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THE ADSORPTION OF HEAVY METALS FROM INDUSTRIAL WASTEWATER USING SARGASSUM CRASSIFOLIUM

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ABSTRACT: *Sargassum crassifolium* is the origin strains of macroalgae from Indonesia which was the most effectively adsorb heavy metals from liquid solution and it was investigated the adsorptive capability of heavy metals in industrial wastewater. The macroalgae were collected at Kotok Besar Island, Seribu Island, North Jakarta, Indonesia and were treated with the variety of pH 2, 3, 4, 5 and 9 for 60 minutes oscillation, in duplicate assessment. The macroalgae were selectively effective for removal of Cd, Hg, and Pb compared with other heavy metals, achieving 75-99.05% adsorption efficiency, mostly in acidic condition for 60 minutes oscillation, except Hg. The selectivity order for other metal ion uptake by *S. crassifolium* was Cu>Fe>Co>Ni>Cr with percentage removal below 56%. Due to the low cost, availability and significantly high adsorption capability, the brown macroalgae *S. crassifolium* is able to be used for selective removal of heavy metals from industrial wastewater effectively in acidic pH range.

Keywords: Adsorption, Heavy metals, Industrial wastewater, Sargassum crassifolium

1. INTRODUCTION

An increase in human population leads to a significant enhancement in waste production. Nowadays, the most notable wastes are the heavy metals because of the high toxicity to living creatures [1]. Along with the development of science and technology, the use of heavy metals, especially in industries is increasing and causing many environmental problems [2]. Heavy metals wastes are discharged into the environment from various industries working on electroplating, leather tanning, water cooling, pulp, and ore and petroleum processing industries that are suspected to be highly toxic to ecology and human health [3], and can also contaminate the environment persistently [4,5]. In Indonesia, Government Regulation of Republic of Indonesia No. 82/2001 on Water Quality Treatment and Water Pollution Control [6] is used to monitor the heavy metals concentrations commonly generated by industries.

The removal of heavy metals from the environment is the urgent action to conserve nature in a sustainable and further to protect human health. The biological agent is one of the most efficient methods used to reduce the pollution of heavy metals in water including macroalgae [7-9], microalgae [10-12], mushroom [13], and agriculture waste [14]. The ion exchange and the formation of complex compounds are the heavy metal adsorption schemes that occur within the walls of algal cells**.** The structure of the cell wall produces a high seaweed binding capacity because it has the various functional groups involved including: (1) alginic acid, with carboxyl groups and sulfated polysaccharides, as well as sulfonic acid, in brown algae matrix [15-17]; (2) sulfated galactans in red algae [15,18,19]; and (3) an external capsule composed of proteins and/or polysaccharides in green algae [20].

Algal materials have been broadly observed for heavy metal adsorption, particularly the potential of marine algae, including macroalgae which has been widely investigated. Non-living seaweed or dried seaweed can be used as an alternative to the low-cost adsorbents [14] compared to other methods such as more expensive physicochemical processes. In Indonesia, the studies of macroalgae as heavy metal adsorbents are limited in an aqueous solution, such as the study of adsorption of Cu(II) on *Sargassum crassifolium* [21], Cr(III) on *Euchema spinosum* [22] and Pb(II) on *Sargassum duplicatum* [23], *S. crassifolium, Euchema spinosum* and *Padina minor* [24]. The study of heavy metals removal from industrial wastewater, however, has not been widely conducted.

An inexpensive material such as marine macroalgae is suitable for removing the heavy metals from industrial wastewater [15,16,18-20,25]. In the present study, the industrial wastewater was used as a medium to test the ability of *S. crassifolium* to adsorb the heavy metals and as a continuation of the previous study that conducted testing with an aqueous solution. *Sargassum crassifolium* is a genuine strain of Indonesian seaweed that grows abundantly in Indonesian waters. Therefore, it is appropriate to use *S. crassifolium* as an adsorbent in this study which aims to analyze its efficiency to absorb the heavy metals from the industrial wastewater. The results of this study can be applied in industrial wastewater treatment and subsequently contribute to reducing pollution in the aquatic environment.

2. MATERIALS AND METHODS

This study was conducted in the Integrated Laboratory Center of the Faculty of Science and Technology, State Islamic University Syarif Hidayatullah Jakarta, including preparation of the samples. The biomass samples of macroalgae *S. crassifolium* were collected from the water surrounding Kotok Besar Island, Seribu Islands, North Jakarta, Indonesia. The samples were used in the dried form. For wastewater sample, it was obtained from the company of hazardous wastes treatment located in Cileungsi region, Indonesia.

2.1. Preparation of Adsorbent

All sediments or small organisms which were trapped in the macroalgae samples were washed with the seawater and put in the zipped plastic bag, then brought to the laboratory. In the laboratory, all samples were washed and rinsed again using distilled water and then dried in an oven at 50 C for 24 hours to get the stable weight. The dried samples were pounded with a mortar and pestle and sieved to the size 250-500 µm. The powder of adsorbent was ready to use and kept it at room temperature.

2.2. Adsorption Test

The experimental design used was Completely Randomized Design with two (2) replications. The eight heavy metals, including Cd, Co, Cr, Cu, Fe, Hg, Ni, and Pb, and 5 variations of pH, namely 2, 3, 4, 5, and 9 which were oscillated for 60 minutes (refer the result from the previous study). The correlation between pH and adsorption efficiency was analyzed using one-way ANOVA.

The characteristic of Industrial wastewater obtained from the company was measured using ICP (Inductively Coupled Plasma), including pH, chemical oxygen demand (COD), Total Dissolved Solids (TSS), NH_3, and initial concentration of heavy metals. Furthermore, 0.5 g of dried *S. crassifolium* was put into 50 mL wastewater and stirred for 60 minutes using a magnetic stirrer at 200 rpm. The solution pH was adjusted to 2, 3, 4, 5 and 9 with HCl (0.1 M) and NaOH (0.1 M). Then, the solution was filtered with Whatman paper BF/C code and the filtrate was analyzed using ICP to determine the metals concentration at 543 nm wavelength.

Metals uptake and removal were calculated as the difference in the metal concentration(s) before and after sorption [26], according to Eq. (1-2).

$$q = (\boldsymbol{C_i} - \boldsymbol{C_f})\ x\ \frac{\boldsymbol{V}}{\boldsymbol{M}} \qquad (1)$$

$$R = \frac{\boldsymbol{C_i} - \boldsymbol{C_f}}{\boldsymbol{C_i}}\ x\ 100\% \qquad (2)$$

Where q =metal adsorption (mg/g); M = dry biomass (g); V= volume of the initial lead solution (L); C_i= initial concentration of lead in aquatic solution (mg/L); C_f = final concentration of lead in the aquatic solution (mg/L) at given time (t; min); R = removal percentage (%).

3. RESULTS AND DISCUSSION

The preliminary research of the present study had been conducted in an aqueous solution on various types of macroalgae, *Euchema spinosum, Padina minor* and *Sargassum crassifolium* on Pb and Cr adsorption using various initial concentrations of 200, 300, 400, 500, 750, and 1000 mg/L and 7 variations of contact time, namely 10, 20, 30, 45, 60, 90, and 120 minutes [24] at pH 5 [9]. Based on the study, *S. crassifolium* achieved 97-98% (Fig. 1) as the highest adsorption of Pb at 60 minutes contact time (Fig. 2). This was higher and relatively faster than the study of Sweetly et al [9] which achieved 89.75% removal. This is due to the high availability of active sites to bind adsorbent such as -COOH, -OH and -NH2 [27]. The ability of heavy metal adsorption by *S. crassifolium* tends to decrease after 60 minutes oscillation because the metal binding on the active site has already saturated.

The highest Cr adsorption by *S. crassifolium* was 99% achieved at 30 minutes contact time. *Sargassum crassifolium* showed the fastest time to

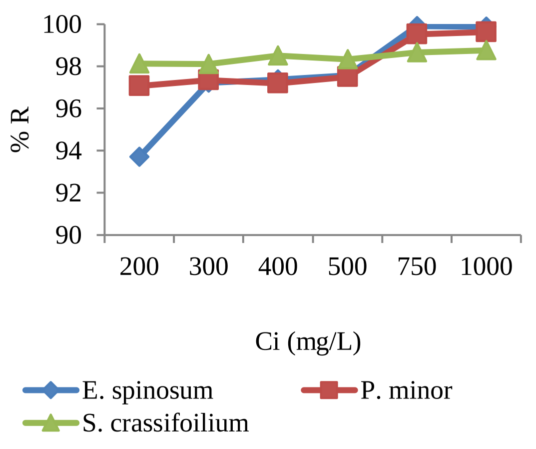

Fig. 1 Adsorption percentage of heavy metal in an aqueous solution at different initial concentration [24]

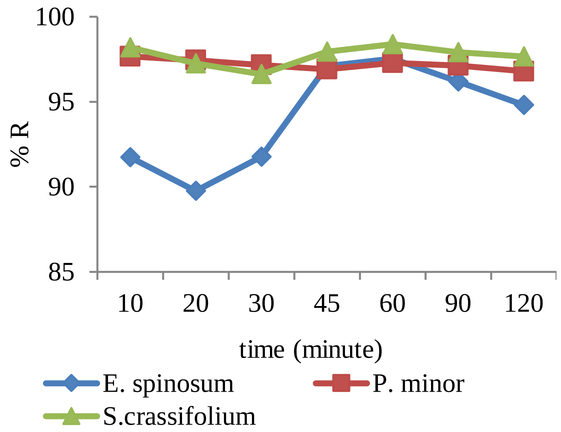

Fig. 2 Adsorption percentage of heavy metal in aqueous solution at various contact time [24]

reach the highest adsorption percentage than *E.*active sites (carboxyl, amine, and hydroxyl) on the surface of the *S. crassifolium's* epidermis.

Therefore, of the previous study' results, *S. crassifolium* belonging to *Phaeophyta* division could be claimed as the most suitable adsorbent for lead supported by a stronger form of the cell wall, predominantly alginic acid or alginate, compared to *E. spinosum* (*Rhodophyta*) and *P. minor*. Macroalgae *S. crassifolium* was able to absorb 97-98% of lead (Fig. 1) and capacity of adsorption 55.56 mg/g fit Freundlich model (Tabel 1). This was assumed *S. crassifolium* had high adsorption capacity on heavy metal. Other studies have also supported by these findings [8,9,15,17,21,23,25].

The observation was continued using industrial wastewater to analyze the capability of macroalgae *S. crassifolium* on heavy metals adsorption, in various pH, 2, 3, 4, 5 and 9 for 60 minutes oscillation as the optimal contact time refer to the previous study.

The pH value is one of the most important parameters in an experiment on a biosorption.

Table 1 Constant Value of Langmuir & Freundlich Isotherm

Adsorbent	Langmuir		
	a	b	R^2
E. spinosum	32.26	0.0046	0.984
P. minor	40.00	0.0026	0.934
S. crassifolium	55.56	0.0008	0.882
	Freundlich		
	k	n	R^2
E. spinosum	3.322	2.841	0.892
P. minor	4.846	1.390	0.862
S. crassifolium	2.453	1.379	0.976

Source: [24]

Various studies have been conducted but using different biomass, such as Sweetly et al [9] using *Sargassum*, while Vimala and Das [13] using mushrooms as an adsorbent. The concentration of hydrogen ions in adsorption is one of the important parameters affecting the ionization rate of the adsorbent during the reaction and the replacement of positive ions in the active site [22].

The industrial wastewater used was assumed containing a high concentration of heavy metals as end-product of the industrial process. The wastewater samples obtained from the company of hazardous wastes treatment in Indonesia is a mixture of wastewater solution from various industrial companies. Thus, it absolutely contains a lot of heavy metals which have specific biochemistry characteristics and should be given serious attention from all stakeholders.

The types of heavy metals contained in the industrial wastewater were Cd, Co, Cr, Cu, Fe, Hg, Ni, and Pb with initial pH 7.91. Several parameters, including pH, COD, TDS, NH_3, and heavy metals from the industrial wastewater were also analyzed using standard methods (APHA-AWWA) [28]. The initial concentration of the various parameters for the untreated industrial wastewater is exhibited in Table 2. The untreated concentration was then compared to the treated industrial wastewater with *S. crassifolium* is presented in Table 3. In order to determine the safe limit of wastewater discharged into the environment, the Government Regulation of the Republic of Indonesia No. 82/2001 [6] is used as a guideline.

Based on the regulation, most of the concentration of the heavy metals from the sample of industrial wastewater in various pH were above the threshold limit, but not too far away, except Cd, Co, Hg and Pb were safe to discharge to the

Table 2 Characterization of the untreated industrial wastewater

Parameter	Initial Concentration (mg/L)
pH	7.91
COD	8160
TDS	18890
NH_3	656
Heavy metals:	
Cd	0.10
Co	0.0505
Cr	0.41
Cu	0.102
Fe	2.69
Hg	0.27
Ni	1.1125
Pb	0.04

Table 3 The concentration of heavy metals after treated by *S. crassifolium*

Heavy Metal	Treshold Limit (mg/L)*	Cf (mg/L)				
		pH 2	pH 3	pH 4	pH 5	pH 9
Cd	0.01	0.001	0.001	0.001	0.001	0.01
Co	0.2	0.026	0.027	0.031	0.036	0.043
Cr	0.05	0.252	0.269	0.35	0.412	0.403
Cu	0.02	0.044	0.045	0.088	0.481	0.058
Fe	0.3	2.608	1.546	1.3	1.186	1.169
Hg	0.001	0.008	0.0072	0.006	0.0065	0.002
Ni	0.003	0.62	0.619	0.69	0.805	0.916
Pb	0.03	0.033	0.01	0.026	0.031	0.046

Note: *: Based on Government Regulation of Republic Indonesia no 82/2001

environment (Table 3). Although the final concentration on those metals was still above the regulated threshold limit, the adsorption efficiency of those metals was quite good reaching 37.78-56.86% removal (Cu>Fe>Ni>Cr). However, in terms of environmental quality, this condition is not good and needs serious attention especially on treatment process, including pH, type of adsorbent, and contact time to get an optimum heavy metals adsorption by the adsorbent.

Regarding the adsorption capability of heavy metals by *S. crassifolium*, the percentage removal of Cd, Hg, and Pb from industrial wastewater was the highest, reaching 75-99.05%, mostly in acidic pH range, except Hg were in alkaline pH, as shown in Fig. 3. It further was followed by Cu>Fe>Co>Ni>Cr was found 37.78-56.86% removal in an acidic pH range, as similar as reported by Davis et al [15]. It is was assumed due to the dissolution of some cytoplasmic components or ions, such as carbonates released into the solution and anion types, similar to the study by Latinwo et al [29] for chromium uptake.

The carboxylic groups in the brown algae are generally the most abundant acidic functional group, such as in *S. crassifolium.* They constitute the highest percentage of binding sites (more than70%), especially in dried brown algal biomass [30]. Thus, the adsorption efficiency of the brown algae displays at pHs near the apparent dissociation constant of carboxylic acids as the majority of metals of interest (i.e. Cd, Co, Cu, Fe, Ni, Pb) [15], which is similar to this study result.

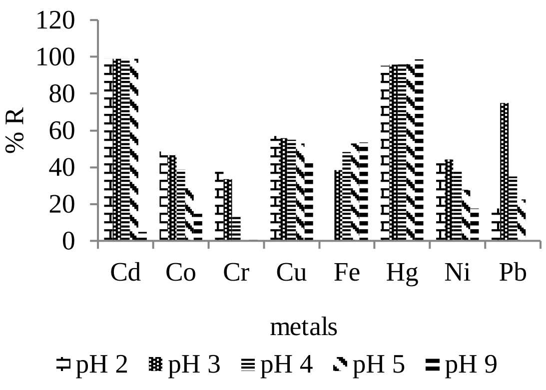

Fig. 3 Removal percentage of heavy metal on industrial wastewater

The optimum removal of those metals from the industrial wastewater was completely achieved at 60 minutes contact time which was contrary to the review by Davis et al [15] in red and brown macroalgae which were achieved in 180 minutes, although the adsorption efficiency was almost the same. The contact time did not show significantly correlated, as reported by Putri [24], however, it has a very significant effect on the cost of the industry for the processing of waste, including electricity and tools use. Besides, the metal hydrolysis constants, the ion size (ionic radius) and electronegativity have to be considered for the affinity of the adsorbent for adsorbing heavy metals [31].

Each type of heavy metal has the ability to bind with the cationic group on different macroalgae active sites depending on the chemical properties of the heavy metal and is highly dependent on the acidity (pH) conditions. The highest adsorption capability of *S. crassifolium* was achieved at range pH 2-5 for Cd, Co, Cr, Cu, Ni, and Pb, which similar to Sudiarta and Diantariani [22] and Susanti [32], except for Hg and Fe which was removed as much as 98.81% and 53.48 from the solution at pH 9. This is similar to the study reported by Cossich et al [33] for Cr, Latinwo [29] for Cr and Fe, and Davis et al [15] for Cd, Pb, Ni, and Cu. However, the affinity of the adsorbent for adsorbing Hg from industrial wastewater is quite rare studied. Huang and Lin [8] studied Hg adsorption by *Sargassum* in an aqueous solution which found pH 8-10 was the optimum adsorption capability with 70% removal of heavy metals. Another study was reported for red algae which could adsorb 99% Hg at pH 7-10 [34]. Similar to this study, the adsorption efficiency of Hg by *S. crassifolium* had almost reached 98% removal from industrial wastewater at pH 9.

The studies reported above were tested in the aqueous solution which was differed from this study trialed in industrial wastewater. In this study, several heavy metals were removed from industrial wastewater below 56.86%, only Cd, Co, Hg, and Pb with 75-99.05% removal. Nevertheless, all the heavy metals were safely discharged to the environment because the concentrations were still below the threshold limit in accordance with the regulation [6]. The different adsorption capacity was due to the affinity of adsorbent to adsorb heavy metal which was related to the cationic binding system. At acidic pH (pH<5), it had a positive surface charge and electrostatic repulsion from a number of H^+ ions and cationic species caused a lower attachment of metal ions. On the contrary, the adsorbent's sites carried negative charges at alkaline pH (pH>5) which favoring metal cationic complexes attachment.

Thus, it can be confirmed that this experiment considers selective removal of heavy metals from industrial wastewater, in suitable pH ($p<0.05$) and contact time, in order to reach the optimum removal percentage. The safe concentration of heavy metals discharged into the environment, especially into the waters, is an important point because it can interfere with the life of living organisms and ultimately lead to death. That is why the international institutions develop standardization for food consumption that could expose through food to human through the food chain in nature. Those international institutions are the World Health Organization (WHO), Food and Agriculture Organization (FAO), European Medicine Agency (EMA) and Joint Expert Committee on Food Additives (JECFA) providing the guidelines of tolerable daily intake on exposure of heavy metals for the human being based on body weight (Table 4).

Table 4 Tolerable Daily Intake (TDI) for heavy metals

Metals	TDI (mg/50-60 kg bw)		
	WHO/FAO [35]	EMA [36]	JECFA [37]
Cd	0.06	-	1
Co	-	-	-
Cr	0.05-0.2	0.3	-
Cu	3	0.05	-
Fe	0.8	0.26	0.8
Hg	0.005	-	1
Ni	1.4	0.3	-
Pb	0.214	-	2

The concentration of heavy metals in food is the important information that has to be obeyed by all stakeholders in the industries. Foods from the fishery products are mostly exposed to pollutants in the waters, which are mainly derived from industrial wastewater. Many studies reported contamination of heavy metals in the organism [38-40], in sediments and waters [40,41]. They reported that industrial wastes contribute to water pollution and is responsible for solving that problem.

In connection with the results of this study, the final concentration of heavy metals in the wastewater after the treatment by *S. crassifolium* was relatively safe for Cd, Cu, Hg, Ni, and Pb in accordance with TDI guideline, except for Cr and Fe. Therefore, the treatment of industrial wastewater is an important process to conserve the aquatic ecosystem which further has a serious impact on human health. The regulations set by the government have to be obeyed by all industry stakeholders and prioritize law enforcement for those who violate.

4. CONCLUSIONS

Sargassum crassifolium from the Phaeophyta division showed the best ability to selectively adsorb 37.78-99.05% of heavy metals from industrial wastewater at pH 2-3 or 9 and contact time of 60 minutes. The highest adsorption capability was 75-99.05% for Cd, Hg, and Pb. Most of the heavy metals are still above the standard, both by the Government Regulation of the Republic of Indonesia No. 82/2001 and TDI standard, however, it exhibits high adsorption efficiency.

This finding is useful for the industries which produce hazardous wastewater in the production process. This study is able to reduce the cost and time of wastewater treatment efficiently, although the selectivity of the heavy metals adsorption is one of the limitations of this study. However, the slightest reduction of a pollutant from industrial wastewater is very influential on environment condition and will give a significant contribution to the conservation of living organisms in the ecosystem sustainably.

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