ENHANCED KNOWLEDGE IN SCIENCES AND TECHNOLOGY
e-ISSN: 2773-6385

EKST

Vol. 4 No. 1 (2024) 229-237
https://publisher.uthm.edu.my/periodicals/index.php/ekst

Geopolymer Material with Coconut Husk Additive as Heavy Metal Absorbances for Malacca River Water

Gokkul Pillay V Tamilvenan[1], Zaidi Embong[2]*

[1] *Department of Physics, Faculty of Applied Sciences and Technology, Universiti Tun Hussien Onn Malaysia, 86400 Pagoh, Johor, Malaysia.*

[2] *Research Centre for Soft Soil (RECESS) Institute Integrated Engineering (I2E) Universiti Tun Hussein Onn Malaysia (UTHM) 86400 Parit Raja, Batu Pahat, Malaysia.*

*Corresponding Author: zaidi@uthm.edu.my
DOI: https://doi.org/10.30880/ekst.2024.04.01.026

Article Info

Received: 27 December 2023
Accepted: 15 January 2024
Available online: 27 July 2024

Keywords

Geopolymer Material, Coconut Husk, River Water, Heavy Metal Absorbance, ICPMS, SEM-EDX

Abstract

The Malacca River, a vital water resource, faces a critical challenge posed by escalating high level of nickel and other heavy metals pollution. As the adverse effects of this contamination on both aquatic ecosystems and human health become increasingly evident. The research objectives aimed to validate the best geopolymer pellet for polluted river water remediation. Hydrothermal method was employed for synthesis process of geopolymer and curing process was employed to form concrete geopolymer pellets. SEM/EDX and ICPMS characterization employed, to analyse the pore size and heavy metal absorption capacities of the geopolymer pellets. Water samples from the Malacca River were initially analysed to establish baseline of heavy metal concentrations. Subsequently, geopolymer pellets with varied compositions were immersed in the water samples to analyse heavy metal content before and after immersion using ICPMS. This research used Inductively coupled plasma mass spectrometry (ICP-MS) to detect the presence and concentration of selected heavy metal. Findings revealed distinct variations in heavy metal absorption capacities among different geopolymer pellets, with promising results in reducing contamination levels. The successful synthesis of a geopolymer material, enhanced with coconut husk, underscores its potential as an eco-friendly remedy for mitigating heavy metal contamination. The results show significant removal efficiency of nickel heavy metal. The best geopolymer pellet is the geopolymer pellet with addition of 3% coconut husk which demonstrated the highest removal efficiency, indicating enhanced porosity from coconut husk addition facilitating heavy metal absorption by the geopolymer pellet.

1. Introduction

The issue of water pollution caused by heavy metals has become a noteworthy worldwide concern, with widespread consequences for human health and environmental balance. Industrial activities, mining operations, and improper waste disposal frequently emit toxic pollutants, specifically heavy metals like lead, mercury, cadmium, and arsenic, into the environment [1]. Persisted presence of hazardous compounds in water bodies can lead to extended exposure, which can have considerable health effects when consumed [2]. Numerous papers exist referring to the harmful effects of heavy metal exposure on human health. The consumption of contaminated food or drinking water that has high levels of heavy metal concentrations can lead to many health problems including liver malfunction, neurological disorders, development abnormalities, renal damage, and an increased risk of cancer. Heavy metals can cause detrimental influences on vulnerable groups, such as youngsters and pregnant women [3].

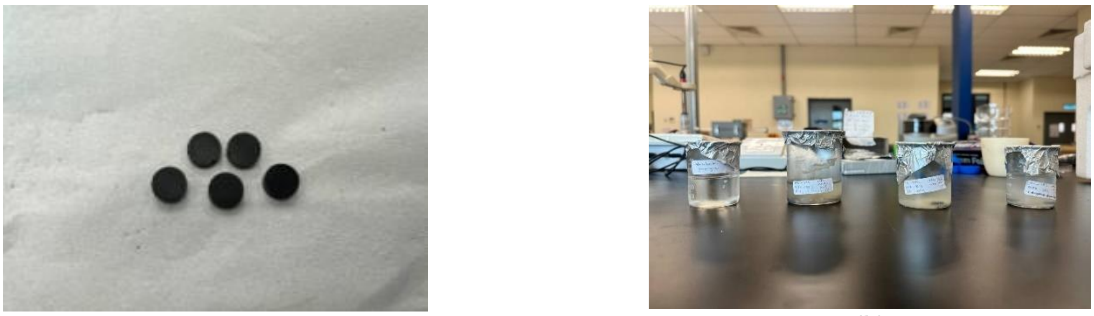
Conventional methods of water purifications, possibly will result in high expenses, require substantial energy usage, and cause detrimental effects on the environment. Thus, it is priority to develop alternative approaches that are both environmental and efficient in mitigating the risks correlated with heavy metal pollution [4]. The main aim of this study is to examine and assess the effectiveness of using geopolymer-coconut husk composites as a material that attracts and eliminates heavy metals from polluted water [5]. The objective is to validate the best geopolymer pellet for polluted river water remediation. This will be accomplished by utilizing coconut husk, an abundant agricultural byproduct, in combination with geopolymer, an environmental and eco-friendly substance [6,7].

The study aims to promote the development of ecologically conscious and enduring solutions to water pollution and to improve water quality. The reason for including coconut husk in this situation is due to its natural ability to absorb and store heavy metals [8]. The fibrous composition of the coconut husk consists of cellulose, lignin, and hemicellulose molecules that naturally possess the ability to adsorb heavy metals. Through the utilisation of inherent characteristics in conjunction with cutting-edge geopolymer methodology, the objective is to produce a new substance with the ability to effectively diminish high levels of heavy metals present in contaminated bodies of water [9,10].

2. Materials and Methods

2.1 Sample Preparation

Prior to sample preparation water samples were collected from Melaka River situated near Kampung Mortem, map location of water sample collection shown in Fig. 2. The raw materials Palm Oil Fuel Ash (POFA), Metakaolin (MK), and Coconut Husk (CH) underwent transformation into powder through ball milling process and sieving process using 150-micron meter sieve. The preparation of alkaline activator involves the following methodical steps. Initially, the alkaline activator is prepared with precise molar ratios of sodium silicate and sodium hydroxide, utilizing a 10 M NaOH solution and 1 M sodium silicate solution and addition of distilled water. These alkaline activator and raw materials were meticulously combined using mechanical stirring, the mixing process involved the gradual formation of a composite material consisting of geopolymer and coconut husk to form the geopolymer paste ensuring homogenization. The resulting paste is cast into a mold, sealed, and left in the oven for 24 hours at 80°C. Fig. 1 shows the geopolymer pellet after curing process. Four different composition of geopolymer pellets were introduced namely (MK-100%), (MK-65% and POFA-35%), (MK-65% and POFA-35%, 0.1g CH) and (MK-65% and POFA-35%, 0.3g CH) , the combination of all raw materials resulted in total mass of 10 grams, the separation of raw materials were made by different in weight percentage of metakaolin, palm oil fuel ash and coconut husk based on four different pellet compositions



(a) **(b)**

Fig. 1 *(a) Geopolymer pellets after curing process; (b) Immersion of geopolymer pellets into water samples.*

Penerbit UTHM

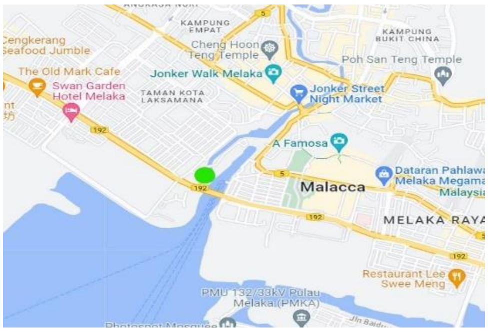


Fig. 2 *The location of water sample collection Melaka River water at Kampung Mortem Melaka.*

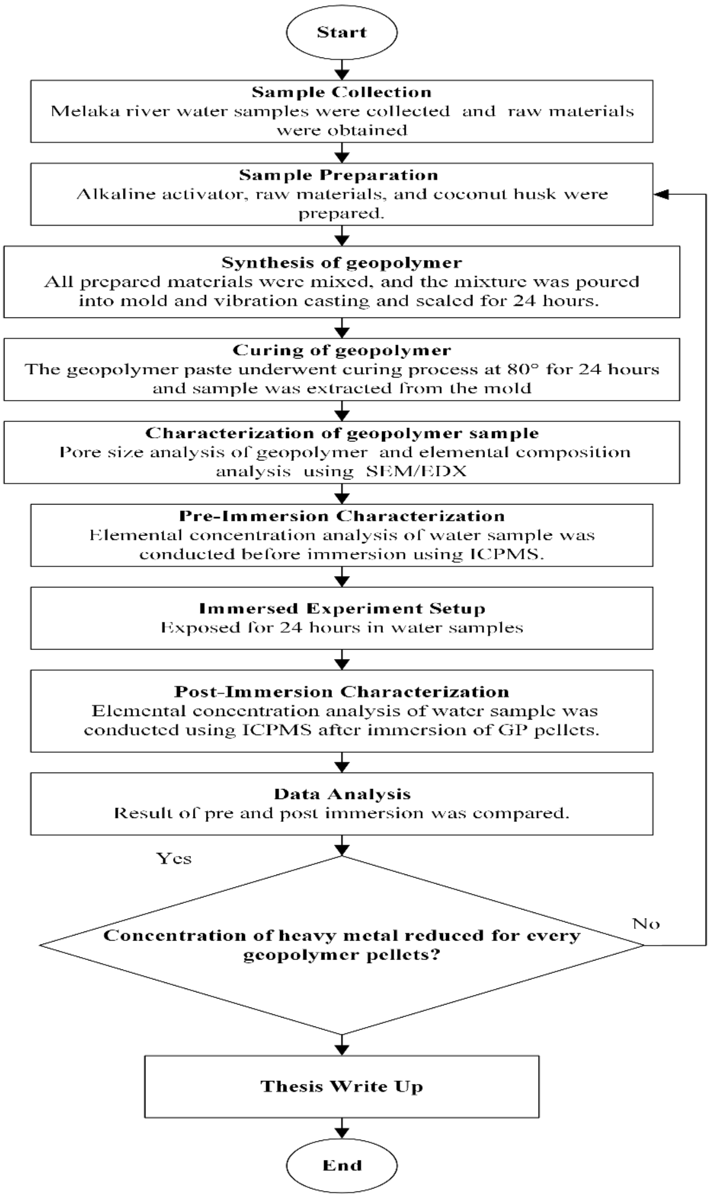


Fig. 2.1 *Flowchart for this research work.*

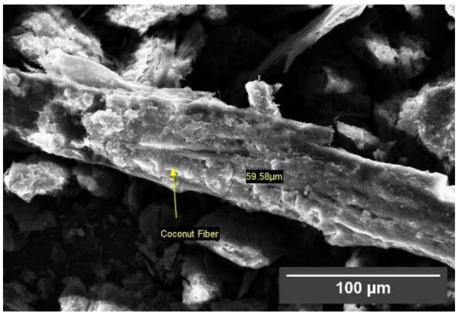
2.2 Sample Characterization

Prior to the sample characterization process, powder form of Palm Oil Fuel Ash (POFA), Metakaolin (MK), and Coconut Husk (CH) dispersed on a sample holder. This ensured uniform surface coverage. Gold sputtering was applied to facilitate Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray Spectroscopy (EDX) analyses, enabling the investigation of microstructural properties and elemental compositions. SEM/EDX analysis at various magnifications, along with quantitative evaluations using Image J software, provided insights into structural composition and pore size of the raw materials. The same analytical techniques were extended to the geopolymer-coconut husk composite, focusing on surface morphology and pore size. Additionally, pre-, and post-immersion analysis were employed, before and after immersion of geopolymer pellets into water samples were analysed using the ICP-MS machine. Inductively Coupled Plasma Mass Spectrometry (ICP-MS) was employed for quantifying heavy metal concentrations in the water sample after the immersion test, with rigorous calibration of ICPMS ensuring precision in the determination of heavy metal concentrations.

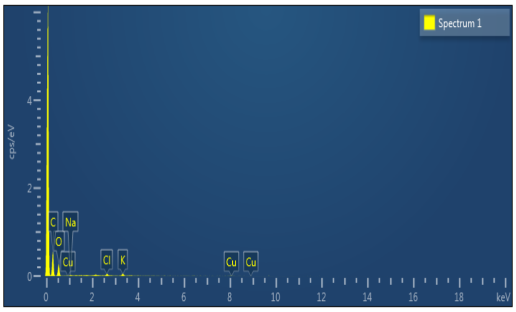
3. Results and Discussion

3.1 SEM/EDX analysis of raw coconut husk

The length of the raw coconut fibre was measured using Image J software in Fig. 3 (a) SEM image, and it was found to be 50.58µm. A detailed analysis was performed to accurately measure the fibre length. The EDX analysis of the raw coconut fibre sample in Fig. 3 (b) shows that it is primarily composed of carbon (57.01%) and oxygen (41.32%), indicating that the fibre is organic in nature. The presence of trace elements such as chlorine, potassium, copper, and sodium are observed in low concentrations. Elements can come from environmental factors or processing methods. The analysis is comprehensive, as the total atomic percentage adds up to 100%. The raw coconut fibre sample has a high carbon percentage of 57.01%, which is typical for organic materials like plant fibres. Carbon is an essential component of organic compounds, particularly in plant tissues where it plays a significant role in complex molecules like cellulose and lignin.



(a)

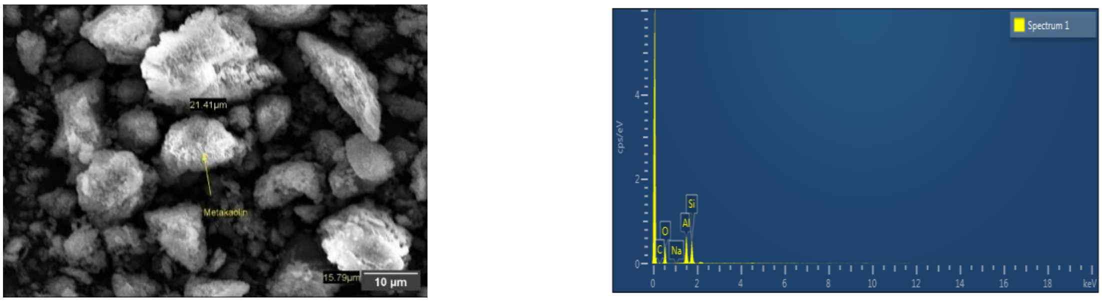


(b)

Fig. 3 *(a) SEM image of raw coconut fibre sample and (b) EDX analysis of raw coconut fibre sample.*

3.2 SEM/EDX analysis of raw metakaolin

The SEM analysis of the raw metakaolin sample which has reveals a maximum dimension of 21.41 µm in the microscope image which is shown in Fig. 4 (a). Additionally, the absence of visible pores in the microscopic structure suggests a relatively dense or non-porous nature of the metakaolin sample at the observed scale. The Energy Dispersive X-ray (EDX) analysis of the raw metakaolin sample in Fig. 4 (b) reveals a diverse elemental composition, with the following atomic percentages: Aluminium (Al) at 12.57%, Oxygen (O) at 57.75%, Silicon (Si) at 11.72%, and Carbon (C) at 17.97%. The total atomic percentage adds up to 100%, indicating a comprehensive assessment. The high oxygen content is consistent with the mineral-rich nature of metakaolin, while aluminium and silicon further suggest a clay mineral composition. Metakaolin is a high-reactivity aluminosilicate pozzolan derived from the calcination of kaolin clay. This reactivity is essential in the geopolymerization process, where metakaolin reacts with an alkaline activator to form a geopolymer binder. The presence of carbon, though relatively substantial, may be attributed to organic impurities or a carbonaceous component in the mineral structure. This elemental breakdown provides valuable insights into the composition of raw metakaolin.

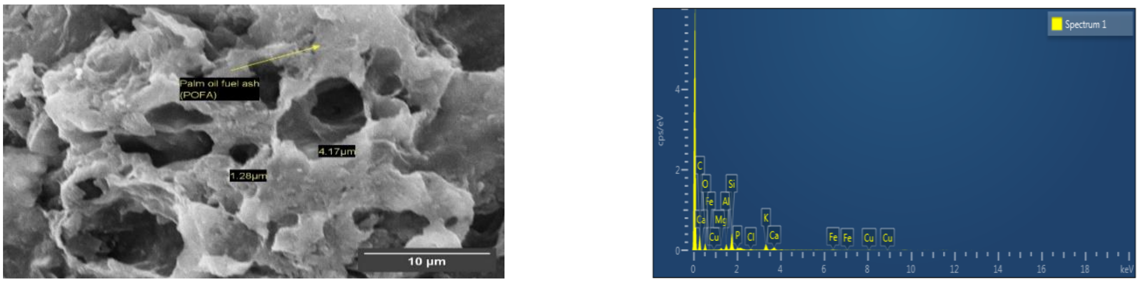


(a) **(b)**

Fig. 4 *(a) SEM image of raw metakaolin sample and (b) EDX analysis of raw metakaolin sample.*

3.3 SEM/EDX analysis of palm oil fuel ash

The SEM image of the palm oil fuel ash (POFA) in Fig. 5 (a) reveals the presence of pores. Two specific pore sizes where the first pore has a size of 4.17 µm, and the second pore has a size of 1.28 µm. These dimensions provide information about the scale of the pores observed in the material. The Energy Dispersive X-ray (EDX) analysis of the raw palm oil fuel ash sample in Fig. 5 (b) depicts a diverse elemental composition, with the following atomic percentages: Silicon (Si) at 4.53%, Aluminium (Al) at 1.69%, Oxygen (O) at 28.58%, Potassium (K) at 1.65%, Calcium (Ca) at 0.82%, and Carbon (C) dominating at 61.07%. The total atomic percentage sums to 100%, indicating a comprehensive analysis. The significant presence of carbon suggests the existence of organic or carbonaceous components in the palm oil fuel ash. By utilizing POFA, a waste product of the palm oil industry, can contribute to waste reduction and potentially lower the overall cost of raw materials for geopolymer mixture. Additionally, the presence of silicon, aluminium, oxygen, potassium, and calcium implies a diverse mineral composition.

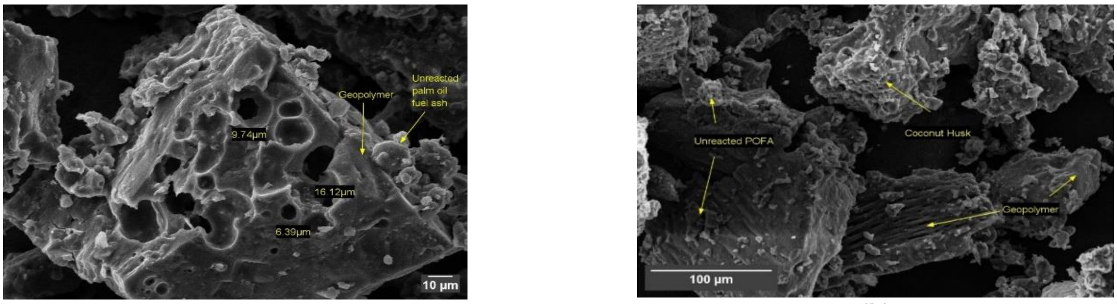


(a) **(b)**

Fig. 5 *(a) SEM image of raw palm oil fuel ash sample and (b) EDX analysis of raw palm oil fuel ash sample.*

3.4 SEM/EDX analysis of geopolymer pellet

The SEM analysis of the MK-65% and POFA-35% geopolymer sample with different weight percentage of metakaolin and palm oil fuel ash without addition coconut husk. Fig. 6 (a) revealed visible pores, and their sizes were quantified using Image J software. The measured pore sizes were noteworthy, with the first pore at 9.74 µm, the second at 6.39 µm, and the third at 16.12 µm, all observed under a magnification of 2500x. In Fig. 6 (b), which depicted the geopolymer sample with 1% coconut husk addition, the SEM image revealed some unreacted palm oil fuel ash and successful integration of coconut husk with the geopolymer sample. The magnification for this analysis was 1500x, providing an overall view of the three components. The amalgamation of coconut husk with the geopolymer suggested a successful combination process. This integration was expected to increase the porosity of the material, potentially enhancing heavy metal absorption capabilities. The SEM analysis, along with the introduced coconut husk, presented a promising avenue for improving the material's properties, particularly in applications involving heavy metal absorption.



(a) (b)

Fig. 6 *(a) SEM image of MK-65% and POFA-35% geopolymer sample and (b) SEM image of MK-65% and POFA-35% geopolymer with 1% of coconut husk sample.*

3.4.1 Geopolymer Pellets Absorption

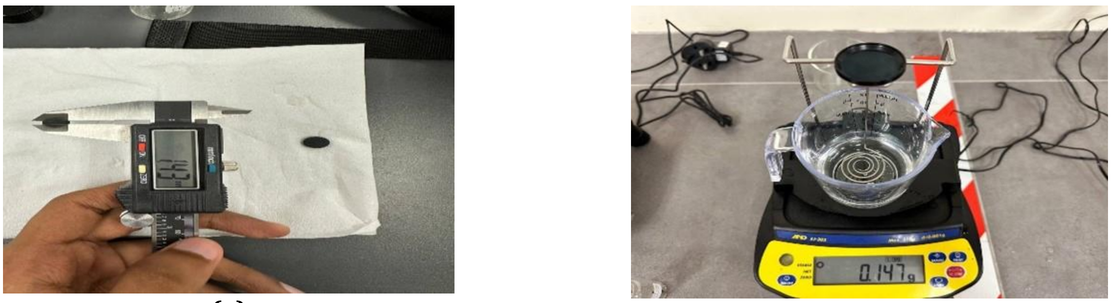
Fig. 6 shows the testing of the capability of geopolymer pellets to absorb nickel ion. The absorption of nickel ion using geopolymer pellets was analyzed as the nickel nitrate solution turned colorless after immersion of geopolymer pellets for 5 days. The nickel ion precipitated on the outer surface of the geopolymer pellet. Two geopolymer pellets were used in this test, namely (MK-100%) and (MK-65% and POFA-35%). The resulting solution was filtered to remove the residue from the solution.

(a) (b)

Fig. 6 *(a) Pre immersion of GP pellet into nickel nitrate and (b) Post immersion of GP pellet after 5 days.*

3.4.2 Physical Analysis

Fig. 7 (a) the thickness of the pellet was measured using Vernier calliper for all the geopolymer samples, and in Fig. 7 (b) shows the density of the geopolymer samples was measured using electronic density measurement kit. The density of the geopolymer samples were calculated by measuring the mass of the sample in air and in water for every geopolymer pellet, and the density of each geopolymer sample was obtained. The average thickness of the pellets was 1.43 mm and the mass of the pellet in air noted was 0.337 g whereas the mass of pellet in water noted was 0.147 g which by using the electronic density kit the density of the pellets observed was 1.760 g/cm^3.



(a) (b)

Fig. 7 *(a)Thickness measurement of the pellet and (b) density measurement of the pellet.*

Enhanced Knowledge in Sciences and Technology Vol. 4 No. 1 (2024) p. 229-237

3.5 Data Analysis of heavy metal concentration between various geopolymer pellets

The concentrations of heavy metals in the geopolymer samples before and after immersion shown in Table 1, with varying compositions of metakaolin (MK), palm oil fuel ash (POFA), and coconut husk (CH), provide insights into the potential for these materials to mitigate heavy metal contamination. Notable trends include a general decrease in concentrations post-immersion, indicating the capacity of the geopolymer to adsorb or interact with heavy metals. The addition of coconut husk, particularly with 0.1g, leads to a significant reduction in several heavy metal concentrations, including Nickel, Chromium, and Iron. This suggests a potential synergistic effect between coconut husk and the geopolymer matrix in sequestering these metals. However, with 0.3g coconut husk addition, there are slight fluctuations in concentrations, highlighting the nuanced influence of coconut husk dosage. The observed changes underscore the intricate interplay between the geopolymer matrix and coconut husk in affecting heavy metal concentrations, demonstrating promise for environmental applications such as heavy metal remediation.

Table 1 *Heavy metal concentration between various geopolymer pellets water.*

Heavy Metals/ Concentration of MRW with immersed GP pellet (ppb)	Nickel	Chromium	Iron	Zinc	Manganese	Copper	Arsenic
(Pre-Immersion) (ppb)	196	12.10	434	33.00	138.00	32.95	21.20
(Post-Immersion with MK-100%)(ppb)	56.8	2.42	256	30.7	45.92	25.31	19.70
(Post-Immersion with MK-65%, POFA-35%)(ppb)	107	9.89	206.00	28.7	123	8.40	18.20
(Post-Immersion with MK-65%, POFA35%,0.1g-CH) (ppb)	49.70	4.98	133.00	14.00	46.90	21.00	18.67
(Post-Immersion with MK-65%, POFA-35%,0.3g-CH) (ppb)	39.10	5.56	153.00	10.00	37.79	5.36	15.00

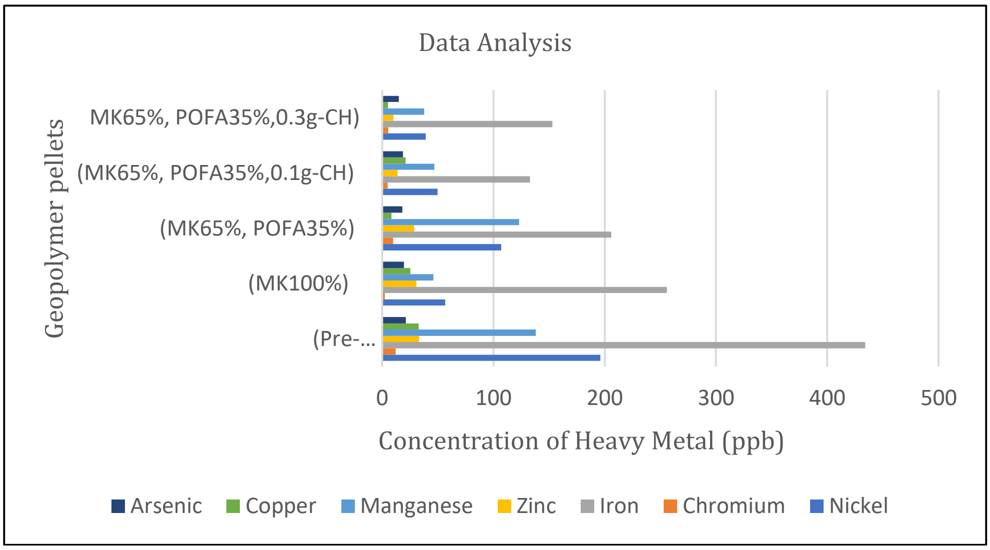


Fig. 7 *Post analysis of heavy metal concentrations between various geopolymer pellet*

The table 2 shows the removal efficiencies of heavy metals using four distinct pellet composition namely MK-100%, MK-65% POFA-35%, MK-65% POFA-35% 0.1g-CH (coconut husk), and MK-65% POFA-35% 0.3g-CH. For MK-100% pellets, chromium exhibits the highest removal efficiency at 80.00%. Introduction of POFA in MK-65% POFA-35% pellets shifts the spotlight to copper, demonstrating a removal efficiency of 74.51%. The subsequent incorporation of coconut husk in (MK-65% POFA-35% 0.1g-CH)and (MK-65% POFA-35% 0.3g-CH)pellets reveals nickel (74.64%) and copper (83.73%) as the heavy metals with the highest removal efficiencies. This underscores the efficacy of coconut husk as an adsorbent, influencing the selective removal of specific heavy metals in these formulations. These findings have implications for designing environmentally sustainable and effective pollutant remediation strategies. The calculation of the percentage removal will be done using the formula:

$$\text{Removal efficiency}(\%) = \frac{(C_o - C_t)}{C_o} \times 100 \quad (1)$$

Table 2 *Removal efficiency between various geopolymer pellet*

Removal Efficiency of Heavy Metals/ Different GP pellets	Nickel	Chromium	Iron	Zinc	Manganese	Copper	Arsenic
MK-100%	71.02%	80.00%	41.00%	7.06%	66.72%	23.18%	7.08%
MK-65%, POFA-35%	45.41%	18.26%	52.53%	13.03%	10.87%	74.51%	14.15%
MK-65%, POFA-35%, 0.1g-CH	74.64%	58.84%	69.35%	57.58%	66.01%	36.27%	11.93%
MK-65%, POFA-35%, 0.3g-CH	80.05%	54.05%	64.75%	69.70%	72.62%	83.73%	29.25%

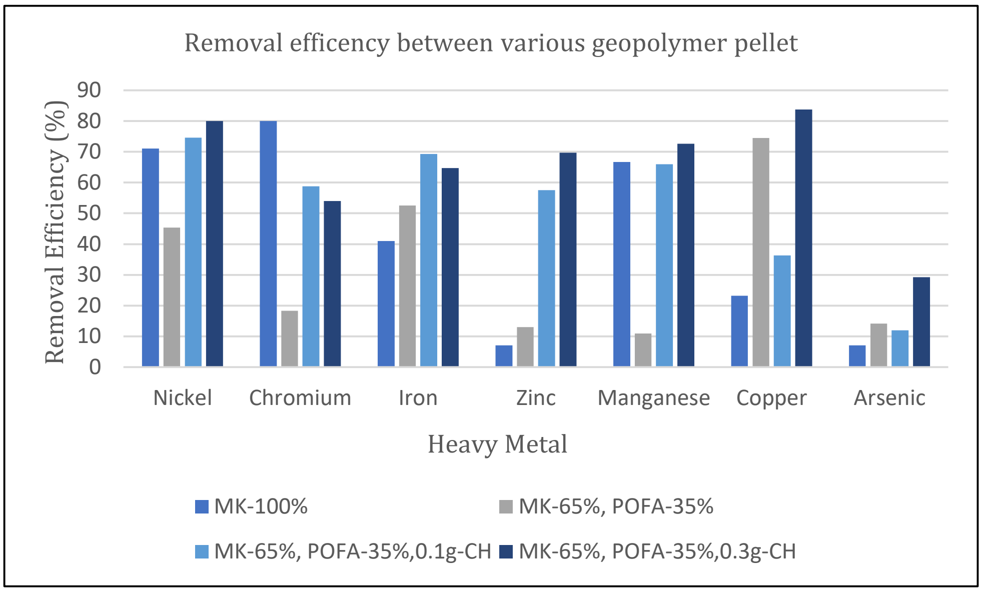


Fig. 8 *Removal efficiency between various geopolymer pellet*

4. Conclusion

As a conclusion, this study aimed to explore the potential of geopolymer-coconut husk composites as adsorbents for heavy metal removal from contaminated water sources. Incorporating coconut husk into the geopolymer matrix was investigated for enhanced adsorption capabilities. Characterization using scanning electron microscopy-energy dispersive X-ray analysis (SEM-EDX) confirmed successful integration of coconut husk particles within the geopolymer matrix. Batch adsorption experiments demonstrated the composites' remarkable capacity in removing heavy metals from aqueous solutions. Varied adsorption efficiency was noted with different weight percentages of geopolymer composite and coconut husk, influenced by factors such as pH, adsorbent dosage, initial concentration, contact duration, and temperature. Immersion studies, utilizing SEM-EDX and inductively coupled plasma mass spectrometry (ICP-MS), revealed the effective removal of heavy metals by geopolymer-coconut husk composites in real-world scenarios. The best geopolymer pellet is the geopolymer pellet with addition of 3% coconut husk which demonstrated the highest removal efficiency, indicating enhanced porosity from coconut husk addition facilitating heavy metal absorption by the geopolymer pellet.

Acknowledgement

The authors would like to thank the Faculty of Applied Sciences and Technology, Universiti Tun Hussein Onn Malaysia, for its support in completing this project.

Conflict of Interest

Authors declare that there is no conflict of interests regarding the publication of the paper.

Author Contribution

The authors confirm contribution to the paper as follows: ***study conception and design, data collection, methodology, analysis and interpretation of results:*** *Gokkul Pillay V Tamilvenan and Zaidi Embong. All authors reviewed the results and approved the final version of the manuscript.*

References

[1] K. Komnitsas and D. Zaharaki, "Geopolymerisation: A review and prospects for the minerals industry," *Minerals Engineering*, vol. 20, no. 14. pp. 1261–1277, Nov. 2007. doi: 10.1016/j.mineng.2007.07.011.

[2] S. Mishra *et al.*, "Heavy Metal Contamination: An Alarming Threat to Environment and Human Health," in *Environmental Biotechnology: For Sustainable Future*, Springer Singapore, 2019, pp. 103–125. doi: 10.1007/978-981-10-7284-0_5.

[3] R. Proshad *et al.*, "Appraisal of heavy metal toxicity in surface water with human health risk by a novel approach: a study on an urban river in vicinity to industrial areas of Bangladesh," *Toxin Rev*, vol. 40, no. 4, pp. 803–819, 2021, doi: 10.1080/15569543.2020.1780615.

[4] J. E. G. van Dam *et al.*, "Process for production of high density/high performance binderless boards from whole coconut husk. Part 2: Coconut husk morphology, composition, and properties," *Ind Crops Prod*, vol. 24, no. 2, pp. 96–104, Sep. 2006, doi: 10.1016/j.indcrop.2005.03.003.

[5] M. S. Islam, M. K. Ahmed, M. Raknuzzaman, M. Habibullah -Al- Mamun, and M. K. Islam, "Heavy metal pollution in surface water and sediment: A preliminary assessment of an urban river in a developing country," *Ecol Indic*, vol. 48, pp. 282–291, 2015, doi: 10.1016/j.ecolind.2014.08.016.

[6] Othman, S. A., & Fadzil, N. F. (2021). Utilization and valorization of Citrus Fruit by-products: A Review. *Enhanced Knowledge in Sciences and Technology*, *1*(2), 170-176.

[7] K. Unadkat and P. Parikh, "A Review on Heavy Metal Absorption Capacity of Aquatic Plants: Sources, Impact and Remediation Technique Molecular Characterisation of Plants of Dangs Umbrella programme GSBTM View project International Journal of Allied Practice, Research and Review A Review on Heavy Metal Absorption Capacity of Aquatic Plants: Sources, Impact and Remediation Technique," *UGC JOURNAL NUMBER 44557 IJAPRR International Peer Reviewed Refereed Journal*, vol. IV, [Online]. Available: www.ijaprr.com

[8] M. Alshabib and S. A. Onaizi, "A review on phenolic wastewater remediation using homogeneous and heterogeneous enzymatic processes: Current status and potential challenges," *Separation and Purification Technology*, vol. 219. Elsevier B.V., pp. 186–207, Jul. 15, 2019. doi: 10.1016/j.seppur.2019.03.028.

[9] *Heavy metal pollution status in the Malaysian aquatic environment* (no date) *Scholarly publishing collective.org*. Availableat: https://scholarlypublishingcollective.org/msup/aehm/article-abstract/9/2/137/168938/Heavy-metal-pollution-status-in-the-Malaysian (Accessed: 18 January 2024).

[10] SATRIA, A. H. B. M. (2022). Isolation and characterization of biosurfactant-producing lactic acid bacteria from sauerkraut. *Enhanced Knowledge in Sciences and Technology*, *2*(2), 060-069.