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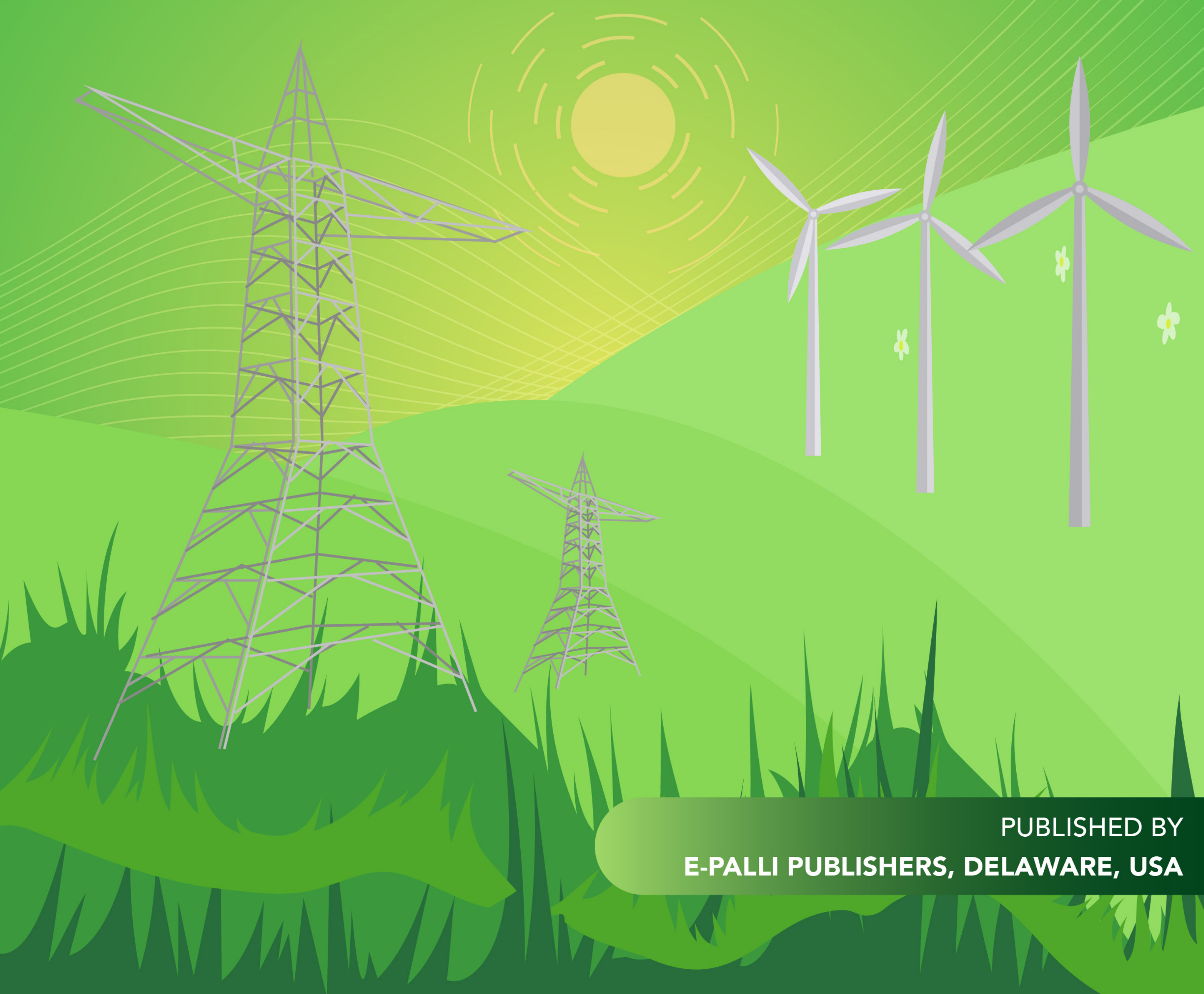

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Remediation of Contaminated Bauxite Residue Using *Gracilaria Sp.* Adsorbents

Edward Hingha Foday Jr[1*], Augustine Daniel Adu[2], Alpha Amara Sei[3], Anthony Amara Golia Seseh Kanneh[3],
George Musa Missailie[3]

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ABSTRACT

Bauxite residue, often referred to as red mud, is the main waste product of alkaline alumina extraction from bauxite that includes high concentrations of heavy metals. The ecosystems, plants, animals, and human health are all at risk from these heavy metals. The removal of heavy metals (As, Ni, Cr, and Fe) from bauxite residue using *Gracilaria sp.* is the main topic of this article. By employing the oil spherification process, *gracilaria sp.* was used to create alginate beads that effectively removed heavy metals from bauxite waste. The contaminated bauxite residue was cleaned up using these beads as adsorbents. According to the findings, *gracilaria sp.* were successful in eliminating Cr, As, Ni, and Fe. The best concentration for removing iron (Fe) from 30 millilitres of bauxite residue solution at 25 °C was 1g of alginate beads. The optimal concentrations for removing chromium (Cr) were 2g and 3g, whereas 3g of *gracilaria sp* adsorbents at a constant temperature of 25 °C were ideal for removing arsenic (As) and nickel (Ni).

INTRODUCTION

The naturally occurring, heterogeneous substance known as bauxite is mostly made up of one or more minerals of aluminium hydroxide, together with trace quantities of silica, iron oxide, titanium, alumina silicate, and other impurities (Zainudeen *et al.*, 2023). This lateritic ore is rich in aluminium and iron oxide and is usually found in the tropics between 30° north and 30° south of the equator (Elias, 2002). Alumina is extracted from bauxite via the Bayer process, wherein sodium hydroxide is combined with the ore and heated to temperatures between 200 °C and 270 °C in a pressure chamber, resulting in the dissolution of alumina (chemically known as aluminium oxide, Al_2O_3), which is subsequently filtered out, leaving behind the waste by-product known as bauxite residue (Hind *et al.*, 1999; Kamara *et al.*, 2025). The residue from bauxite is therefore a reddish-brown, extremely alkaline, non-flammable material that is loaded with heavy metals and occasionally contains naturally occurring radionuclides (Klauber *et al.*, 2009). Red mud is the primary by-product produced during the caustic digestion of bauxite ores in the alumina manufacturing process. Metallurgists consider red mud to be a polymetallic raw material, comprising a complex composition of oxides, including aluminium, iron, titanium, and silicon, along with other valuable elements such as scandium, uranium, and thorium (Klauber *et al.*, 2011). However, because red mud is very caustic, contains heavy metals, is alkaline, and radioactive, these disposal methods may pose complications. These include the seeping of red mud slurry and the leaching of alkaline solution from the enclosing barriers. All red mud has a rust tint, which is a result of the oxidised iron in the mud. This is true even if the physical, chemical, and mineralogical characteristics of bauxite waste vary depending on the mineral sources and refining methods utilised. Furthermore, solid elements of red mud include predominantly iron oxides (particularly haematite), alumina, and several hazardous heavy metals (Li *et al.*, 2023).

If radioactive materials were present in the original bauxite, it may also be somewhat radioactive. A few of the main environmental issues for the safe and cost-effective disposal of red mud include its high water content, strong alkalinity, and hazardous heavy metals. Because of its inherent characteristics (high pH, saline, heavy metals, and radioactivity), the disposal of tailings seriously harmed the surrounding populations' ecosystem (Foday Jr *et al.*, 2021). The caustic leftovers (sodium) from these "red mud lakes" seeped into the nearby subterranean aquifers. Heavy metals, naturally occurring radioactive material (NORM), and soda/alkalinity are the three main techno-environmental components that are the main source of danger, according to the Commonwealth Scientific and Industrial Research Organisation (CSIRO) (Gore, 2015). Another significant environmental hazard is dust, which can spread kilometres outside the containment area and harm the health of nearby inhabitants and employees of the alumina producing plant. Another critical issue is the seepage of leachate from the containment area, which can infiltrate the local groundwater table, transporting caustic solutions and trace metals into the ground and consequently affecting the groundwater supply. Bauxite

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[1] Department of Civil and Environmental Engineering, Faculty of Engineering and Innovation, Eastern Technical University of Sierra Leone, Combema Road, Kenema City, Sierra Leone
[2] Department of Business Administration, Faculty of Business and Entrepreneurship Studies, Sierra Leone
[3] Department of Social Studies, Faculty of Teacher Education, Eastern Technical University of Sierra Leone, Combema Road, Kenema City, Sierra Leone
[*] Corresponding author's e-mail: hinghaja@gmail.com

residue is known to contain considerable levels of trace metals, including arsenic, cadmium, chromium, lead, mercury, vanadium, and zinc (Jones & Haynes, 2011; Liu *et al.*, 2009). Other works have listed the major components concerning bauxite residue leachate. According to Klauber *et al.* (2011), when utilising bauxite residue as a construction material, the concentration of Ac, At, Bi, Pa, Pb, Po, Ra, Th, Tl, and U should be evaluated.

As, Ba, Cd, Cl, Cr, Cu, Fe, Hg, Mo, Ni, Pb, Sb, Se, SO_4, Th, U, and Zn were identified by URS/Alcoa as elements of importance in the analysis of bauxite residue (Gore, 2015). Chronic health concerns, as well as abrupt environmental risks to surrounding soil, air, water, and ecosystem, are among the environmental consequences of bauxite residue use. Considering the issue of heavy metal impacts, they can have long-lasting and severe effects on the environment at local levels (Mayes *et al.,* 2016; Roadcap *et al.*, 2005). Concern over environmental pollution by these heavy metals has grown in recent years, both ecologically and in terms of public health worldwide. Furthermore, human exposure has increased considerably as a result of an exponential expansion in their usage in numerous industrial, agricultural, residential, and technical applications(Bradl, 2004; Kamara *et al.*, 2023). The environment is known to include heavy metals from geological, industrial, agricultural, pharmaceutical, domestic wastewater, and atmospheric sources (He *et al.*, 2005; Jagaba *et al.,* 2024). Environmental contamination is highly prevalent in point source regions such as mining, foundries and smelters, and other metal-based industrial processes (Bradl, 2004; Foday Jr *et al.,* 2017). Despite being naturally occurring elements present in the earth's crust, the majority of environmental contamination and human exposure is caused by anthropogenic activities like mining and smelting, industrial production and use, and the use of metals and metal-containing compounds in homes and farms (Jomova *et al.*, 2025; Shallari *et al.*, 1998). Metal corrosion, air deposition, soil erosion of metal ions and leaching of heavy metals, sediment resuspension, and metal evaporation from water resources to soil and groundwater are other ways that the environment can get contaminated (Briffa *et al.*, 2020). Heavy metal contamination has also been found to be greatly influenced by natural processes like weathering and volcanic eruptions (Ali *et al.*, 2021). Heavy metals are also termed trace elements due to their existence at trace amounts (ppb range to less than 10 ppm) in diverse environmental matrices (Gupta *et al.*, 2023). Metals like cobalt (Co), copper (Cu), chromium (Cr), iron (Fe), magnesium (Mg), manganese (Mn), molybdenum (Mo), nickel (Ni), selenium (Se), and zinc (Zn) are essential nutrients, according to reports. Numerous deficiencies or disorders are caused by either excessive or insufficient exposure to certain micronutrients (Organization, 1996). The toxicity of heavy metals is dependent on several variables, including the chemical species, exposure route, and dose, in addition to the age, gender, genetics, and nutritional state of those who are exposed. Iron, nickel, mercury, arsenic, and chromium are among the priority metals of public health importance due to their high levels of toxicity (Mulware, 2020). Even at lower exposure levels, these metallic elements are known to cause numerous organ damage and are regarded as systemic toxicants. It is in this regard, a low-cost, easily available, and eco-friendly seaweed has been selected as an ideal biosorption material compared to other sorption materials. This article seeks to throw light on the importance of seaweed biomass as a sorption material and to summarise its biosorption mechanism. This article also pinpoints the toxic effects of heavy metals on environmental resources, as well as comparing the Gracillaria sp adsorption efficiency to other seaweed biomass. Also, the concentration of target heavy metals (As, Ni, Cr and Fe) before and after adsorption was determined using, Inductively Coupled Plasma Optical Emission Spectrophotometer (ICPOES). The optimum dosage concentration of adsorbents for effective and efficient heavy metal removal was also determined. It is in this vein that this research seeks to use *gracilaria sp* adsorbent to remove heavy metals from bauxite residue.

MATERIALS AND METHODS

Chemical Reagents

The chemical reagents used in this experiment were defined as analytical reagents (AR). All the aqueous solutions were prepared using deionised distilled water, while 7.5mL of phosphoric acid (H_3PO_4) and 0.5mL of nitric acid (HNO_3) were used for the preparation of samples as a standard solution in determining the heavy metals concentration.

Composition of Red Mud

The Bauxite Residue used for this study was collected from Bukit Goh in Pahang, Malaysia. It was kept in a plastic container for subsequent experiment at a room temperature of 15-20°C for 150 days. The Bauxite Residue was crushed/ grinded with a hand to ensure homogeneity and sieved to pass through a 200-300 mesh sieve. Iron (Fe), Chromium (Cr), Arsenic (As) and Nickel (Ni) concentrations were determined before and after the experiment. Microwave acid digestion was done to enhance the operation of the ICPOES. An Inductively Coupled Plasma Optical Emission Spectrophotometer (ICPOES, Agilent 710 model) was used to characterise the concentration of various heavy metals in the Red Mud.

Microwave Digestion Treatment

The Elemental composition of the Red Mud used for this study was determined through total microwave digestion according to US EPA SW 846 Method 3051A (US EPA, 1998). The heavy metals (Fe, Ni, Cr and As) contents in the soil were determined by adding H_3PO_4 and HNO_3 for the acid digestion method at a temperature of 220 °C. The digested solution was analysed for the heavy metals content using an Inductively Coupled Plasma Optical Emission Spectrophotometer (ICPOES, Agilent 710 model). The advantage of microwave-assisted treatment technology is to reduce the processing time with consequent reduction in energy consumption and the amount of gases employed for treatment (Lozano Pérez *et al.*, 2024). It allows variations in reagents and methodology, making it ideal for a variety of matrices and elements. Microwave energy is derived from electrical energy with a conversion efficiency of approximately 50% for 2,450 MHz and 85 % for 915 MHz (Haque, 1999). Microwave treatment affects the porosity of the adsorbents, causing modification of surface chemistry and thus enhancing the metal removal capacity of the adsorbent (Shi *et al.,* 2018).

Study Area

The sample of bauxite residue was taken from Bukit Goh, Pahang, Malaysia, which is located around 20 kilometres northwest of Kuantan, the biggest city on the east coast of West Malaysia, as shown in Figure 1. This location is notable for bauxite mining, which has a negative influence on the socioeconomic environment. Significant waterways such as Sungai Kuantan, Sungai Mabuk, and Sungai Riau exhibit a range of possible hazards (Kusin *et al.,* 2017). These rivers' physical alterations, such as their hue and odour, are indicators of how seriously polluted they are. The South China Sea receives the mining site's effluent. The most necessary components for human survival such as safe food, clean air, safe water and shelter are under jeopardy due to the pollution of these natural environment. Aggressive, unregulated bauxite mining in the research region, if not managed over time, may produce irreversible changes to the environment, threatening ecosystems to reach the Ultimate Environmental Threshold (UET).

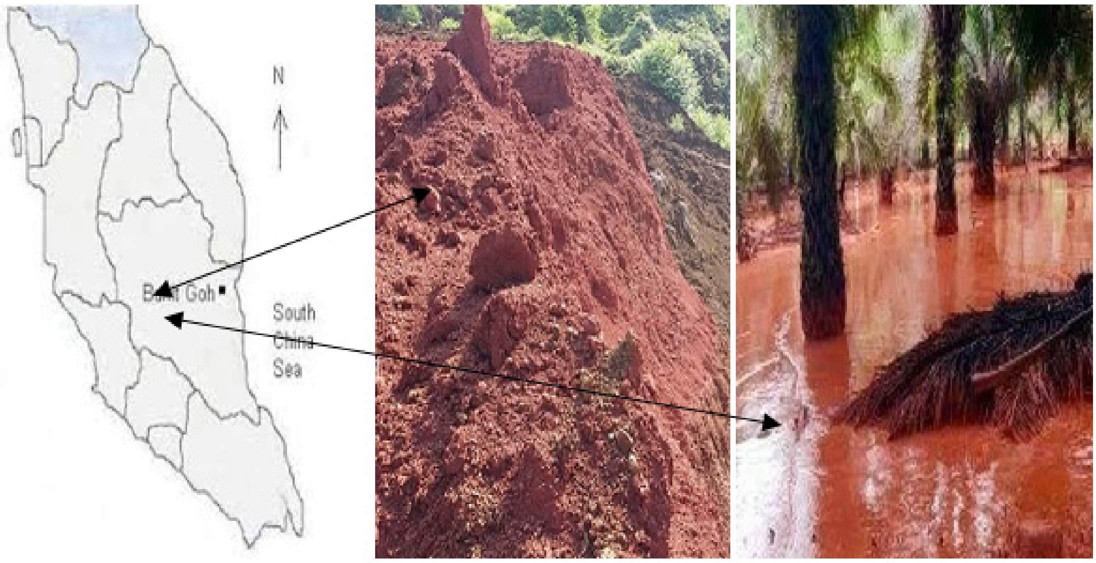

Figure 1: Study area showing Bauxite Residue

Seaweed Collection and Beads Formation

The seaweed (*Gracilaria sp*) was collected from Dickson Port, Kuala Lumpur, as it was used for the development of alginate beads that were used as adsorbents to remove heavy metals from Bauxite Residue. The dried local seaweed species (*Gracilaria sp.*) was made into powder by drying, grinding and sieving before the formation of spherical alginate beads. The spherical shape provides a high surface area to volume ratio, which increases the efficiency in the treatment of pollutants from leachate. Cold sunflower oil was used to develop the alginate beads. An optimum amount of 10g of dried seaweed species known as *Gracilaria sp* was introduced into a 1 L beaker with 400 ml of distilled water, and the mixture was left to boil for one hour. After one hour of boiling, the hot extraction was left to cool for 15 minutes. The cooled extraction solution is immediately introduced into cold sunflower oil (refrigerated for 24 hours before the experiment) using a syringe to produce spherical beads. Due to the low temperature and insolubility in oil, the boiled extraction will gelify in spherical shapes at the bottom of the beaker.

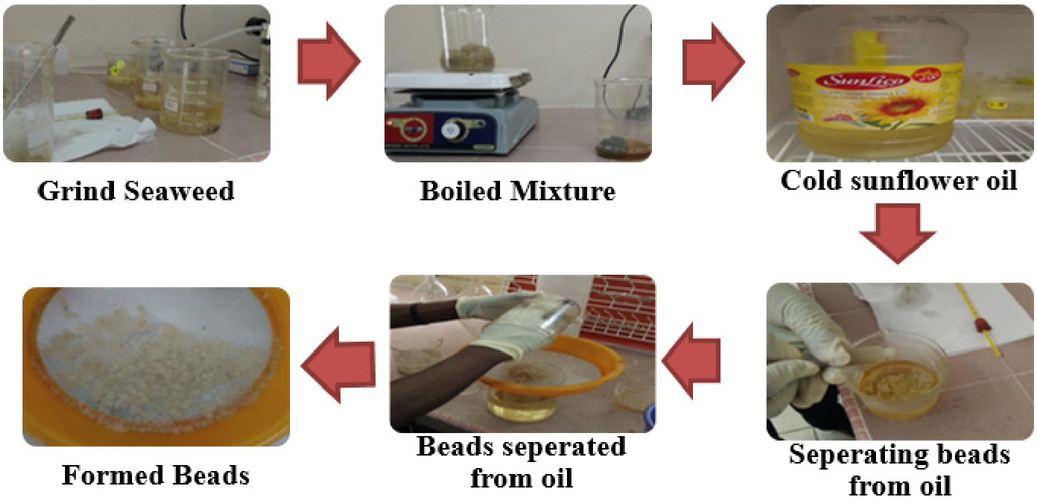

Figure 2: Steps showing spherical bead formation

Jar Test

The alginate beads of different weights (1g, 2g and 3g) from *Gracilaria sp.* were used during the Jar Test. The alginate beads were added to 3 different 50ml beakers filled with 30mL digested bauxite residue solution. The solutions were left to undergo the Jar Test for 1 hour and 30 minutes. The concentrations of heavy metals were again measured by using an Inductively Coupled Plasma Optical Emission Spectrophotometer (ICPOES). The Jar Test was stirred at 100rpm for 1hr 30min to satisfy the Langmuir and Faundlich adsorption theories, which deal with homogenous adsorbents concerning adsorbates and rough surface of adsorbates for adsorbents, respectively.

RESULT AND DISCUSSION

Table 1: Heavy metal concentrations before using *gracilaria sp.* Adsorbent

Heavy Metals	Wavelength	Heavy conc. before adsorbent (*gracilaria sp*)	Percentage Relative Standard deviation (%RSD)	Standard Deviation
Nickel (Ni)	230.299	0.26ppm	0.3	0.000905
Chromium (Cr)	267.716	3.80ppm	1.2	0.045874
Iron (Fe)	238.204	1029ppm	0.9	2.76048
Arsenic(As)	193.696	-0.50ppm	24.4	0.015012

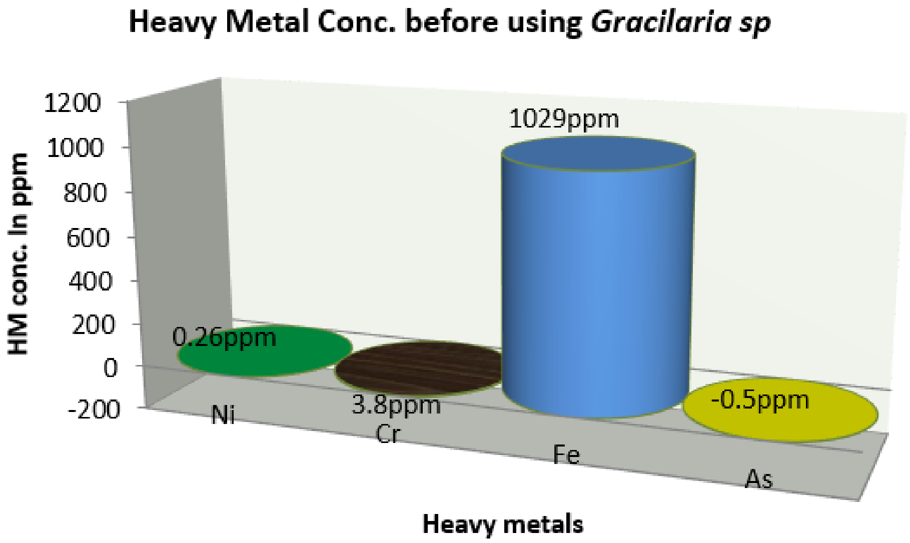

Figure 3: Heavy metal concentration before using *gracilaria sp.*

Figure 3 shows the heavy metal concentration in Bauxite Residue before the use of *Gracilaria sp* absorbent. Among the four(4) elements studied, Iron (Fe) was in high quantity, accounting for 1029ppm, with chromium (Cr), Nickel (Ni) and Arsenic (As) accounting for 3.8ppm, 0.26ppm and -0.5ppm respectively. The negative value for Arsenic (As) (-0.50ppm) indicates that the Arsenic (As) concentration was insignificant in the bauxite residue and with no health and environmental threat to the Bukit Goh's environment. Heavy metal concentrations in Bauxite Residue from Bukit Goh, Pahang, Malaysia were analysed using an ICPOES spectrometer (Agilent 710 model) and the results are shown in Table 1 above. The presence of Iron (Fe) in the study area is high, which is contrary to the Malaysian regulatory standard.

Heavy Metal Concentration after using *gracilaria sp.* Adsorbent

Table 2: *Gracilaria sp.*Adsorbents effect in the adsorption of heavy metals

Heavy Metals	**EQR 2009**	*Gracilaria sp* **(g)**	**Initial conc. ppm**	**Jar test time**	**Removal Conc.(ppm)**
Nickel (Ni)	0.20	1	0.26	1hr 30min	0.22
		2			0.24
		3			0.25
Chromium (Cr)	0.05	1	3.80	1hr 30min	3.40
		2			1.80
		3			3.65
Iron (Fe)	5.0	1	1029	1hr 30min	330
		2			248
		3			191
Arsenic (As)	0.05	1	-0.50	1hr 30min	-0.50
		2			-0.052
		3			-0.025

Nickel (Ni)

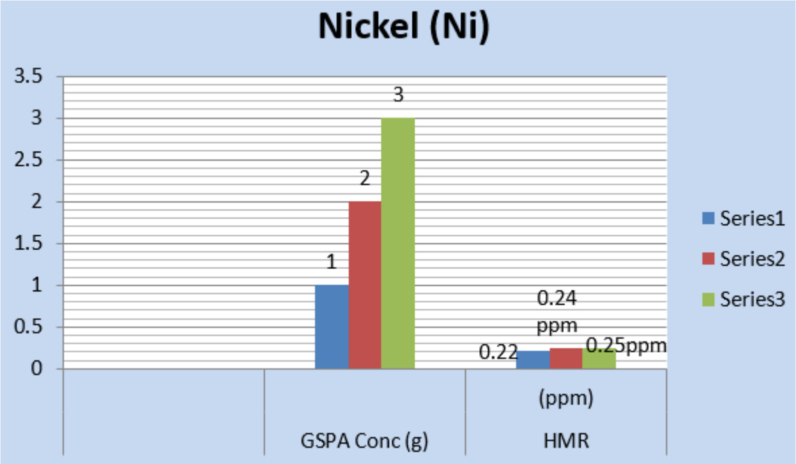

Figure 4: Nickel (Ni) removal concentration by using *gracilaria sp*

According to the Environmental Quality Regulation 2009 standard of Malaysia (Mustafa, 2022), Nickel (Ni) concentration before the use of *gracilaria sp* adsorbent was high, and this indicates an environmental threat to the community. The *Gracilaria sp.* adsorbent concentrations of 1g, 2g and 3grams were used to remediate 30ml contaminated digested Bauxite Residue Solution, respectively. After the use of *Gracilaria sp.* adsorbent at respective dosage concentrations 1g, 2g and 3g, the Nickel (Ni) concentrations decreased to 0.22ppm, 0.24ppm and 0.25ppm, respectively, when compared to the initial concentration (0.26ppm). The removal outcome of the heavy metals after the use of the *gracilaria sp.* was in the following order: 3g > 2g >1g. Thus, more grams of alginate beads can remove a higher concentration. Nickel (Ni) removal by *Gracilaria sp.* extract was possible due to the sulphide and hydroxide bonds. The ICPOES spectrometer also reveals that Nickel (Ni) has a low relative standard deviation of 0.3%.

Chromium (Cr)

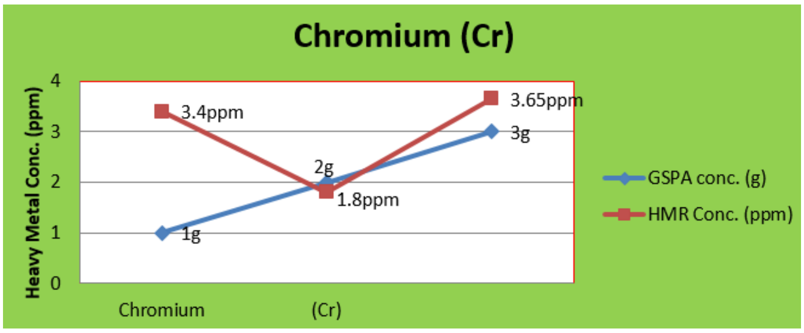

Figure 5: Chromium (Cr) removal by *Gracilaria sp.* Adsorbents

Figure 5 explains the removal efficiency of Chromium. Chromium (Cr) initial concentration was 3.8ppm (Table 2). The application of *gracilaria sp.* adsorbent of 1g, 2g and 3g shows an effective and efficient removal rate, especially with 3g alginate beads. The application of *gracilaria sp* adsorbents to the digested red mud solution gives the following chromium (Cr) removal concentration results with the respective concentration of adsorbents added (1g=3.4ppm, 2g=1.8ppm, 3g=3.65ppm). This indicates that chromium (Cr) can best be removed at both 1g and 3g of *gracilaria sp* adsorbent, as this enhances a healthy environment. The adsorption capacities of the heavy metals were in the following order: 3g > 1g > 2g. According to Akbar Esmaeili (Esmaeili *et al.*, 2010), the adsorption of heavy metals is dependent on the concentration of metal ion, retention time and pH of the metal solution and the Langmuir adsorption model.

Iron (Fe)

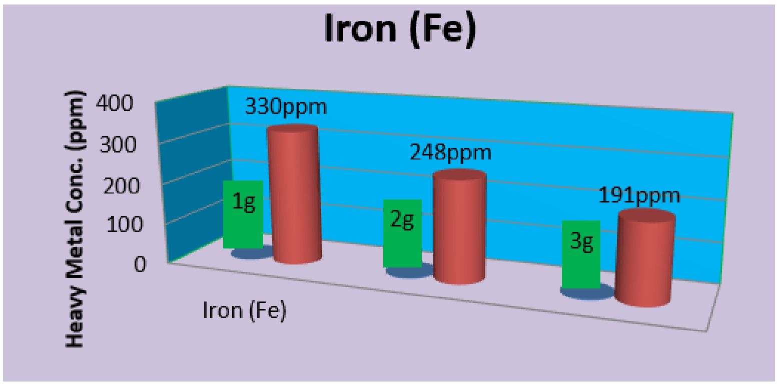

Figure 6: Iron removal by *Gracilaria sp.* Adsorbents

From the above figure, the iron (Fe) concentration was so high that it caused a very serious environmental threat, which will impair the health of the dwellers in the study area. The adsorption of Iron (Fe) by *gracilaria sp.* adsorbents shows significant changes, as shown in Figure 6. The lower the adsorbent concentration, the greater the removal efficiency. Iron (Fe) removal efficiency and effectiveness were more pronounced with 1g of adsorbents compared to 2g and 3g (Figure 6). Iron (Fe) is also capable of forming bonds with various chelating agents, such as sulfides, glycosides and hydroxides. The high availability of metal chelating bonds and high competitiveness of Fe (reactivity) explain the efficiency of the adsorption process (Gajski *et al.*, 2012). The adsorption capacities of the heavy metals using *gracilaria sp.* adsorbents were in the following order: 1g > 2g > 3g. It can be summarised that *gracilaria sp* adsorbents are efficient and effective in removing heavy metals from bauxite residue, especially at low concentrations of alginate beads.

Arsenic (As)

Arsenic is a very toxic heavy metal which is normally found in groundwater sources and mercury mines (Varank *et al.,* 2011). Despite being non-reactive in the red mud solution, Arsenic (As) could be adsorbed due to the presence of sulphide bonds in the *gracilaria sp.* extract (Kurniawan *et al.*, 2006; Kurniawan *et al.*, 2010). The Arsenic (As) concentration before the application of *gracilaria sp* adsorbent indicates none non-reactive value, which means the concentration of arsenic (As) shows no threat to the environment. Even though Arsenic poses no environmental threat in the study area, precautionary measures should be considered, as Arsenic is a ubiquitous element that is detected at low concentrations in virtually all environmental matrices. Arsenic (As) exposure affects virtually all organ systems, including the cardiovascular, dermatologic, nervous, hepatobiliary, renal, gastrointestinal, and respiratory systems (Shiowatana *et al.*, 2001; Shiowatana *et al.*, 2001).

CONCLUSION

The treatment of Bauxite Residue using *gracilaria sp.* demonstrated that *gracilaria sp.* was effective and efficient in removing heavy metals from bauxite residue. However, the treatment effectiveness of nickel (Ni) removal from the bauxite residue employing *gracilaria sp* was so small that its removal rate can scarcely be observed when compared to chromium and iron. From the overall observation, *Gracilaria sp.* adsorbent was most suitable for the treatment of heavy metals from bauxite residue. However, the concentration of arsenic was insignificant compared to the Iron (Fe), Chromium (Cr) and Nickel (Ni), which were of great concern because of their high concentration and concomitant effect on the environment and health of inhabitants in Pahang, Malaysia. The adsorption of heavy metals in Red Mud was due to the presence of glycoside bonds in agar, sulphide bonds in carrageenan, polyphenols and hydroxide bonds (Blaga *et al.*, 2021). *Gracilaria sp.* adsorbents were found effective and efficient at various concentrations of 1g, 2g and 3g.In the removal of Iron (Fe), 1g of *gracilaria sp.* adsorbents was the most ideal concentration for the purification of 30ml bauxite residue solution at a temperature of 25 °C. Chromium (Cr) element was best removed at a concentration of 2g and 3g, while Nickel (Ni) was ideal with 3g of *gracilaria sp* adsorbents.

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