J. Mesin Vol. 29, No. 2, 2023, 117-131

A Review of Valorization of Okara and Okara Nanocelullose Derived from Soybean Industrial Waste for Biosorbent Applications

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Abstract. Okara, the solid waste from tofu production, holds abundant potential in Indonesia. This review explores its application as a biosorbent for wastewater treatment, specifically as aerogels and hydrogels. Wastewater poses environmental risks due to pollutants like heavy metals, organic compounds, and microbes. This review discusses okara biosorbent, either in its natural form or modified to obtain cellulose, that proves effective in binding pollutants. Nanocellulose-based okara biosorbent enhances the absorption of heavy metals and textile dyes, surpassing traditional biosorbents. Its low lignin concentration simplifies its process of obtaining cellulose, compared to other lignocellulosic materials.

Keywords: *aerogel; biosorbent; hydrogel; okara; wastewater treatment.*

1 Introduction

Soy-based food products have become one of the primary sources of protein for the Indonesian population. In addition to the low cost of soy-based foods, soy also offers good nutritional value. It is well known that soy is one of the legumes with a high protein content. Just from the husk itself, soy contains 29-51% cellulose, 10-25% hemicellulose, 1-4% lignin, 4-8% pectin, 11-15% protein, and other extracts [1].

According to data from the Indonesian Ministry of Agriculture in 2013, tofu is the most consumed protein source by the Indonesian population. Tofu production itself reaches a minimum of 2.560.000 tons per year. However, it is not widely recognized that the tofu industry significantly contributes to waste. Research by

Diterima 27 November 2023, Direvisi 13 Desember 2023, Diterima untuk dipublikasikan 15 Desember 2023

ISSN:0852-6095, DOI: 10.5614/MESIN.2023.29.2.2

Faisal et al. in 2016 revealed that small-scale tofu industries produce at least 20.000.000 m^3 of liquid waste and 1.024.000 tons of solid waste annually. This waste equals 1.000.000 tons of CO_2 gas emissions [2].

Solid and liquid waste from the tofu industry in Indonesia is still seldom utilized. Yet, tofu waste can be used for various high-value-added applications. Solid waste from tofu production, or okara, is commonly used as livestock feed due to its high protein content, and leaving okara exposed to open fields can produce unpleasant odors that pollute the environment. However, compared to other lignocellulosic materials, the application of okara is more versatile due to its extremely low lignin content (okara 1-4% [1], sugar cane bagasse 14-24% [3], and eucalyptus 27% [4]). Lignin is a major hindrance to the enzymatic hydrolysis of biomass [5]. Okara can also be processed into soil fertilizer [6] and food products.

One of the most fascinating applications of utilizing okara is one of the components in freshwater wastewater treatment. This benefit is undoubtedly exciting as wastewater issues are a global challenge that persists today. Wastewater is hazardous to the environment and living organisms due to its saturated salts, heavy metals, organic compounds, oil emulsions, dyes, and even microbes as pollutants that can cause various diseases. To address this issue, it is necessary to implement wastewater treatment for two primary purposes. Firstly, to efficiently recycle and reuse wastewater for human and other living organisms' consumption. The other is to guarantee adherence to environmental sustainability standards for wastewater discharge to minimize soil contamination [7].

Several methods can be employed to treat wastewater: chemically (chemical precipitation, oxidation, and reduction, electrochemical), mechanically (sedimentation, filtering), biologically (biofilters), or combination (coagulation, sorption, ion exchange, electrodialysis, reverse osmosis), etc. [8]. Additionally, adsorptions remain the most economical and efficient method as their concept and application are simple and flexible. Using biosorbents can efficiently remove both inorganic and organic pollutants [1].

Based on the previously explained issues, this review discusses the production of biosorbents by utilizing abundant waste in Indonesia, okara or soybean waste.

2 Composition of Okara

Soybean waste, known as okara, is a byproduct generally from the tofu and soymilk industry that generates tons worldwide annually. After curding and processing, every kilogram of soybean will produce 1.1-1.2 kg of fresh okara [6] that contains up to 84.5% moisture [9]. This review focuses on okara from the tofu industry, as shown in Figure 1. Okara contains up to 15.2-33.4% protein, 8.3-10.9% fat, 10.9% lipid, 3.8-5.3% low molecular weight carbohydrates, 42.4-58.1% dietary fiber including cellulose, hemicellulose, and lignin [9,10]. Other than that, every 100 grams of dry okara contains minerals, as listed in Table 1 [9,11,12].

Table 1 Minerals composition of dry okara in mg.

K	Na	Ca	Mg	Fe	Mn	Zn	Cr	P
935-1350	16.2-96	260-428	130-257	0.6-8.2	0.2-3.1	0.3-6.4	0.2	396-444

Okara reported having large amounts of hydroxyl and carboxyl groups in its components of cellulose and protein amino acids. This fact showed that okara is a good candidate for chemical modification, possibly being developed into biosorbent hydrogel and aerogel material [6]. These gel materials can be used for various applications such as soil supplements [6], tissue engineering, reduction of oil spills [13], and wastewater treatment [10,14].

Figure 1 Fresh okara from tofu production.

3 Biosorption using Okara Biosorbent

Biosorption refers to a metabolic-independent and passive process involving diverse interactions between a sorbate and a biological matrix called a biosorbent [15]. Biosorption is essential in numerous wastewater treatments. Evaluating the suitability of biosorbents is a crucial step in their selection. The use of biomass residue has gained significance for circular economic considerations. The environmentally friendly nature of biomass residue offers several benefits, addressing disposal concerns and providing economic advantages by generating revenue for various industries.

Many biomass residues can be used as biosorbents, including those based on cellulose [16], chitosan [17], algae [18], and okara [19]. The latter, okara-based biosorbents, are very interesting to develop, this biomass is very large in quantity, has functional groups that can bind heavy metal ions and organic pollutants, and is relatively easy to process, and can be used directly without further modification (Figure 2). Several types of pollutants that okara-based biosorbents can adsorb include heavy metal ions (Cd^{2+}, Zn^{2+}, Pb^{2+}) [19,20], organic pollutants (methylene blue, safranin orange, reactive brilliant blue, brilliant green) [14] and anionic pollutants (PO_4^{3-}) [21]. Another approach even utilized okara as it is without further modification, while others processed them to extract cellulose and nanocellulose content.

4 Preparation of Okara as Biosorbent

Some works have reported different approaches to fabricating okara-based biosorbents. Okara can be used as a biosorbent with or without further modification and functionalization (Figure 2). One of the advantages of okara is that it does not dissolve in water so that it can be separated after the adsorption process. To improve the performance of okara biosorbent, several studies reported modification and functionalization of okara, as seen in Table 2 and Table 3.

Hiew et al. [20] reported simple okara biosorbent preparation. Okara from industrial waste was rinsed and dried. The dried paste was blended to reduce it to smaller particles. The okara powder was then ground in an ultra-centrifugal mill and was screened through multiple sieves to obtain the biosorbent with particle sizes ranging from 125– 200 µm. Nguyen et al. [21] reported okara biosorbents loaded with metal salt to activate their capability to capture phosphorous. In this work, fresh okara was treated with NaOH, followed with Fe (III) or (Zr(IV)). More than one metal can be incorporated into okara biosorbent. Yu et al. [22] loaded the surfactant onto okara to increase its adsorption capability towards methylene blue. okara was impregnated in sodium dodecyl sulfate, and then dried okara was powdered and sieved (<0.25 mm).

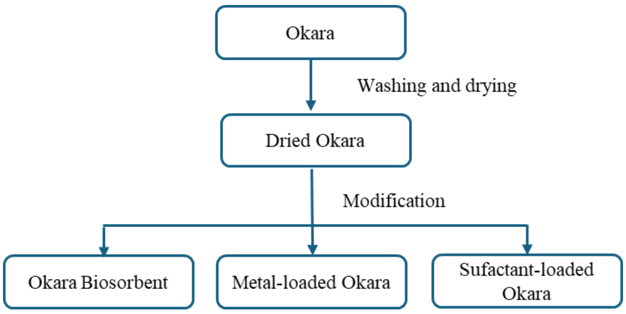

Figure 2 Okara utilization.

5 Heavy Metals Biosorption

Water contamination with heavy metals often comes from industries like battery manufacturing, metalwork, and plastic processing [23]. These heavy metals engage with active functional groups on the surface of the biosorbents. The biosorption mechanism includes the reduction of heavy metal ions, transforming them into less toxic forms. Table 2 shows several types of biosorbents and their capacity to bind heavy metals. Okara demonstrates moderate biosorption compared to other biomass biosorbents. It shows a biosorption capacity lower than rice husk, tomato waste, and pinecone but higher than banana peel and rice straw.

Table 2 Biosorbents from various types of biomass.

Biosorbent	Heavy Metal	Biosorption Capacity (mg/g)	Reference
Banana Peel	Pb (II)	0.5	[24]
	Cd (II)	5.71	[25]
Rice Husk	Cr (VI)	33.68	[26]
Tomato Waste	Pb (II)	152	[27]
Pinecone	Pb (II)	100.01	[28]
	Cd (II)	78.73	
	Cu (II)	33.55	
	Cr (VI)	57.36	
Rice Straw	Cd (II)	3.77	[29]
Okara	Cd (II)	14.80	[20]
	Zn (II)	16.31	[30]
	Pb (II)	11.13	[30]

Several studies have indicated that okara can be used as a precursor for the removal of pollutants; for example, okara was used to remove Cd^{2+} [20], Zn^{2+}, and Pb^{2+} [19] from aqueous solution [30]. The Okara-metal biosorption may involve mechanisms, such as ionic exchange, hydrogen bonding, and electrostatic interactions [20].

The effects of contact time, solution pH, biosorbent dose, and initial solution concentration were studied. The results showed that the optimum was at Cd^{2+} solution pH 6.0 and Zn^{2+} solution pH 7.0, with a solution concentration 50 mL. Okara biosorbent has an absorption capacity (q) of up to 14.80 mg/g of Cd^{2+} [20], 16.31 mg/g of Zn^{2+}, and 11.13 mg/g of Pb^{2+} [30]. On the other hand, approximately okara has biosorption efficiency around 88.93-89.75% Cd^{2+} , 69.69-71.39% of Zn^{2+}, and 95.18-99.91% of Pb^{2+}. The okara biosorbent also demonstrates regenerative capabilities. According to a study [20], despite a decrease in desorption efficiency from 89.21% to 32.56% after four cycles of regeneration using an HCl solution.

6 Organic Pollutant Biosorption

Okara is known to contain CHO, CO, and COOH functional groups on its surface, which are crucial in attracting different contaminants based on their surface charge. This property makes okara highly effective in eliminating heavy metals and organics such as azo dyes such as methyl orange, Eriochrome black-T, and rhodamine-B from textile industry effluents. Okara also has excellent removal capacity even at low pH (2.0), a distinct advantage compared to other biosorbents [14]. It has also been proven that okara effectively removes several organic pollutants with high adsorption capacity, as shown in Table 3. For example, Okara demonstrated adsorption of methylene blue up to 238.10 mg/g, while surfactant-modified okara increased the adsorption to 334.83 [22]. Okara also showed an adsorption capacity that reached 402.58 for Reactive Brilliant Blue KN-R.

Table 3 Okara biosorbent for organic pollutants.

Num	Okara Biosorbent	Pollutant	Biosorption Capacity (mg/g)	Reference
1	Okara and Sodium dodecyl sulfate activation okara	Methylene blue	238.10 and 334.83	[22]
2	Okara	Reactive Brilliant Blue KN-R (RBB)	402.58	[31]
3	Okara	Brilliant green	64.33	[32]
4	Okara modified with iron oxide nanoparticles	Cyanotoxin	2.84 µg/L	[33]
5	Acid treated okara	Acid Red 14 and Reactive Red 15	217.39 and 243.90	[34]
6	Okara	Methylene Blue and Safranin Orange	93.201 and 184. 592	[35]

7 Phosphorus Biosorption

Current research indicates the presence of phosphorus in okara, as shown in Table 1, suggesting a potential opportunity for phosphorus recovery from both okara and wastewater. Recognizing the limited capacity of untreated okara to remove phosphorus from aqueous solutions, modified okara was created and employed for specific purposes. Raw okara exhibits a low *q* of 0.08 mg P/g. The previous study with iron load okara (ILO) demonstrated *q* of 16.39 mg phosphate/g. The presence of iron (Fe (II)) significantly improved phosphorus capture, facilitated by carboxymethylation that allowed the attachment of Fe (II). Zirconium and Iron/zirconium-loaded okara showed higher adsorption capacity than ILO, reaching 47.88 and 40.96 mg/g, respectively (Table 1) [21]. Compared to other metal-loaded biomass biosorbents, these adsorption values are relatively high.

Table 4 Okara biosorbent in binding phosphorus(PO_4^{3-}) [21].

No	Okara Biosorbent	Biosorption Capacity (mg/g)
1	Iron loaded okara	16.39
2	Zirconium loaded okara	47.88
3	Iron/Zirconium loaded okara	40.96

Phosphate adsorption by metal-loaded okara-based biosorbents likely occurs through the ligand exchange mechanism. This mechanism involves a process where there is an exchange between PO_4^{3-} ions in the solution and OH^- ions coordinated with the metal ions loaded on okara-based biosorbents. The loaded metal ions can easily transform into hydrated forms due to the abundant presence of OH^- ions and H_2O molecules. During hydrolysis, H_2O molecules undergo deprotonation, releasing H^+ ions to generate exchangeable OH- ions. These OH- ions can then be replaced by PO_4^{3-} ions through the ligand exchange mechanism.

8 Okara Cellulose and Nanocellulose-based Biosorbent

As mentioned, okara contains a high amount of cellulose [10], a biopolymer composed of repeating β-1,4- glycoside-linked D-glucopyranose units [36]. Okara cellulose can be isolated via several processes; one example is shown in Fig. 3. Okara is treated by dispersing, drying, and washing several time to obtain its cellulose.

Okara can be used as nanocellulose precursors. The advantages of nanocellulose-based biosorbents include reduced adsorption time due to the nano-cellulose structure, low energy requirements for production, biodegradability, regeneration ability for reuse, and no generation of toxic sludge [1]. Nanocellulose is extracted from cellulose, resulting in less than 100 nm in size, as shown in Fig. 4 [37].

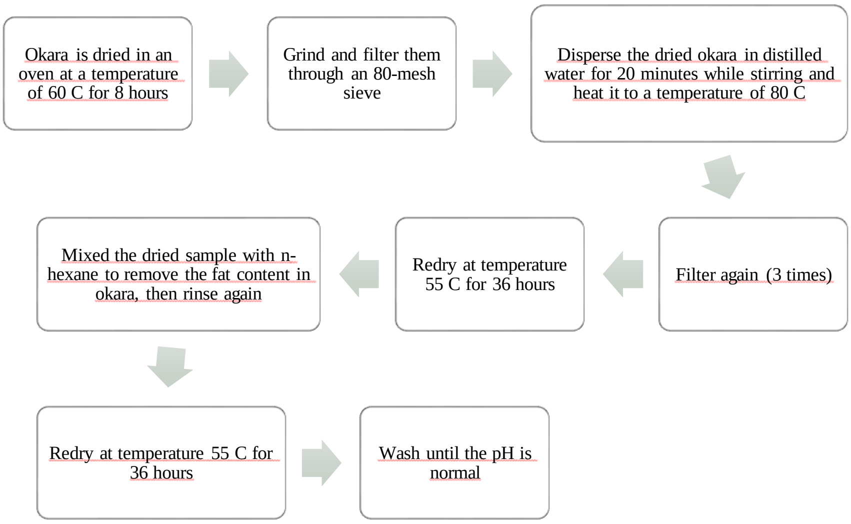

Figure 3 Isolation process of okara cellulose [36].

Nano-sized cellulose has significantly different properties compared to bulk cellulose. Nanocellulose is a sustainable nanomaterial with outstanding mechanical properties, biocompatibility, biodegradability, etc. The surface modification of nanocellulose can easily create new binding sites that provide specific characteristics for absorbing several types of pollutants. Surface modification of nanocellulose is emerging as a current and future research field in developing new biosorbents for environmental improvement [38].

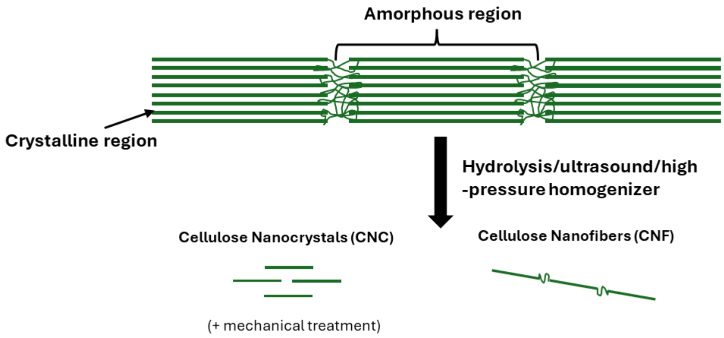

Figure 4 Nanocellulose structures.

Nanocellulose can be prepared via various routes, such as acid and base hydrolysis. Besides that, nanocellulose can also be produced using ultrasonic homogenizer underwater bath conditions to obtain nanocellulose [39]. Research conducted by Wu et al. in 2020 showed that ultrasonic and pressure homogenizers effectively disintegrate and disrupt thick cellulose fibers [36]. Nanocellulose that has undergone the sonication process is subsequently dried to obtain nanocellulose powder. Li et al. [40] prepared nanocellulose from okara to result in the form of nanofiber. They employed different mechanical homogenization, as shown in Fig. 6. It shows the formation of cellulose nanofiber with a diameter range of 5-45 nm.

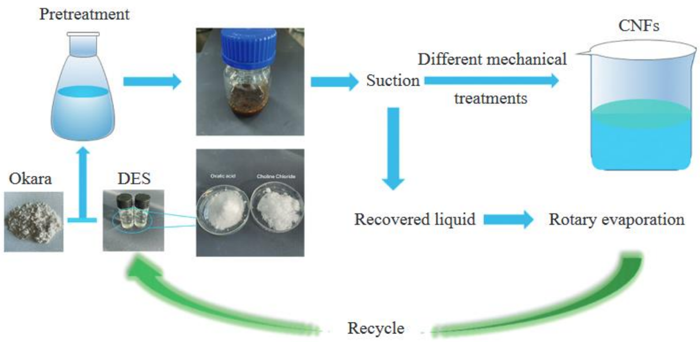

Figure 5 Isolation of cellulose nanofiber from okara [40].

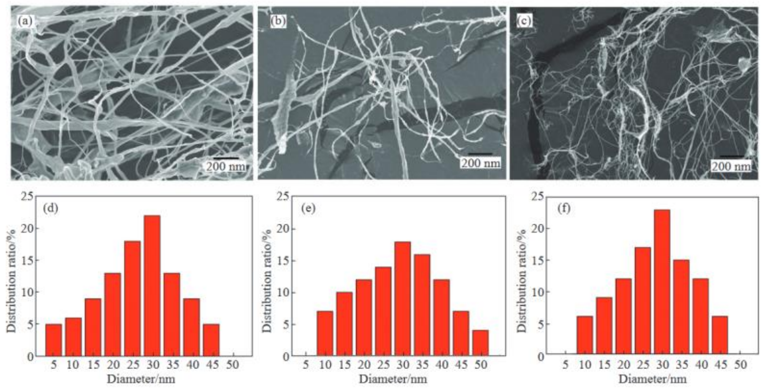

Figure 6 Morphology and diameter of okara cellulose nanofibers prepared by high-pressure homogenization (a, d), ultrasonic treatment (b, e), and high-speed stirring (c, f) [40].

In a study conducted by Cui et al. (2019) [10], FT-IR characterization was performed for samples of okara powder, okara cellulose, okara cellulose hydrogel, sigma cell cotton cellulose, and cotton cellulose hydrogel. All these samples exhibit the same absorption region at a wavenumber 3410 cm^{-1}, corresponding to the stretching of -CH3 groups, which are not found in the okara cellulose sample. The peak at 1744 cm^{-1} in the okara sample suggests the presence of C=O groups, which are also absent in okara cellulose. Additionally, the peak at 1530 cm^{-1} corresponds to the stretching vibration of C-C bonds in aromatic rings, which is not found in the okara cellulose sample. These three peaks are associated with lipids, proteins, hemicellulose, and lignin in okara.

Okara nanocellulose powder obtained from the process can be formed into two biosorbents: aerogel and hydrogel. Aerogel and hydrogel compromise a solid three-dimensional network that forms the structure and a medium inside it, while the medium of aerogel is air and hydrogel is liquid [41]. Nanocellulose-based aerogels have attracted significant interest lately because of their remarkable adsorption capabilities, environmentally sustainable potential, and cost-effectiveness.

The porosity of the fabricated nanocellulose hydrogels and aerogels is important. Pores in wastewater treatment have a role in adsorbing and absorbing pollutants into the gels and trapping them [42]. The porous nanocellulose aerogel base has been proven in the research conducted by Jiang and Hsieh in 2014 [43]. Fabricating okara nanocellulose biosorbents into aerogels or hydrogels will result in abundant advantages, as gels absorb vast amounts of liquids and contents. Other than that, nanocellulose gels-based biosorbents have high porosity, specific surfaces that can be adjusted according to our needs, high mechanical strength, low density, biodegradability, and hydrophilic [41].

Nanocellulose in gels has proven to have excellent adsorption capacity and increased effectiveness in binding compared to other biosorption materials due to the specific characteristics of nanocellulose, such as its high area specificity and the abundant amount of -OH groups present in its structure. These -OH groups contribute to binding pollutants and enable potential modification processes such as pollution remediation [44].

Ji et al. [45] reported that cellulose nanofiber aerogel efficiently captured diverse oils and organic solvents from water, demonstrating maximum absorption capacities reaching up to 108 g/g. Mo et al. [46] demonstrated that modified nanocellulose aerogel displays excellent absorption capability towards Cu (II), reaching an uptake capacity of 300 mg/g. Other research reported that biosorbents from other nanocellulose, such as hardwood kraft pulp and PVA aerogel, have the maximum capacity adsorption for Hg^{2+} (157.5 mg/g), Pb^{2+} (22 mg/g), Cu^{2+} (110.6 mg/g), and Ag^{2+} (24.5 mg/g) where these numbers were much higher than pure PVA aerogel [16].

Furthermore, okara showed cost-effectiveness as most of them were dumped and burned since there were no proper utilization techniques to process them. This abundant source is very effective for synthesizing value-added products, and since okara is considered a waste, the price will be lower than unprocessed soybeans [14]. Dumping and burning okara without any treatment can cause other problems, such as water and air pollution, since the waste contains a lot of organic substances [2].

9 Conclusion

Okara, the byproduct of tofu production, is potentially processed for various applications, including as a biosorbent in water through aerogels and hydrogels. Compared to other lignocellulosic materials, okara stands out due to its lower lignin content, enabling a quicker enzymatic hydrolysis cycle in obtaining cellulose. This ease of delignification makes okara versatile in applications. Okara-based gels potentially bind heavy metals and even other organic compounds produced by various industries. Repurposing soybean waste as a biosorbent adds value to agricultural residues and addresses environmental concerns, particularly in Indonesia, where okara waste is abundant.

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