Shrimp Waste as an Effective Biosorbent – A Review

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ABSTRACT

Due to increased industrialization, a lot of effluents containing toxic chemicals have been generated and low-cost treatment is needed for the removal of these heavy metal ions. One such is the Shrimp industry that has been commercialized for more than a century producing huge waste and these wastes are detrimental to the environment if not disposed of properly. This waste can be efficiently used in biosorption. This has been an interesting area in recent days due to its ease of operation and flexibility. The purpose of this review is to present the significant role of shrimp shells as effective biosorbents for the removal of noxious heavy metals from effluents. The literature review has been done on the thermodynamic characteristics of heavy metal (HM), adsorption behavior, equilibrium, and kinetics of the various industry effluents and their adsorption capacity.

Keywords: Shrimp Shell, Adsorption, Biosorbent, Heavy Metals (Hm), Effluents.

INTRODUCTION

The most precious natural resource for sustaining human, animal, and plant life is water. However, the water on the earth now is hazardously polluted because of the rapid growth of industry, increase in the human population, and domestic and agricultural activities coloring and non-coloring dyes have been widely used in industries not limited to leather, paper, printing, textiles and cosmetics, leading to serious environmental issues (Tsiourtis, 2001). The wastewater effluents from almost all printing and dyeing industries are found to contain toxic, and carcinogenic compounds like aromatic, polycyclic aromatic or heterocyclic dye, that are difficult to be discarded and decomposed (Maximous *et al*., 2010).

Heavy metals are non-biodegradable, carcinogenic, and chemically stable, allowing them to integrate into biological systems and disrupt entire food chains (Rajput et al., 2016). Dyes, on the other hand, play a critical role in the production of paints, coatings, pigments, and textiles. Currently, over 105 types of dyes are commercially available. To meet industrial demands, an estimated 1,600 kilotons of dye components are produced annually, with 10%–15% of this amount discharged into local water systems. The primary chemical technologies for processing heavy metals include methods such as chemical deposition, ion exchange, reverse osmosis, photocatalytic reduction, electro-desorption, membrane filtration, evaporation, solvent extraction, and adsorption (Sarı and Tuzen, 2008b; Huang and Chen, 2009; Ahmad et al., 2012b). Among these, adsorption is recognized as the most cost-effective and efficient approach for recovering metals from aqueous solutions. This is due to its reversible nature, desorption capabilities, straightforward implementation, flexibility, and resistance to toxic contaminants (Abas et al., 2013; Halnor, 2015).

Adsorbents used in water treatment processes are typically non-toxic and environmentally friendly, minimizing their negative impact on ecosystems (Tamjidi et al., 2019). One promising adsorbent is waste shrimp shell (WSS), a by-product of the shrimp processing industry. WSS is a valuable biomass resource due to its composition, which includes chitin (15–30%), protein (41%), and minerals (34%) (Kannan et al., 2018). Shrimp waste (SW), comprising cephalothorax and exoskeleton, accounts for approximately 40% of a shrimp's total weight and serves as a widely used source of chitin. Globally, the production of SW is significant, with China alone generating around 500 kilotons annually. The recovery of chitin from SW could be further optimized with the development of effective technologies, unlocking its full potential as a resource. Mine-impacted water (MIW), a critical environmental issue, poses severe threats to water resources and aquatic ecosystems. Shrimp shell has been identified as a viable treatment agent for MIW due to its affordability, high chitin content, and the presence of calcium carbonate, which acts as an acid-neutralizing agent. In regions near mining operations, surface and groundwater pollution represent significant environmental challenges that demand sustainable and effective remediation solutions.

The oxidation of pyrite (FeS_2), the primary cause of acidic water formation or "acid mine drainage" (AMD), has been extensively studied (Nordstrom and Alpers, 1999). AMD is characterized by a low pH (ranging between 2 and 4), high concentrations of suspended solids, and elevated levels of sulfates and dissolved metals, including iron (Fe), aluminum (Al), manganese (Mn), zinc (Zn), copper (Cu), and lead (Pb) (Morin and Hutt, 2001). Streams impacted by mining waste acids are complex environmental systems requiring chemical treatments for both physical and biological remediation (Daubert and Brennan, 2007). In Brazil's Southern region, hydric resources are severely affected by coal mine-impacted water (MIW). Rivers within the coal basin of Santa Catarina State are particularly contaminated by AMD, rendering the water unsuitable for any practical use. A research initiative aims to transform coal MIW into water suitable for non-potable purposes, thereby preserving limited high-quality water resources for potable needs.

Shrimp shell, an abundantly available by-product in Santa Catarina State, has been selected as a cost-effective treatment agent for MIW. Its high chitin and calcium carbonate content make it an excellent acid-neutralizing material. Preliminary studies have demonstrated that raw shrimp shell performs better as an MIW treatment agent compared to processed chitin, yielding significant metal recovery rates and effectively increasing river water pH under various experimentalconditions, such as varying shrimp shell content-to-water volume ratios, contact time, and mixing rates. However, raw chitin presents some limitations, including low surface area, pore volume, and average pore size. To enhance its performance, structural modifications are necessary. One promising approach involves the production of chitin nanowhiskers, a class of nanomaterials with a high surface-to-volume ratio and functionalized groups. These nanowhiskers can be synthesized via acid hydrolysis of native chitin (Gopi et al., 2016).

ANALYZING THE PROPERTIES OF SHRIMP SHELL

To thoroughly evaluate the physical and chemical properties of shrimp-based adsorbents, a range of advanced analytical techniques is employed. These include Brunauer–Emmett–Teller (BET) analysis, dynamic light scattering (DLS), Fourier transform infrared spectroscopy (FTIR), zeta potential analysis, energy-dispersive X-ray spectroscopy (EDX) with mapping, scanning electron microscopy (SEM), field emission scanning electron microscopy (FESEM), transmission electron microscopy (TEM), scanning tunneling microscopy (STM), atomic force microscopy (AFM), nuclear magnetic resonance (NMR), and X-ray diffraction (XRD). FTIR analysis is utilized to identify the functional groups present in the adsorbent material, providing insights into its chemical composition. BET analysis measures the surface area and pore volume of the adsorbent, critical factors influencing adsorption capacity. DLS analysis is used to examine the bio-adsorbent's nanostructure and to determine the particle size distribution. Collectively, these techniques provide a comprehensive understanding of the adsorbent's structural, chemical, and physical characteristics, supporting its optimization for effective pollutant removal.

The phase and crystalline structure of materials can be effectively analyzed using X-ray diffraction (XRD), while nuclear magnetic resonance (NMR) provides insights into the purity and concentration of the material. Techniques such as transmission electron microscopy (TEM), scanning tunneling microscopy (STM), and atomic force microscopy (AFM) are employed to investigate aggregation, structure, particle size, surface heterogeneity, and particle dispersion. Scanning electron microscopy (SEM), field emission scanning electron microscopy (FESEM), and energy-dispersive X-ray spectroscopy (EDX) are essential for studying surface morphology, structure, and particle properties. These analyses also enable the visualization of elemental distributions through color mapping and the quantification of constituent elements. Zeta potential analysis, on the other hand, is used to measure the surface charge of particles, which is crucial for understanding colloidal stability and adsorption behavior (Tamjidi et al., 2019; Foroutan et al., 2020). Table 1 summarizes the findings from detailed studies on shrimp shells. The properties of shrimp shells as adsorbents are significantly influenced by surface treatment methods and pyrolysis temperatures. Process temperature plays a critical role in determining the physical and chemical properties of the adsorbent. Modification techniques focus on altering hydrophobicity, polarity, surface area, and other structural and morphological features of the biosorbents. These changes directly impact the adsorption efficiency.

Table -1: Various Studies of shrimp waste for biosorption

Bio-adsorbent	SEM/FESEM	FTIR	BET	Ref.
Raw shrimp shell particle (RSSP)	Surface roughness	O-H	Surface area (3.618 m^2 g^{-1})	(Youzhou *et al.*, 2018)
Treated shrimp shell particle (TSSP).	Surface roughness	N-H	Surface area (66.35 m^2 g^{-1})	(Youzhou *et al.*, 2018)
Chitosan NP (extracted from shrimp shell)	Porous layer is composed of network	C-N C-O $C\text{-}NH_2$	-	(Ali *et al.*, 2018)
shrimp peel species of Vanami	Cavities with different size	Fe-O CO_2 CO^{2-}	-	(Foroutan *et al.*, 2015)
Magnetic chitosan particle	Irregular shape with aggregation	N-H O-H C-O-C C-O	-	(Thuan *et al.*, 2018)

Factors influencing the biosorption of pollutants by Shrimp shells as an effective adsorbent:

Adsorption of heavy metal is dependent on the following: Temperature of reaction, Contact time, Adsorbent dosage, Metal ion initial concentration, pH.

The temperature of reaction and contact time:

The most significant step in Biosorption is to analyze the effect of contact time on the sorption process. The reaction rate and the equilibrium of the process are highly dependent on the temperature. In general, adsorption is always carried out with the heat evolution. By Le-Chatelier's principle, the adsorption efficiency decreases with the temperature increase (Tamjidi and Ameri 2020). The effect of temperature on the adsorption capacity is well explained with an initial and final temperature of 310 and 320 K respectively. There was a decrease in the adsorption capacity of methyl orange with the temperature increase (Chao et al., 2020). Coal acid mine drainage (AMD) is a condition where low pH and high concentrations of sulfate and different metallic ions are present. Most of the studies on kinetic parameters have revealed that a 200 min contact time transforms AMD into water for non-potable reuse.

Adsorbent dosage:

The adsorption capacity of an adsorbent is directly influenced by the concentration of the material being adsorbed. Generally, adsorption efficiency improves with an increase in the adsorbent dose due to the higher availability of active sites for binding. Núñez-Gómez et al. (2019) explored an innovative method for synthesizing cross-linked magnetic chitosan particles (MCPs) using steel slag and shrimp shells, with green tea extract serving as a cross-linking reagent. The study demonstrated that these MCPs exhibit enhanced adsorption capacities for metal ions, achieving 66.23 mg/g for Ni(II) and 126.58 mg/g for Cu(II). Moreover, the MCPs showed remarkable regeneration stability, maintaining an efficiency exceeding 83% even after five cycles of the adsorption–regeneration process.

Metal ion initial concentration:

The initial concentration of a contaminant is a critical parameter influencing the entire adsorption process. Since the concentration of heavy metals in aqueous solutions can vary, the recovery rate also differs accordingly (Fabbricino et al., 2016). The effect of the initial Congo red (CR) dye concentration on adsorption capacity and removal efficiency was studied under equilibrium conditions using 0.1 g of adsorbent and 30 ml of dye solution. The equilibrium adsorption capacity for treated shrimp shell particles (TSSP) was 225.9 mg/g, significantly higher than that of raw shrimp shell particles (RSSP). When the initial pollutant concentration is low, the removal efficiency reaches its maximum. However, as the initial concentration increases, the efficiency decreases. This phenomenon occurs because, at low concentrations, the ratio of active sites on the adsorbent surface to the number of contaminant molecules is high, leading to maximum adsorption. At higher concentrations, this ratio diminishes, reducing the availability of active sites and thereby lowering the adsorption efficiency (Kandra et al., 2012).

Effect of pH:

The adsorption process is generally more effective at higher pH levels, as an increase in pH enhances the adsorption rate. This occurs because the competition between protons and heavy metal ions for active sites diminishes, along with a reduction in repulsion forces (Rodrigues et al., 2020). However, beyond a certain pH threshold, adsorption efficiency declines due to the formation of metal ion complexes with hydroxide ions (Tamjidi and Esmaeili, 2019). Rostamian et al. investigated a continuous descending flow system for metal ion removal. Their findings revealed a removal efficiency of up to 90% for Fe and 88% for Mn, with a corresponding increase in pH from 3.5 to 6.8. This study highlights the suitability of the proposed treatment method as a low-cost solution for addressing acid mine drainage (AMD).

Table – 2: List of Pollutants removed and other associated parameters

Bio-adsorbent	pollutant	pH	Contact time (minutes)	Temperature (°C)	Pollutant concentration (mg/L)	Adsorbent dose (g/L)	Ref.
Chitosan NP (extracted from shrimp shell)	Mn(II)	5	100	25	40	10	(Ali et al., 2018)
Chitosan NP (extracted from shrimp shell)	Fe(II)	4	100	25	40	10	(Ali et al., 2018)
shrimp peel species of Vanami	Pb(II)	6	130	30	60	5	(Foroutan et al., 2015)
Chitin (extracted from shrimp shell)	Zn(II)	7	180	–	500	0.5	(Jaafarzadeh et al., 2013)
Chitosan nanoparticles	Fe(II)	4	120	30	120	0.5	(Ali et al., 2018)
	Mn(II)	6	120	30	120	0.5	(Ali et al., 2018)
chitosan nanofibers	U	6	120	30	170	5	(Rostamian et al., 2019)

Various pollutant removal and recent improvements using Shrimp waste:

Druzian *et al*. analyzed the preparation and the characterization of chitin nanowhiskers to remove crystal violet (CV) dye from an aqueous solution**.** The rod-shaped ChNW and the increased pore size are reported to be the main characteristics that contributed to the increase in adsorption potential when compared with raw Chitin. Also reported were the adequate adsorbent dosage of 5 $g.L^{-1}$ and an optimum pH was 8. Nirmal *et al*. studied the direct dye adsorption on the chitin-containing residues. As a representative waste, the shells of two shrimp species are tested. Removal percentage up to 90% Dye removal rate is obtained in about 2 h, using 2.1 $mg.L^{-1}$ of shells, simply dried and grinded. Comparative analyses are also conducted on commercial chitin and chitosan flakes. Coal acid mine drainage (AMD) is usually characterized with low pH and has been reported to contain significant concentrations of sulfate and metallic ions (Nunez-Gomez *et al*., 2017).

Further research has been conducted to evaluate the sorption equilibrium for the removal of metal ions and acids in coal acid mine drainage (AMD) using shrimp shell (SS), in both batch and continuous-flow removal processes. Five different adsorption isotherms were examined, with the results indicating that the Freundlich isotherm model provided the best fit. Two adsorbents synthesized from shrimp shells were characterized, and their adsorption performances for Congo red dye were investigated (Youzhou et al., 2018). The findings showed that the treated shrimp shell adsorbent exhibited a higher removal capacity than raw shrimp shell powder. The maximum adsorption capacity for the treated shrimp adsorbent was 288.2 mg/g, as predicted by the Langmuir isotherm model. Notably, this capacity is significantly higher than that of chitin, highlighting the effectiveness of the treated shrimp shell adsorbent. The adsorption process was well-described by the pseudo-second-order kinetic model, further confirming the potential of shrimp shell biosorbents for applications in dye wastewater treatment. Additionally, the Langmuir adsorption isotherm was found to accurately model the adsorption behavior on both treated (TSSP) and raw shrimp shell powder (RSSP).

Andreazza et al. studied the removal potential of copper ions using various chitin extracts derived from shrimp culture residues. Four different extracts were prepared using processes such as milling and simple boiling, demineralization, deproteinization, and deacetylation. After the adsorption tests, samples were collected to measure the copper concentration remaining in the solution, allowing for the evaluation of each extract's effectiveness in removing copper ions. The possible extraction was investigated by thermal degradation analysis, X-ray diffraction, infrared spectroscopy, scanning electron microscopy and dispersive energy X-ray spectroscopy and was found to show removal of 52.66 $mg.g^{-1}$, 23.05 $mg.g^{-1}$, 19.31 $mg.g^{-1}$ and 67.81 $mg.g^{-1}$, respectively. Rodrigues et al. explored the use of sulfate-reducing bacteria (SRB) and the role of chitin in the development of a comprehensive biological system for treating mine-impacted water (MIW), specifically river water contaminated by coal acid mine drainage (AMD), an extremely acidic effluent. Their findings indicated highly effective removal rates, with sulfate removed at a fraction of 0.998, iron at 0.99, aluminum at 0.98, and all manganese ions successfully eliminated. Additionally, the sorption kinetics of Fe ions by the sediments were investigated, and the results revealed pseudo-second-order kinetics.

The acidic level of wastewater from coal mining can be subsequently neutralized by Chitosan microspheres. 0.001 kg of chitosan in a spherical shape was capable of increasing wastewater pH from 2.5 to 4.0 and removing approximately all of the Fe (III) contents. Vishali *et al*. showed the usage of *Fenneropenaeus indicus* (Shrimp) shell, as a coagulant. This is used in the treatment of simulated paint factory effluent (SPFE) for color and turbidity removal. The color and turbidity removal was found to be 93.67% and 81.77% respectively. The results were compared with those of chemical coagulant chitosan, concluding that, due to its biodegradability and widespread availability, shrimp shell powder (SSP) has the potential to serve as a sustainable, green alternative to chemical coagulants. Chao et al. synthesized a hydrochar adsorbent from waste shrimp shells (WSS) by undergoing deproteinization and deacetylation, followed by hydrothermal carbonization (HTC). A similar hydrochar adsorbent was prepared by HTC of commercial chitosan under the same conditions. Further analysis revealed that the waste shrimp shell hydrochar (WSH) contained nitrogen-rich functional groups and exhibited a long aliphatic chain structure, enhancing its adsorption capabilities.

Calcium carbonate-acid etching in WSS led to a higher specific surface area of WSH (12.65 m^2/g) which was nearly 6 times higher than that of CCH (2.13 m^2/g). With an initial solution of pH 4.0, WSH could rapidly achieve a relatively high adsorption capacity of 755 mg/g for methyl orange molecules. Excellent adsorption potential and regeneration performance suggest WSH to be a promising and affordable adsorbent candidate for anionic dye removal. Thuan et al. (2018) investigated the preparation of cross-linked magnetic chitosan particles (MCPs) from steel slag and shrimp shells, using green tea extract as a crosslinking agent. These particles were employed to study the adsorption properties of Cu(II) and Ni(II) ions in aqueous solutions. The adsorption isotherms, kinetics, and thermodynamics for Cu(II) and Ni(II) were measured and analyzed. The results indicated that the synthesized MCPs exhibited high adsorption capacities for both metal ions, with 126.58 mg/g for Cu(II) and 66.23 mg/g for Ni(II). Rostamian et al. (2019) examined the production of chitosan nanofibers (CS-NFs) using the force spinning method, where centrifugal force replaces electric force, significantly improving the efficiency and scalability of nanofiber production. The study explored uranium (U) removal from aqueous solutions by adsorption onto the CS-NFs surface. Furthermore, equilibrium models were applied to fit the experimental data. Kousha et al. optimized conditions for the removal of Acid Blue 25 (AB25) dye using Penaeus indicus shell biomass. The study analyzed key variables, including dye concentration (90–130 ppm), salinity (10–30%), biomass amount (0.2–0.4 g/L), and reaction time (30 min), using central composite design (CCD) at an optimal pH of 2. The results showed a maximum dye removal of 95.64% at the highest salinity concentration, which aligned with the calculated values.

Núñez-Gómez et al. examined the effectiveness of shrimp shell (SS) in treating coal acid mine drainage (AMD), characterized by low pH and high concentrations of sulfate and various metallic ions. A central composite rotatable design (CCRD) was employed to optimize parameters for AMD remediation using a combination of shrimp shell (SS) and mussel byssus (MB). SS was chosen due to its high chitin and calcium carbonate content, while MB was selected for its potential synergistic effects in the treatment process. In this study, five kinetic models were tested to explore the adsorption mechanisms. The kinetic analysis revealed that a 200-minute contact time was sufficient to transform AMD into water suitable for non-potable reuse. To determine the optimal conditions for metal removal, a statistical study was conducted. The removal of elements such as Fe, Al, Mn, Co, and Ni from mine-impacted water (MIW) was investigated. The CCRD approach, with a quadruplicate at the midpoint, was used in the experiment. Adjustments to the model were made for a 90% and 75% confidence interval for Fe and Mn removal, respectively. The optimal conditions for pollutant removal were found to be 188 rpm and 9.36 g/L of shrimp shells.

CONCLUSION

This review examines the potential of shrimp waste as an environmentally friendly, efficient, and low-cost alternative to conventional adsorbents. In recent years, it has garnered increasing attention due to its effectiveness. A wide range of shrimp shells has been analyzed, and various experiments have been conducted to assess their adsorption capacity. The structure of shrimp shells is highly similar to that of the cost-effective biopolymers chitosan and chitin, making them promising bio-adsorbents. Various modification processes have been explored to enhance the properties of these biosorbents. Notably, adjusting the pyrolysis temperature has been found to significantly improve the adsorption process by increasing the energy of active binding sites and creating additional functional groups. Additionally, the influence of various factors such as pH, contact time, temperature, initial metal ion concentration, and adsorbent dose on the adsorption process has been thoroughly studied.

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