer waste, is challenging to decompose in nature and is typically either incinerated or landfilled. Indonesia, as indicated by Ministry of Environment and Forestry data (2021), generates a substantial amount of medical waste, and this figure is expected to rise without proper waste management. Carbonization of polymer waste, especially polypropylene from medical masks, has proven to be an effective processing method (Min *et al*., 2019). Polypropylene boasts an 85.7% carbon content by weight, making it a promising source for carbon-based nanomaterials (Fitria *et al*., 2022). Given that carbon is a primary component of polymer materials, various carbon forms, such as activated carbon, carbon fiber, carbon balls, graphite, and nano carbon, can be produced through carbonization (Chen *et al*., 2020). Previous studies have demonstrated the efficacy of porous carbon derived from polypropylene waste in adsorbing substances like methylene blue dye (Li *et al*., 2022) and heavy metal ions (Sharma *et al*., 2021).

Building on this existing body of research, there is a pressing need for further investigation into synthesizing porous carbon from medical mask polypropylene waste as an adsorbent for iron metal (Fe) in water used in tin mining in Bangka Belitung. The objective of creating such material is to mitigate iron (Fe) levels in mining water, considering its toxic properties to the human body. Given that poisonous substances can enter the body through inhalation, ingestion, and skin contact, reducing or eliminating heavy metal Fe levels in mining water is crucial for environmental sustainability and ensuring the availability of clean water in Bangka Belitung.

Research Methods

Materials

The materials utilized in the research comprised medical mask waste, sulfuric acid (H_2SO_4) 98% (Merck Pro Analysis), potassium hydroxide (KOH) (Merck Pro Analysis), deionized water, distilled water, Polyvinyl Alcohol (PVA) (Merck Pro Analysis), hydrochloric acid (HCl) 1 M (Merck Pro Analysis), bleach, and detergent.

Instrumentation

The tools utilized included a Teflon vessel autoclave (TEP-70067-01085), beaker (Iwaki), Erlenmeyer flask (Pyrex), analytical balance (ACZET CG 2002L), measuring cup (Iwaki), dropper pipette, oven (Memmert UN 55 53L), tubular furnace (Thermo Scientific AAF 3/11/PID 301), Scanning Electron Microscope (SEM) TM 3000 (Hitachi with SwiftED 3000 X-Ray Microanalysis), Fourier Transform Infrared Spectroscopy (FTIR) (PerkinElmer Frontier), X-ray Diffraction (XRD) (PANalitycal MPD PW3030/60 type X'Pert PRO), and Atomic Absorption Spectrophotometer (AAS) (Shimadzu type AA 600).

Procedure

The methods to be used in this research are sulfonation and carbonization. Broadly speaking, this research includes the preparation of mask waste raw materials, sulfonation, activation, carbonization, adsorption tests on tin mining water, water and ash content tests, and porous carbon synthesis.

1) Preparation of medical mask waste

   The decontamination process for medical mask waste involves soaking it in a water and detergent mixture for 3–4 hours to eliminate attached bacteria and viruses. Subsequently, the waste is dried, cut into the smallest possible size, washed four times with water, and then dried again, as

outlined in the methodology (Victory *et al* 2021).

2) Sulfonation of sample

For the sulfonation of samples, 0.5 grams of the waste is weighed and placed in a 150 mL Erlenmeyer flask with 10 mL of sulfuric acid (H_2SO_4). The flask is then transferred to a Teflon vessel autoclave and heated using the autoclave at a temperature of 110 °C for 12 hours. This process aims to sterilize medical mask waste that may be contaminated with viruses and bacteria through exposure to high temperatures, aligning with the methodology proposed. The sulfonated samples were then washed with deionized water five times to remove solvent residues and dried in an oven at 70 °C for 12 hours (Hu and Lin 2021).

3) Activation and carbonization

The dried samples are immersed in a KOH solution with varying ratios (1:2; 1:3; 1:4) and subsequently undergo carbonization (pyrolysis) through combustion at 750 °C for 2 hours using a furnace. This process aims to eliminate water content and other unwanted materials in mask waste, such as hydrogen and oxygen, or volatile materials (Ridhuan and Suranto, 2016). The samples, now transformed into black (carbon), are cooled to room temperature, washed with a 1 M HCl solution, and deionized water for five cycles until the pH reaches around 7. Subsequently, they are dried in an oven at 70 °C for 12 hours (Silalahi and Hendrasarie, 2021).

4) Characterization of porous carbon

The material's morphology is examined using a scanning electron microscope (SEM). The chemical structure is analyzed through Fourier-transform infrared spectroscopy (FTIR), and the crystal pattern is assessed using X-ray diffraction (XRD) (Hu and Lin, 2021).

5) Heavy metal adsorption test on stagnant water

A 100 mL sample of stagnant water is taken and added to an Erlenmeyer flask containing 10 grams of activated carbon. The shaker is set at 30 rpm. The mixture is stirred for 30 minutes, left to stand at room temperature for 3 hours, and then filtered. The obtained filtrate is measured using atomic absorption spectrophotometry (AAS) (Kurnia, 2023).

6) Water content determination

A quantity of 1 gram of carbon is weighed and placed in a known-weight porcelain dish (sample weight denoted as W1). Subsequently, the porcelain dish containing the sample is placed in an oven at 100–110 °C for 2 hours. Afterward, the sample is cooled in a desiccator and weighed until a constant weight is achieved (expressed as W2) (Ramayana *et al*., 2017). The water content can be calculated by Eq. (1).

$$\text{Water content} = \frac{W1-W2}{W1} \times 100\% \quad (1)$$

7) Ash content determination

A quantity of 1 gram of carbon is weighed and placed in a known-weight platinum dish (sample weight denoted as W1). Subsequently, the platinum dish containing the sample is placed in a furnace at 800–900 °C for 3 hours. After cooling in a desiccator, the sample is weighed until a constant weight is achieved (expressed as W2) (Ramayana, *et al*., 2017). The ash content can be determined using Eq. (2).

$$\text{Ash content} = \frac{W1}{W1} \times 100\% \quad (2)$$

8) Adsorption Efficiency

The adsorption efficiency can be calculated by Eq. (3).

$$\%E = \frac{Ci - Ce}{Ci} \times 100\% \qquad (3)$$

Information:

%E = Adsorption efficiency

C_i = Heavy metal content before adsorption

C_e = Heavy metal content after adsorption

Results and Discussion

Preparation of medical mask waste

Collected mask waste is initially separated into rope and wire components. Subsequently, it undergoes a 3–4 hours soak in a water and detergent mixture to eliminate bacteria and viruses attached to the outer surface of the mask. The sanitized waste is then dried in an oven at 160 °C for 30 minutes. Following the drying process, the mask waste is cut into smaller pieces for further use.

Sulfonation of samples

The prepared medical mask waste is subjected to sulfonation in this research to achieve sterilization and eliminate bacteria and viruses on the mask's surface. The utilization of a Teflon vessel autoclave is instrumental in introducing sulfonate groups. This introduction is facilitated by the strong oxidation of concentrated sulfuric acid, which breaks down the carbon chain, allowing for the incorporation of sulfonate groups. These groups play a crucial role in the production of porous carbon, enhancing carbon yield and improving the physical and chemical properties of the resulting materials. During the sulfonation process, polymer chains are broken, leading to the generation of gases such as CO_2, CO, and low molecular weight alkanes (Hu and Lin, 2021).

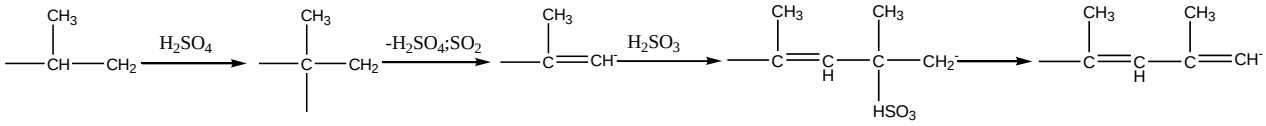


Figure 1. Propylene sulfonation reaction

During the sulfonation stage, a reaction occurs between the medical mask waste, primarily composed of polypropylene and sulfuric acid (Figure 1). This chemical interaction involves the sulfuric acid attacking the surface of the polypropylene fibers, leading to the formation of sulfonate and olefinic unsaturated groups in the initial stage. However, it is important to note that the sulfonic acid group formed in this process is inherently unstable at the reaction temperature of 120 °C. This instability results in the production of by-products such as SO2, H_2O, and olefinics. The loss of sulfonic acid groups, known as desulfonation, occurs, creating C=C double bonds and initiating the carbonization of the polypropylene fibers. As a consequence, isolated C=C double bonds combine, and their sequences become conjugated as the reaction progresses (Hu and Lin, 2021). The culmination of the sulfonation-desulfonation reaction yields carbon fibers with a robust and solid structure.

Activation and carbonization process

The activation process utilizing KOH serves to generate micro-sized pores, enhancing the surface area of the carbon framework and producing a dense pore structure. The subsequent carbonization process, conducted at high temperatures, plays a crucial role in releasing gases (CO, CO_2, H_2O, and SO_2), further augmenting the development of surface pores formed during the activation process, as detailed by Sun *et al*. in 2023. Following the

carbonization process, the sample undergoes washing with a sufficient amount of 1 M HCl and distilled water until reaching pH 7. This stage is conducted to eliminate unnecessary substances. Importantly, the carbonization stage is sequenced after the sulfonation stage to preserve the carbon quantity in the sample, as this phase may result in substantial carbon loss or may not withstand high temperatures. The objective of washing is to ensure the removal of any residual substances. Distinct colors are observed for each sample variation post-carbonization and washing stages with 1 M HCl. Specifically, variations (1:2) and (1:3) exhibit a gray color, while variation (1:4) appears black, classifying it as carbon. This coloration aligns with findings by Said (2007), which assert that carbon is a porous solid characterized by a black color.

Water content and porous ash carbon test

Determination of the quality of propylene porous carbon is carried out according to SNI-06-3730-1995 regarding the requirements and quality of activated carbon. Determination of quality is done in the form of water content test to determine the hygroscopic nature of activated carbon and ash content test to determine the content of metal oxides that are still present in polypropylene activated carbon after the activation process. The results of determining the quality of propylene porous carbon are in Table 1.

Based on the analysis results in Table 1, it was determined that the water content in porous carbon is 1.2%, and the ash content is 2.72%, indicating the presence of residual water and ash particles. A lower water content suggests reduced water within the porous carbon, while a lower ash content signifies diminished residue. Excessive water and ash presence can obstruct carbon pores, leading to a reduced surface area and hindering the adsorption process (Ramayana *et al*., 2017). The obtained analysis results align with the standards outlined in SNI 06-3730-1995.

Table 1. The results of water content and ash content analysis

Factor	Parameter	Results of Analysis
Water Content	Max.15%	1.2%
Ash Content	Max.10%	2.72%

The characteristics of porous carbon

1) The analysis of functional groups using Fourier Transform Infra-Red (FTIR)

Figure 2. The FTIR spectra analysis of carbon variations at ratios 1:2, 1:3, and 1:4 reveals the presence of the S-O-C functional group, wave number of 822 cm^{-1} characteristic of polypropylene, with the sulfinyl group (O=S=O) exhibiting bending vibrations at 1001 cm-1 and 1003 $cm^{-1,}$ and the S=O functional group identified at 1404 cm cm^{-1} and 1048 cm^{-1}. Stretched C=C functional groups representing aromatic ring bonds are evident at 1632 cm^{-1} and 1652 cm^{-1}. The oxidized C=O functional group, a result of the sulfonation process, is observed at 1842 cm^{-1} and 1849 cm^{-1}. Activated carbon, featuring hydrophilic properties due to hydroxyl and carbonyl groups (Efiyanti *et al*., 2020), displays the C=O functional group in the 1800s cm^{-1} range. The alkyne group's presence is indicated by curved C≡C at 2119 cm^{-1} and narrowed C≡C at 2179 cm^{-1}, aligning with research associating C≡C with wave numbers between 2067–2900 cm^{-1}. The O-H functional group, broadening between 3154 cm^{-1} and

3636 cm^{-1}, results from activation using KOH, facilitating metal ion connections with carbon and oxygen on the carbon surface.

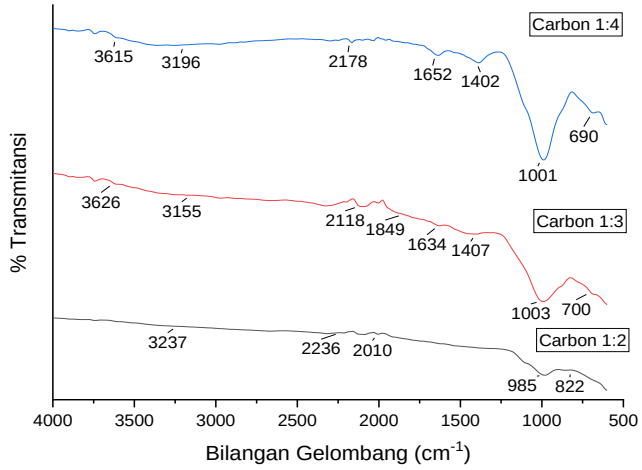


Figure 2. The FTIR spectra of propylene porous carbon

2) Crystallinity analysis using X-ray Diffraction (XRD)

The XRD spectrum analysis of the 1:2 variation sample reveals a distinctive pattern with a rugged and narrow peak at 28.794°, indicative of graphitic carbon with a high crystallinity phase (Figure 3). Additionally, a smooth and widened peak at 50.33° further supports the presence of graphitic carbon in a crystalline phase. Similarly, the 1:3 variation sample exhibits a rugged and narrow peak at 29.355° and a smooth and widened peak at 58.91°, characteristic of graphitic carbon with a crystalline phase. In contrast, the 1:4 variation sample displays a rugged and narrow peak at 20.8° followed by a smooth and wide peak at 40.44°, suggesting a variation of porous activated carbon with an almost amorphous spectrum, as evidenced by the smooth and wide peak indicative of reduced crystallinity.

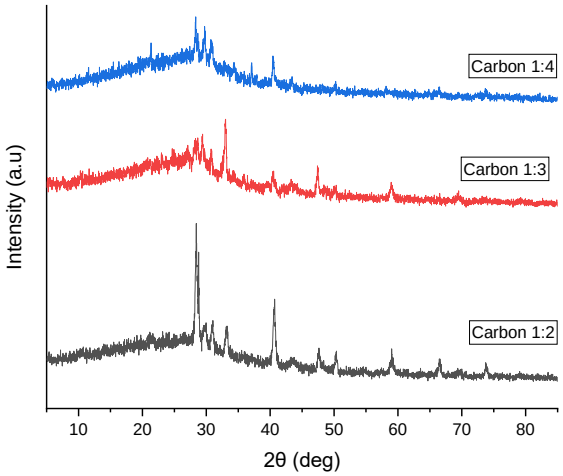
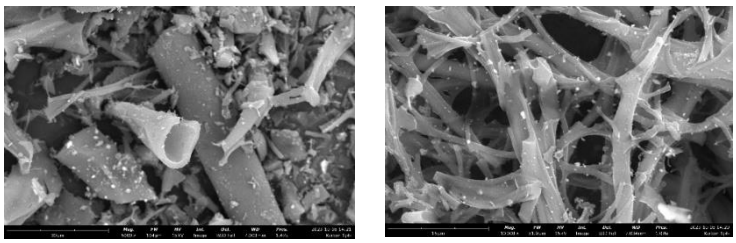


Figure 3. XRD spectra of propylene porous carbon

3) Morphological analysis using Scanning Electron Microscope (SEM)
The morphological analysis of propylene porous carbon reveals distinctive features in the 1:4 sample variation (Figure 4). At 5,000 times magnification, shown in Figure 4a, a solid rod-shaped structure with visible cavities indicates the presence of pores. Meanwhile, 10,000 times magnification displays interconnected coarse fibers (Figure 4b). These findings align with the research conducted by Sun *et al*. (2023) and Laos *et al*. (2016), confirming that porous carbon exhibits a pore surface area within the range of 300–2000 m^2/gr.



(**a**) (**b**)

Figure 4. The results of SEM analysis on propylene porous carbon at a magnification of (a) 5,000× and (b) 10,000×

4) Adsorption test of the heavy metal in the stagnant water and adsorption analysis
The Fe heavy metal content in the water under the basin was measured using atomic absorption spectroscopy. The measurement results are in Table 2. The initial concentration of the heavy metal Fe in stagnant water is recorded at 1,377 mg/L, surpassing the maximum permissible level of 1 mg/L as stipulated by the Regulation of the Minister of Health (PERMENKES) of the Republic of Indonesia Number 32 of 2017 regarding environmental health quality standards and water health requirements. The untreated stagnant water exhibits a noticeable odor and a clear yellow color, categorizing its heavy metal Fe content as high and polluted. Subsequently, through a 30-minute adsorption process involving shaking 0.4 grams of porous activated carbon in a 100 ml sample, the bottom water undergoes a transformation, becoming clear, odorless, and registering a heavy metal content of 0 mg/L. This represents a remarkable adsorption efficiency of 100%. The success of propylene porous activated carbon in adsorbing heavy metal Fe is attributed to its inherent properties (Idrus *et al*., 2013).

Table 2. The results of the adsorption test on the stagnant water

Variation of Porous Carbon	C_i	C_e	Adsorption Efficiency (%E)
1:4	1,377 ppm	0 ppm	100 %

Conclusions

The FTIR characterization results indicate the presence of a characteristic propylene group at a wave number of 822 cm^{-1}, underscoring the porous nature of propylene. Ash and water content tests further affirm the porous structure of carbon, with notable heavy metal content, specifically iron (Fe). While iron is essential for the body, excessive doses can lead to damage to the intestinal walls and lung alveoli. The adsorption test demonstrates exceptional efficacy, achieving 100% adsorption efficiency. This remarkable performance is evidenced by the carbon's ability to purify water, eliminate odors, and absorb iron metal (Fe), particularly in the water under tin in Bangka Belitung. The findings highlight the potential of propylene porous carbon for effective water treatment and heavy metal removal.

References

Badan Standardisasi Nasional., 1995. SNI 06-3730-1995 Tentang Arang Aktif Teknik. Jakarta: Badan Standardisasi Nasional.

Chen, W. M., Luo, M., Yang, K., Zhou, X. Y., 2020. Microwave-assisted KOH activation from lignin into hierarchically porous carbon with super high specific surface area by utilizing the dual roles of inorganic salts: microwave absorber and porogen. *Microporous Mesoporous Mater,* 300:110178.

Efiyanti, L., Wati, S. A. and Maslahat, M., 2020. Pembuatan Dan Analisis Karbon Aktif Dari Cangkang Buah Karet Dengan Proses Kimia Dan Fisika. Jurnal Ilmu Kehutanan, Volume 14, Pp. 94-108.

Fitria, H., Ahmad, T. L. and Rizaq, S. U., 2022. Pemanfaatan Masker Limbah COVID-19 Sebagai Upaya Mengurangi Pencemaran Lingkungan. *Metode Jurnal Teknik Industri*, 8(1), Pp. 41-50.

Hu, X. and Lin, Z., 2021. Transforming Waste Polypropylene Face Masks Into S-Doped Porous Carbon As The Cathode Electrode For Super Capacitors. *Ionics*, Volume 27, Pp. 2169-2179.

Idrus, R., Lapanporo, B. P. and Putra, Y. G., 2013. Pengaruh Suhu Aktivasi terhadap Kualitas Karbon Aktif Berbahan Dasar Tempurung Kelapa. *Jurnal Prisma Fisika*, Volume 1, pp. 50-55.

Kurnia, A., 2023. Identifikasi Logam Berat Pada Air Kolong Dan Mikroba Potensial Untuk Bioremediasi Di Lahan Pasca Penambangan Timah. *Jurnal Geominerba*, 8(1), pp. 36-43.

Laos, L. E., Masturi and Yulianti, I., 2016. Pengaruh Suhu Aktivasi Terhadap Daya Serap Karbon Aktif Kulit Kemiri. *Snf*, Volume 5.

Min, J. K., Zhang, S., Li, J, X., Klingeler, R., Wen, X., Chen, X. C., Zhao, X., Tang, T., Mijowska, E., 2019. From polystyrene waste to porous carbon flake and potential application in supercapacitor. *Waste Manag*, 85:333–340.

Meyzilia, A., 2018. Pemanfaatan Air Kolong Bekas Tambang Timah Sebagai Penambah Sumber Air Tanah Menggunakan Lubang Kompos Di Bangka Belitung. *Jurnal Pendidikan Ilmu Sosial*, 27(1), Pp. 22-30.

Murraya, Taufiq-Spj, N. and Supriyantini, E., 2018. Kandungan Logam Berat Besi (Fe) Dalam Air, Sedimen Dan Kerang Hijau (Perna Viridis) Di Perairan Trimulyo, Semarang. *Journal Of Marine Research*, 7(2), Pp. 133-140.

Permenkes RI No 32 tahun 2017. Peraturan Kesehatan RI Nomor 32 tahun 2017 tentang Standar Baku Mutu Kesehatan Lingkungan dan Persyaratan Kesehatan Air Untuk Keperluan Higiene Samitasi, Kolam Renang, Solus Per Aqua, dan Pemandian Umum.

Ramayana, D., Royani, I. and Arsyad, F. S., 2017. Pembuatan Carbon Black Berbasis Nanoserbuk Tempurung Biji Karet Menggunakan High Energy Milling. *Jurnal MIPA*, 40(1), Pp. 28-32.

Ridhuan, K. and Suranto, J., 2016. Perbandingan Pembakaran Pirolisis Dan Karbonisasi Pada Biomassa Kulit Durian Terhadap Nilai Kalori. *Jurnal Teknik Mesin (TURBO)*, 5(1), Pp. 50-56.

Said, N. I., 2007. Pengolahan Air Minum dengan Karbon Aktif Bubuk (Prinsip Dasar Perhitungan, Perencanaan Sistem Pembubuhan dan Kriteria Disain). JAI, 3(2), pp. 96-110.

Sharma, M. D., and Krupadam, R. J., 2021. Adsorption-desorption dynamis of synthetic and naturally weathered mirofibers with toxic heavy metals and their ecological risk in an estuarine ecosystem, 112198.

Silalahi, A. S. and Hendrasarie, N., 2021. Pemanfaatan Limbah Masker Bedah 3 Limbah Plastik Polyetilen Sebagai Adsorben Untuk Menurunkan Kandungan Deterjen Dan Fosfat Pada Limbah Industry Laundry. *Teknik Lingkungan*, 2(1), Pp. 51-59.

Sun, Q., Liu, T., Wen, T. and Yu, J., 2023. Porous Carbon Tubes from Recycling Waste COVID-19 Mask for Optimization Of 8 Mol% Y_2O_3-Doped Tetragonal Zirconia Polycrystaline Nanopowder. *Materials Today Chemistry*, Volume 30, Pp. 1-12.

Victory, S. *et al*., 2021. The Utilization of Medical Mask Waste as A High-Quality Nanofiber Material: A Review. *Indonesian Journal of Chemical Science*, 10(2), Pp. 89-94.

Yusuf, M., 2011. Model Pengembangan Kolong Terpadu Pasca Penambangan Timah di Wilayah Bangka Belitung. Jurnal Makalah Ilmiah Sriwijaya, 18(11), pp. 669-681.