



Aquatic Science and Fish Resources

<http://asfr.journals.ekb.eg>

Print ISSN: 2682-4086

Online ISSN: 2682-4108



Bioremoval of phenol from aqueous solution using Seaweed *Sargassum muticum* (Yendo) Fensholt

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ARTICLE INFO

Article history:

Received sept. 10, 2020

Received in revised form oct. 12, 2020

Accepted oct. 24, 2020

Available online oct. 31, 2020

Keywords

Sea weeds

phenol ion

bio sorbent

water pollution

sustainable development

ABSTRACT

In this study, *Sargassum muticum* (Yendo) Fensholt was evaluated as an economic and accessible biosorbent capable of removing phenol ions from aqueous solutions. The effects of the functional parameters, e.g., pH value of solution, dose of biosorbent, and initial concentration of phenol and contact time, were assayed in batch mode in the experiment. The optimum adsorption that allows for the displacement of phenol from aqueous solutions via *Sargassum muticum* was at the amount of 250 mg dry powder of *Sargassum muticum* for 100 mg/L of phenol at pH value of 3 after 24 hours of adsorption. This laboratory treatment strategy for the polluted water could be applied in a wide-scale treatment strategy.

1. Introduction

Water environment could be polluted by herbicides, pesticides, fertilizers, and other hazardous organic and inorganic chemicals according to an anthropogenic impact and human activities on the biosphere (Younis et al., 2014; El Zokm et al., 2015; Amin et al., 2018; Soliman et al., 2018; Younis, 2018, Nafea, 2019, Nafea, 2020).

95 chemicals are defined as toxic materials including heavy metals, oil, phenols, sulfate, nitrate, phosphate, and dissolved and suspended solids according to production volume, exposure, and biological effects which are released into water environment. (Asamudo et al., 2005; Said et al., 2006; Younis and Nafea 2012; Younis et al., 2018; Soliman et al., 2019; El-Naggat et al., 2019). Phenol and Phenolic compounds are one of the most hazardous water pollutants with an inclusive range of distribution and presence in different industrial operations' effluent, including petrochemicals, textiles, dyeing, phenolic resin manufacturing, and steel plant (Metcalf and Eddy 2003), it has highly toxic effects to most microorganisms, fishes, plants, and other animals besides its capability of causing severe environmental damage.

They are listed among priority organic pollutants by the US Environmental Protection Agency (Muftah et al., 2009). Moreover, they are of potential human carcinogenic and considerable health concern, even at a low concentration (Nayyef et al., 2012). Therefore, they must be displaced or treated from industrial wastewater before introduced to drainage systems. Various technologies aimed to experiment with removing and degrading phenol and phenolic compounds in wastewater including the following: solvent extraction (Ruey et al., 2009), adsorption (Frieda and Nava 1997; Saleh et al., 2019), chemical oxidation, activated carbon adsorption (Wang 1992) and biodegradation (Azin and Katayon 2002) however, all these methods are of immense disadvantages such as being costly, incomplete purification, hazardous by-products, and inefficiency and inapplicability to a limited concentration range and are often ineffective. Among the various methods available, biodegradation is economically and environmentally friendly (Pichiahet et al., 2008). Phenol's biological treatment is an important process in pollution prevention. Phenol's biodegradation has been assessed under aerobic (Rontaniet et al., 1999), and anaerobic conditions (Grossiet et al., 1998). Phytoremediation is beginning to garner interest as a passive, green, solar

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DOI: 10.21608/asfr.2020.42455.1007

energy-driven, and cost-beneficial method for cleaning up the environment when cross-referenced with physicochemical approaches. Even other biological methods (Rahul 2015, Nafea, 2019, Nafea, 2020) show promise when using different plants and their related microbes to degrade, collect, or incapacitate contaminants from water and soil (Dietz & Schnoor 2001). Phytoremediation is relatively inexpensive to utilize and easy to manage (Meagher 2000; LeDuc and Terry 2005). The materials of plants used in phytoremediation can be refashioned into bioenergy resources such as wood chips or pulp (Stanton et al., 2002). It is likely for phytoremediation to become popular since due to the innate ease of monitoring plants for effective performance (Nafea, 2019, Nafea, 2020). Catechols tend to damage DNA or destroy some proteins in the body and disrupt electrons transportation in energy transducing membranes. (Penny et al., 2017) Phenols, besides its compounds, are abundant in medicine, leather, paint, textile, oil refinery, disinfectants, and lubricant production wastewater industries (Kidak and Ince, 2006; Mous-savi et al., 2009). Skin absorbs phenol which creates the possibility of eye and skin burns following contact. Convulsions, comas, cyanosis, and death can be caused by being overly exposed to it (Busca et al., 2008; Gholizadeh et al., 2013). Therefore, wastewater containing phenol should not be introduced to open water before being treated (Hameed, and Rahman, 2008; Jia and Lua, 2008). Consequently, low-cost biosorption methods for the effective removal of phenols are quite lucrative. One of the most important topics of this research area is developing efficient methods to displace hazardous pollutants from wastewater (Jung et al., 2015).

Sargassum muticum (Yendo) Fensholt is a genus of brown (class Phaeophyceae) macroalgae (Seaweed) belonging to order Fucales. It usually grows in tropical and temperate oceans, generally inhabiting coral reefs and shallow water; it is commonly known for its planktonic (free-floating) species. Most species within this class are predominantly cold water organisms that utilize nutrients upwelling.

The major concern of this study is to investigate the efficiency of Sea weed Brown macro algae low-cost bio sorbent "*Sargassum muticum*" as Bio-material for removing phenol ions from aqueous solution. The effects of experimental parameters, such as pH of solution, adsorbent dosage, contact time and initial phenol concentration on the removal efficiency of phenol were investigated.

2. Materials and methods

2.1 Materials

The chemicals used for experimental were procured from Aldrich. The stock standard solutions of 1000 mg/L of phenol was prepared in twice-distilled water and placed in brown glass bottles at a temperature of -4°C. The solutions were assorted from an aqueous phenol stock-standard solution diluted with -twice-distilled water to the optimal concentrations.

The tested plant *Sargassum muticum* was collected from the eastern side of the Gulf of Suez (El-Tour) South Sinai.

Before being air dried, the collected *Sargassum muticum* was rinsed with freshwater and then tap water followed by distilled water. Then, the cleaned biomass was air dried for two days.

2.2. Batch adsorption studies

Batch adsorption experiments were conducted by soaking biosorbent *Sargassum muticum* into phenol-containing wastewater to evaluate pH values' effects, contact time, adsorbent dosage, and shaking speed on the efficiency of phenol removal from wastewater by the low-cost biosorbent *Sargassum muticum*.

To analyze the effect of contact time, the suspensions were agitated at optimal speeds via a mechanical shaker at room temperature. Batch experimental protocols occurred in various shaking times of 3, 6, 18, 24, and 30 hours at a pH value of 7.0 while maintaining biosorbent dosage (250 mg) and concentration of phenol (100 mg/l).

To evaluate biosorbent dosage's effect, batch experiments occurred via mixing various biosorbents masses of 10, 50, 125, and 250 mg of *Sargassum muticum* and the other parameters, a pH value of 7.0 and concentration of phenol, were kept constant.

Phenol's adsorption behavior for the exact initial value of concentration and equilibration time was analyzed as a function of pH. 250 mg of a given *Sargassum muticum* material was dispersed into 100 mL solutions containing 100 mg/L of phenol. The initial pH values were appropriated in a range of 3.0–9.0 by utilizing solutions of 0.1 mol/L of H₂SO₄ and 0.1 mol/L of NaOH. Afterward, and for 30 hours, the suspensions were shaken via a mechanical shaker at room temperature.

To assess the effect of initial concentrations, adsorption experiments occurred via shaking 250 mg of the biosorbent with various initial values of concentrations of phenol yielding 10, 50, 125, 250, and 500 mg/L for 30 hours. The supernatant was filtered through a 0.45 μm membrane filter to measure the phenol's final concentration in the solutions; the method used was colorimetric method using VIS-UV Spectrophotometer-19 (SCO-Tech, Germany) (Martin, 1949).

The sorption capacity *q* (mg/g) was obtained using the following equation:

$$Q_t = \frac{(C_0 - C_t)V}{M}$$

Where *C*₀ and *C*_t are the initial and final concentrations (mg/L) of phenol in the aqueous solution, respectively, *V* is the volume of phenol solution, and *m* is the weight of biosorbent *Sargassum muticum*.

3. Results and Discussion

After carrying out the laboratory work for determining adsorption capacity of brown algae for removing phenolic compounds from aqueous solution, four important parameters were investigated such as effect of adsorbent dose (mg), effect of contact time (h), effect of initial concentration and effect of pH of solution. These parameters were discussed in terms of their relation with the removal efficiency.

3.1 Effect of adsorbent dose:

The adsorbent dose is vital and should be appropriated according to the concentrations of phenol as the adsorption process is based on the idea of sufficient surface area for the adsorption of phenolic compounds. Increasing the dosage of adsorbent means more availability of surface area leading to exposure for binding with phenolic compounds. Sorbent dosage's effect on the adsorption of the appropriated phenolic compounds is explained in Figure 1. Removing phenol compound depends on the mass algal biomass existing in the solution, which surges with the accumulation of the adsorbent dosage, specifically quicker in the earlier stages. Then, it maintains constancy.

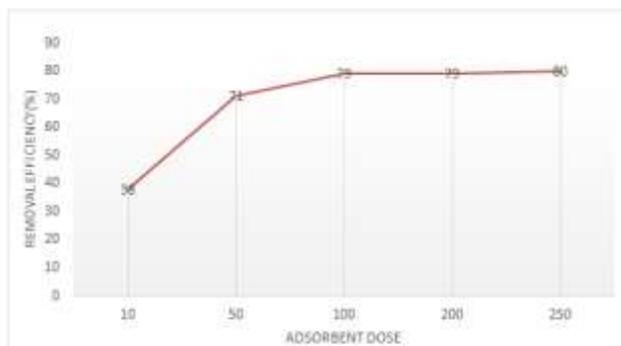


Fig (1): The effect of adsorbent dose on removal efficiency of phenol by Sargassum muticum

3.2 Effect of contact time

Increasing the dose of the adsorbent alongside increasing contact time while shaking the solution means more available surface area and exposure for binding with phenolic compounds. According to the results, contact time affects the sorption process; phenolic compounds' adsorption capacity surges with time where it remains constant at a specific time. After that, no more phenols are displaced from the solution. Phenolic compounds' required time to achieve equilibrium parameter was within 24 to 30 hours for Sargassum muticum. Related data is illustrated in figure 2. The phenolic compounds' sorption was evident in the initial part of the process. However, the process of biosorption on the algal biomass was different; equilibrium was achieved when the rate of sorption was equal to the rate of desorption rate, i.e., after 24 hours.



Fig 2. The effect of contact time on removal efficiency of phenol using Sargassum muticum.

3.3 Effect of initial concentration

The effect of initial phenol concentration is shown in Figure 3, as it shows the effect on phenol removal percentage and the adsorption capacity of Sargassum muticum for every

concentration at 20°C and a pH value of 7. The amount of adsorbed phenol diminished with more concentrated initial phenol values. This could be attributed to adsorption sites' saturation at higher amounts of concentration for phenol. Meanwhile, phenol adsorbed amount per a similar dose of Sargassum muticum surged with higher values of the beginning value of the concentration of phenol. The beginning value of phenol concentration allows for the nullification of all mass transfer resistance. Hence, a higher initial value of concentration of phenol facilitates the enhancement of adsorption capacity. A similar phenomenon was observed for the adsorption of phenol onto organobentonite (Ocampo-Perez et al., 2011; Moy, et al., 2012) and lignite-activated carbon. (Guocheng et al., 2011).

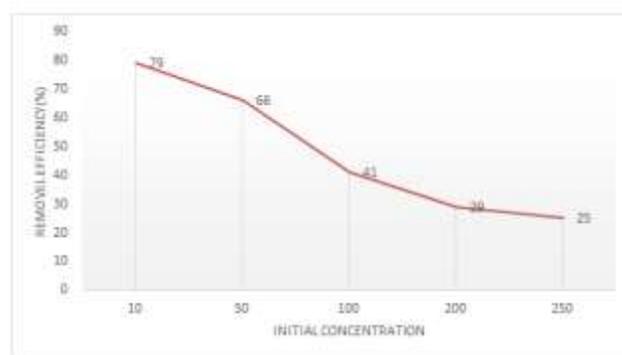


Fig. 3 The effect of initial conc. on removal efficiency of phenol using Sargassum muticum

3.4 effect of PH on removal efficiency

pH values critically affect phenol adsorption; a medium's pH value will control the magnitude of the electrostatic charges that are imparted by ionized phenol molecules. Consequently, adsorption rate fluctuates with the pH value of an aqueous medium. (Senturk et al., 2009; Sudharshi, et al., 2018). Figure 4 shows the effect of a solution's pH value on phenol removal by Sargassum muticum within a pH of 3.0 to 10.0. Phenol removal improves with pH values from 3.0 to 4.0, while it maintains pH values to be within 4.0–9.0. It sharply diminishes at pH values less than nine. The maximum value of adsorption is achieved at a pH value of 4.0. At pH value 2.0, multiple positive charges remain on Sargassum muticum's surface, exuding a large static force of repulsion. As pH increases from 2.0 to 4.0, the static repulsion force fades and the phenol adsorption surges. When the pH value is less than nine, phenol adsorption diminishes; this could result from the following reasons: (i) The negative charges on the surface of adsorbent surge with the pH value and phenol morphs from the molecular state to the ionic state, which creates force of repulsion amidst phenol ions and the Sargassum muticum significant. (ii) Sargassum muticum–adsorbed phenol ions also have a force of repulsion amidst themselves. (iii) The surface negative charges on Sargassum muticum are repulsive, repressing the disaggregation and adsorption of phenol ions. As the percentage of removal fluctuates within minimal a range of 4–9, pH 7, a phenol solution's optimal pH value, was accordingly handpicked as an optimum pH value for further experimentation on adsorption. (Guocheng

et al., 2011 and Asmaly et al, 2015). The beginning concentration allows for a critical driving force to nullify the effect of phenol's mass transfer between the aqueous and solid phases' drawbacks. A higher beginning phenol concentration will embolden the biosorption process.

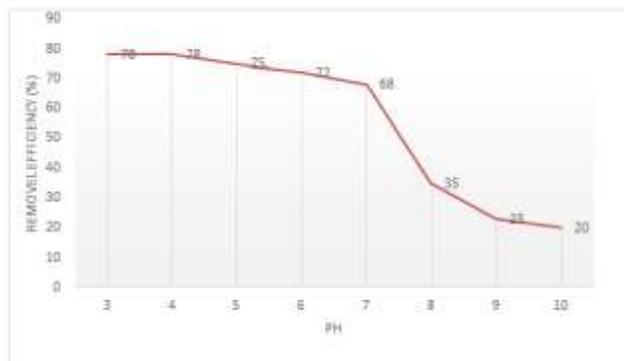


Fig. 4 The effect of pH on removal efficiency of phenol using Sargassum muticum

3.5 Adsorption capacity

The initial concentration provides an important driving force to overcome all mass transfer limitations of phenol between the aqueous and solid phases. Thus, a higher initial phenol concentration will enhance the biosorption process as shown in Figure (5) by the following equation:

$$Q_t = \frac{(C_0 - C_t)V}{M}$$

Where the C_0 is the initial concentration: C_t is the final concentration, V is the volume of the solution and M is the mass of the plant used

Adsorption capacity surged with the increases of the initial concentration values as in figure 5, where the number of receptors increased and were capable of accumulating more phenol ions on their surfaces (Younis and Nafea, 2012; Nafea, 2019).

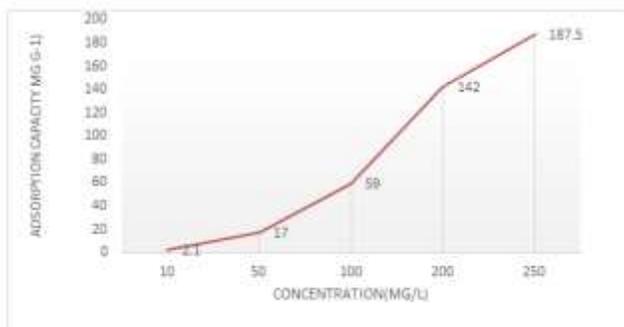


Fig. 5 show effect of initial concentration on adsorption capacity of phenol by Sargassum muticum

Conclusion

The results of this work report batch mode experiment for the removal of phenol from aqueous using Seaweed (Sargassum muticum). The results showed that adsorption efficiency of phenol was dependent on adsorbent dosage of Sargassum muticum, pH of solution, contact time and initial phenol concentration and the adsorption capacity increased with decreasing the initial phenol concentration in aqueous solution, indicating the saturation of the available active sites on the surface of Sargassum muticum

at higher initial concentration of phenol. It can be concluded that low cost bio sorbent (Sargassum muticum) can be used for the removal of phenol from industrial wastewater.

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