RESEARCH ARTICLE | MAY 25 2023

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Household Waste Bioadsorbent for Removing Heavy Metals and Textile Dyes: A Review

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Abstract. Heavy metals and textile dyes are one of the wastes that became an environmental pollution. Meanwhile, another source of pollution is household waste including organic waste. An option to manage pollution by heavy metals and textile dyes is by an adsorption process where the adsorbent used is made with household waste raw materials, namely bioadsorbent. This method is considered effective because their advantage is ability to overcome pollution caused by heavy metals and textile dyes, it also contributes to the utilization of household waste into products that have higher economic value.

INTRODUCTION

Hazardous chemical compounds such toxic heavy metals and dyes contains in wastewater bring seriously problem to environmental as a source of pollution in aquatic ecosystems and human health. Various methods were used to decrease heavy metal and dyes in wastewater. These methods were include chemical precipitation, electrolysis, adsorption, photocatalysis, coagulation-flocculation, ion–exchange, photochemical oxidation, membrane separation [1,2]. Adsorption method has been suggested more effective than chemical or physical methods. Adsorption processes was successfully decreased contaminants such as toxic heavy metal and dyes. Materials that shows high performance in the of heavy metals and dyes in wastewater contaminants treatment are activated carbon [3], silica [4], and bioadsorben [5].

Bioadsorbents could considerate as new materials to adsorb heavy metal and textile dyes in the waste water. The advantage of bioadsorbent are due to the consideration of sustainable, organic and ecofriendly characteristic. The new bioadorbent feedstocks have been reported in recent years such as coffee waste, banana peel, tea waste [6]. Bioadsorbents from household waste can easily found, low cost, and mostly free of charge.

The aim of this work is to evaluate effectiveness of present works on the development of bioadsorbent to remove heavy metals and textile dyes from wastewater, especially bioadsorbents that prepared from household waste. Another advantage by converting household waste to raw material of bioadsorbents is decreased the food wastes and create better environment.

Bioadsorbent

Bioadsorbent was defined as adsorbent that their source came from natural, biological and derived waste. Bioadsorbents are the materials derived from different biological sources such agricultural by-products and household waste Various bioadsorbent feedstock from household waste can be found in TABLE 1.

> *3rd International Seminar on Science and Technology (ISSTEC) 2021* AIP Conf. Proc. 2720, 040027-1–040027-7; https://doi.org/10.1063/5.0136878 Published by AIP Publishing. 978-0-7354-4533-8/\$30.00

040027-1

No	Household waste	Heavy metals	Dyes	% adsorption	References
1	Banana Peel		Methylene Blue Methylene Orange	28 % - 92%	[7,8]
2	Egg Shell		Brilliant Green Congo Red Methylene Orange	41%	[9,10,11]
3	Orange Peel		Deazol Black B EAN Brilliant Green	60% - 70%	[12,13]
4	Tea	Fe Cr IV)		72% - 94%	[14,15]
5	Fish scale	Cr III) Zn		99 %	[2,16]
6	Coffee	Pb II)		99 %	[5, 17]

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The use of household waste has shown many advantages in removing environment pollutants from waste water. Household waste commonly used for preparation of bioadsorbent for removal heavy metals and textile dyes. Due to its ability and high efficiency also easily and freely available.

The adsorption of textile dyes using banana peel has been reported by Lantang *et al* [7] with acid treatment. H_2SO_4 was used for the acid treatment. The adsorption of methylene blue was studied at different parameters such as contact time, efficiency, and temperature, with the highest adsorbent capacity 92,63 % and 90 minutes of contact time. Rahadi [8] prepared the banana peel without any modification. The maximum efficiency were found to be 28 % for methylene orange removal with 120 minutes of contact time. The high amount of bioadsorbent dosage in adsorption will decrease the dye concentration. The usage of acid in the prepared banana peel bioadsorbent could expand the adsorption surface. As shown in the result, bioadsorbent with acid activator reached higher efficiency for dyes removal.

Egg shells were also used to eliminated textile dyes. It is an excellent bioadsorbent for removal such cationic dye in aqueous solution. Reported by Kobiraj that the adsorption efficiency using eggshell were found at 99 % and 15 minutes contact time in brilliant green solution [9], while Ali Zulfikar obtained an absorption efficiency of 93 % for treating Congo Red with 20 minutes contact time [10]. Another result is for methylene orange dyes removal conducted by Nurlaili. Efficiency were found at 41,46 % with 60 minutes of contact time. Many of the bioadsorbents from egshell were prepared without any acid or alkaline treatment. However, the results found were highly effective to remove dyes. This can be influenced by how the bioadsorbent prepared and treated also other compound which is contained in the raw material [11].

The work by Abdurrahman showed that the removal efficiency of Deazol Black B EAN reactive dye using orange peel was 60-70% with a contact time of 120 minutes without using any activator [12]. Different experimental results were obtained on orange peel modified with H_3PO_4 as an activator, leading to the highest increase in the efficiency of brilliant blue removal of 96 % with 30 minutes of contact time [13]. The advantage of adding the activator into prepared bioadsorbent is for helping to increase the ability of the bioadsorbent to adsorp the dyes in the water contaminant solution and shorten the contact time. Both results showed higher efficiency in the orange peel bioadsorbent prepared with H_3PO_4 . Apart of being an activator, the use of H_3PO_4 is for removing volatile compound in bioadsorbent surface.

Application of tea waste bioadsorbent in the presence of HCl could achieve 94.25% during 30 minutes contact time which reliably better bioadsorbent for removing ferrum. [14] While Abriagni observed the maximum adsorption efficiency was 59,46 % for chromium removal, found at 45 minutes of contact time and without using any modification. Bioadsorbent prepared with activator could remove higher percentage of heavy metal than the one prepared without activator [15]

Fish scale as bioadsorbent was possbile for removal of heavy metals from wastewater. It was found that fish scale prepared from H_2SO_4 and found that the chromium adsorption efficiency of 99,7518 % at 90 minutes contact time [2] While fish scale prepared from HNO₃ for zinc removal, observed the maximum adsorption efficiency of 92,3 % and 3 hours contact time [16]. Both of the results showed high efficiency of chromium and zinc removal and conducted at lower pH 5 to 6 because of the acid and alkaline activator. The application of these bioadsorbent can be customined based on the activator and pH parameter used in fish scale bioadsorbents preparation.

Another studied of the biodsorbent is using coffee waste coated by Fe_3O_4 for adsorption purpose. The addition of magnetic material helping the increase of adsorption efficiency. By increasing the Fe_3O_4 load, the surface of the

bioadsorbent will be coated with magnetic nanoparticel. They reported 99,29 % adsorption efficiency of plumbum and be the most effective bioadsorbent among the other coffee waste bioadsorbent [5]. While Ayucitra found the plumbum removal in the presence of HCl and found the maximum adsorption efficiency of 51,6 % at 3 hours of contact time. The addition of magnetic material as an activator into the coffee waste bioadsorbent preparation process is better than the acid activation based on the efficiency removal of the lead [17].

The efficiency results obtained can be influenced by raw material, activator, pH, contact time, bioadsorbent dosage, adsorbat concentration and also pre-treatment. Each of the bioadsorbent prepared have different result. It can be use for further research study by variating and modificate any other material to obtain higher efficiency.

HEAVY METAL ADSORPTION

Mechanism of Heavy Metals Adsorption

Different mechanisms were involved in adsorption process. There are physical and chemical adsorption, ion exchange, reduction, surface precipitation, and etc. Physical adsorption is about the interaction between the adsorbate and the surface of the bioadsorbent by Vander Waals forces. While chemical adsorption comprise ionic exchange, chemical precipitation of functional groups that occur on the surface of bioadsorbent

Effect of Bioadsorbent Dosage

Tea

The work on the tea waste bioadsorbent with HCl acid activation for ferrum removal. According to their paper, the adsorption efficiency goes up to 94,25% during 30 minutes contact time for the tea waste bioadsorbent dosage range of 5, 7,5 and 10 grams [14]. Different variation of tea waste bioadsorbent dosage used to remove chromium. The highest chromium absorption percentage can be obtained that is 59,46 % with bioadsorbent dosage range of 0,2 g to 1 g in the 50 ppm of heavy metal concentrations during 45 minutes contact time [15]

Fish scale

It was reported that a high adsorption efficiency in a study where they removed chromium 99,7518 %). The initial concentration was 150 mg/L chromium ion, and biadsorbent variation dosage from 0.4 g to 1 g, the experiments were conducted at 90 minutes contact time [2]. While the effects of adsorbent dosage on the zinc removal at 3 hours contact time was investigated in 0,020 g amount of bioadsorbent and could remove 85,4 % chromium [16]

Coffee

Edathil studied coffee magnetic bioadsorbent by using Fe_3O_4 as activator. The adsorption efficiency to remove plumbum was found to be 99,29% with range 0-0,030 of bioadsorbent dosage during 30 minutes of contact time [5] Another result of coffee bioadsorbent for plumbum removal was studied, the highest lead removal percentage of 51.6 % and mass to volume ratio of coffee waste bioadsorbent and plumbum solution of 1:10. The adsorption was conducted for 3 hours contact time [17]

Effect of Contact Time

Tea

The contact time was investigated for 10, 15, 20, 25, and 30 minutes. The dyes sample of 250 mL and small amount of bioadsorbent kept in a magnetic stirer with 5 minutes for each dosage of bioadsorbent. The optimum time duration required for colour removal is 30 minutes [14]. While another study using 50 mL dyes sample and contact time variation for 15, 30, 45, 60, 75, and 90 minutes. Optimum contact time found during 45 minutes contact time [15]

Fish scale

Activated fish scale bioadsorbent was achieved at a medium amount of dosage and contact time range of 30–120 minutes with 100 mL of shynthetic chromium solution. High adsorbent efficiency found at 90 minutes contact time [2]. While Zayadi conducted at 3 hours of contact time using small amount of fish scale bioadsorbent and 9.771-11.117 metal ion concentration [16].

Coffee

Variation of contact time studied by Edathil are 0 to 240 minutes. Reached high efficiency at 90 minutes optimum contact time with 99,29 % removal of lead [5]. While Ayucitra found optimum time at 30 minutes. Adsorption process between bioadsorbent and adsorbat was carried out in a shaking water bath [17].

Mechanism of Textile Dyes Adsorption

The mechanism of the dye adsorption process can be explained by assuming that as a determinant of the reaction rate is a chemical adsorption process which includes inter-valence forces or electron exchange between the adsorbent and the adsorbate. This means that as a determinant of the rate of the adsorption process, the uptake process in the form of chemical-physical adsorption with the slowest uptake process, namely during the adsorption process, there is an electrostatic interaction between positively charged methylene blue dye and negatively charged activated carbon. Intra-particle diffusion depends on several factors such as sorbent structure, physical properties of sorbent and sorbate, chemical properties of sorbate, system interactions and system conditions. The mechanism of intra-particle diffusion is based on two mechanisms, pore diffusion and solid diffusion. Pore diffusion depends on solvent transport and the internal structure of the sorbent pores. This mechanism explains the diffusion of the sorbate molecules into the particles in the liquid and in the pores of the liquid [18].

Effect of Bioadsorbent Dosage

Banana peel

It was reported that 0.01 gram to 0.10 gram methylene blue introduced by 5 ppm of banana peel bioadsorbent that activated by H_2SO_4 could remove 92.63 % of the color during 90 minutes adsorption process [7]. While banana peel bioadsorbent without activation process with the same concentration could remove 28% of the methylene orange during 120 minutes adsorption process [8]

Egg shell

The adsorption of the dye on eggshell was studied by varying the eggshell dosage from 0.4 g/L to 6 g/L introduced by 50 mL of brilliant green solutions of 100 mg/L concentration. Almost complete removal of dye at 4 g/L dosage of egg shell bioadsorbent. For congo red removal, different bioadsorbent doses were used 5 g to 30 g). It could remove 93 % of the dyes in the dye concentrations of 20 mg/L. Different amount of bioadsorbent dosage used in removal of methylene orange. It was reported that the use of 7 g to 13 g of bioadsorbent dosage introduced by 20 ppm dye concentration [11].

Orange peel

The experiment was conduct with variation of orange peel bioadsorbent which is increased from 0.5g to 1.5g introduced into 25 ml solution. It was found that optimum amount is 1.5 g of adsorbent. Adsorption efficiency was increased with the increasing of adsorbent dosage and could remove 60 % to 70 % dye in water solution [12]. While another study reported on the use of orange peel as source of bioadsorbent using 0,2 g to 1 g for brilliant blue dyes removal. It could remove 96 % of dyes in dye concentration of 125 ppm [13].

Banana peel

The contact time on a bioadsorbent made from banana peel was studied in 10, 30, 60, 90, and 120 minutes. The optimum time for colour removal is 90 minutes in the dyes sample of 5 ppm[7]. While another studied reached optimum contact time is 120 minutes [8].

Eggshell

Experiment for removal brilliant green dye observed by Kobiraj were carried out in a mechanical stirrer. Optimum contact time found during 15 minutes adsorption. Variation of contact time is 5 to 40 minutes with 5 minutes interval in 50 mL of dye solutions [9]. For congo red removal, variation of contact time is 5, 10, 15, 30, 45, 60, 75, and 90 minutes. The optimum time was found at 20 minutes in 20 mg/L congo red solution [10]. While another study conducted to remove methylene orange. They reported that at 60 minutes of contact time removed higher toxic dye with an initial dye concentration of 20 ppm within contact times from 0 to 100 minutes with 20 minutes [11]

Orange peel

Variation of time for reached high maximum removal of Deazol Black B EAN reactive dye is 20 to 160 minutes with 20 minutes of time interval. The maximum adsorption was found during 120 minutes of contact time [12]. While different maximum contact time was investigated at 30 minutes contact time with 10, 30, 50, 70, 90, and 180 time variation [13]

PREPARATION OF BIOADSORBENT FROM HOUSEHOLD WASTE

Banana Peel

Lantang prepared bioadsorbent from banana peels from species of *Musa acuminafe* sp. The peels were washed, sliced and sun dried for about 2 days. After drying, banana peel were carbonized at 400 °C for 1,5 hours and crushed into small pieces. Banana peel were re-dried for 1,5 hours at 110 °C after soaked into H₂SO₄ solution. The bioadsorbent were washed and dried again for 2 hours for use the purpose to remove of methylene blue removal [7]. While another study prepared unactivated banana peels bioadsorbent using *Musa acuminta* cv. Cavendish for methylene removal [8].

Eggshell

In study by Kobiraj, the eggshells washed and dried at 100 °C for 60 minutes. After drying, the eggshells were grinded into powder and washed again before redried in a microwave oven. The powder used for brilliant green dye removal [9]. The method of preparing eggshells as bioadsorbent remained the same as that used for congo red removal [10] and for methylene orange removal [11]

Orange Peel

Orange peel was used as an bioadsorbent to treat Deazol Black B EAN reactive dye. Waste peels were washed to remove dirt particles and sliced into small pieces for use as an bioadsorbent. These were sundry for 48 hours and grounded using mortar into powder size [12]. Simillar bioadsorbent preparation also prepared for brilliant blue dye removal. The orange peel were cleaned and crushed, then it was shieved into powder. Activator used in this preparation is H_3PO_4 with 1 : 1 ratio of bioadsorbent powder. The activated bioadsorbent chilled in desiccator for a night and heated at 600 °C in furnace for 60 minutes. After that, the bioadsorbent were washed and redried for 6 hours at 150 °C. [13].

A powder prepared from tea waste was utilized for ferrum removal. Tea waste were washed and sundried. To obtain powder size, dried tea waste was shieved into 200 mesh size. After that powdered tea was soaked overnight by HCl for the activation purpose [14]. The simillar work also studied for chromium removal and without adding any activation [15].

Fish scale

In study fish scale were washed and soaked for a day and sundry for 2 days. After sundrying, fish scale redried for a day at 65 °C for moisture purpose and grounded into a powder. Sulfuric acid used for the chemical activation by soaking the powdered fish scale for 2,5 hours and redried for 120 minutes at 80 °C [2]. Nitric acid also used for chemical activation in the preparation of fish scale bioadsrobent [16].

Coffee

Coffee bean waste were washed and dried for a day at 60 °C. The dried bean grinded into a fine powder with 250 μ m particles size. For chemical activation, Fe₃O₄ was used for higher adsorption purpose by soaking the coffee powder. After activating the bioadsorbent, it washed with deionization water for neutralization and redried until it reached constant weight [5]. Another preparation of coffee waste bioadsorbent was carried out. Powdered coffee waste were washed and carbonized for 90 minutes with 400, 500, and 600 °C temperature variaton. Hydrochloric acid was used for chemical activation by soaking the bioadsorbent for a day and then double-washed it [17]

CONCLUSIONS

Household waste were potentially source for bioadsorbent feedstock especially for heavy metals and synthetic dyes that used in textile. The research works that already done shows the efficiency of bioadsorbent is in a range between 80% to 99%. Some household waste was convert into active carbon and then with our without further treatement was use as bioadsorbent, while others household waste was dried and followed by activation step before applied as bioadsorbent. Mostly of the bioadsorbent that produce from household waste spend more than 60 minutes to achieve the highest performance in the adsorbtion process.

ACKNOWLEDGMENTS

This work was funded by Ministry of Research and Technology Indonesian Government, through Hibah Penelitian Terapan Unggulan Perguruan Tinggi (Hibah PTUPT) 2021 no .312/E4.1/AK.04.PT/2021

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