A review of utilization of Mollusca shell for the removal of contaminants in industrial waste water

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Abstract- Bio-sorption is a promising technology for the removal of contaminants like nutrients, textile dyes, organic matter and heavy metals which have hazardous effects on the aquatic ecosystems. In recent years many low cost sorbents such as algae, fungi, bacteria and agricultural byproducts have been investigated for their bio-sorption capacity towards water contaminants. But less attention has obtained my Mollusca shell as a waste material for the utilization of bio- sorbent due to its composition and availability. The studies conducted with the focus of utilizing Mollusca shell has confirmed that the propensity of shells to remove contaminants in waste water as a single component and combinations with other materials.

Index Terms- Bio-sorption, Mollusca shell, Contaminants, Removal

I. Introduction

The environment is the place in where all the living and non-living beings had been interconnected and interrelated with each other with having little interference to others lives. However, with the emergence of the industrial revolution, urbanization and with the increase in technological development, anthropic activities have resulted in substantial and notable increase of toxic pollutants in the environment. Worldwide consciousness of crisis over environmental pollution problems is spreading and especially countermeasures to solve the environmental pollution come into the limelight as concerns of the entire globe beyond any regions and nations.

Water resources are of paramount and exclusive importance to all ecological communities as well as for the human growth and development. According to a report compiled by the World Economic Forum in collaboration with, Oxford University, University of Pennsylvania, and National University of Singapore, water crisis ranks 3rd in the Global Risks in the year of 2014 (Naik *etal*,2016). With the view point of 'dilution is the solution for the pollution' water bodies became as one of the prime sources, when contaminated with noxious pollutants like heavy metals, organic toxicants, reactive textile dyes, and fluorides that disrupt and destroy the fragile ecology in where all the living beings are inter-related. Promiscuous discharge of contaminants have resulted in its accumulation in the aquatic biota leading to its bio-magnification which generates and induces health hazards in humans and other living organisms substantially.

Heavy metals are among the conservative pollutants that are unable to undergo a bacterial attract or bacterial digestion or degradation process and they are considered as permanent additions to the marine environment (El-Nady and Atta, 1996). As a result of this, their concentrations often exceed the permissible levels normally found in soil, water ways and sediments. Hence, they find their way up the food pyramid (Igwe and Abia, 2006). When they accumulate in the environment and in food chains, they can profoundly disrupt biological processes. Therefore, a complete understanding about noxious effects caused by the release of toxic metals into the environment and the emergence of more severe environmental protection laws, have encouraged studies about removal/recovery of heavy metals from aqueous solutions using various techniques and technologies.

A variety of technologies such as chemical oxidation, precipitation, ion exchange, reverse osmosis and membrane separation has been adopted for the removal of contaminants in water such as heavy metals, dyes and organic and inorganic substances (Wang and Chen, 2006). However, chemical precipitation and electrochemistry, ion exchange, membrane technologies and activated carbon adsorption are extremely expensive and pave the way to generate secondary pollution and/or produce a large amount of sludge that is difficult to treat. For instance Activated carbon is only able to remove around 30-40 mg/g of Cd, Zn, and Cr in water and is non-regenerable, which is quite costly to wastewater treatment (Babel and Kurniawam, 2003) but also chemical precipitation and electrochemical treatments are ineffective, especially when the metal ion concentration in the aqueous solution is between 1mg/L and 100mg/L (Wang and Chen, 2006).

Adsorption is the ability of the adsorbate to adhere or attach to the adsorbent. It is a well-established separation technique to remove dilute pollutants as well as to recover valuable products from aqueous solutions. In the conventional adsorption process, the particle size of the adsorbent is restricted because of hydrodynamic phenomena such as pressure drop (Chia-Chang and Hwai-Shen, 2000). Adsorption is divided in two categories according to the forces that are made in between adsorbate and adsorbent. First one is generated due to forces of physical nature called van der Waals force which is relatively weak, since they are not sufficiently strong to influence appreciably the reactivity of the molecule adsorbed. The second type is considerably stronger. In there the adsorbed molecules are held to the surface by valence force of the same type as those occurring between bound atoms in molecules. This is known as chemisorption and the heat evolved is of the order 10 to 100 kcal/mole, compared to physisorption which has less than 5 kcal/mole (Motoyuki, 1990).

Adsorptive removal of heavy metals from aqueous effluents which have received much attention in recent years is usually achieved by using activated carbon or activated alumina (Shim et al., 2001; Ouki et al., 1997; Hsisheng and Chien-To, 1998; Monser and Adhoun, 2002). Activated carbon is a porous material with an extremely large surface area which is capable of generating intrinsic adsorption potential with many chemicals. Polymer resins that can form complexes with the heavy metal ions are the best adsorbents (Lu et al., 1994). These are called conventional adsorbents and many others have been reported such as silica gel, active alumina, zeolite and metal oxides (Motoyuki 1990).

Biosorption or bioremediations consists with wide range of applications which involve the detoxification of hazardous substances instead of transferring them from one medium to another by methods of microbes and plants. This process is considered as less disruptive and can be often carried out on site, eliminating the need to transport the toxic, materials to treatment sites (Gavrilescu, 2004). Bio sorbents are prepared from naturally abundant waste biomass. Due to the high uptake capacity and very cost-effective source of the raw material, biosorption is a progression towards a perspective method not only utilization of naturally available materials as emergent need of present and future to reduce the waste generation not only in industrial processes but also from pollution remediation. The use of naturally available materials as biosorbents has many advantages over the aforementioned methods (Wang and chen, 2006) as they are considered as an effective remediation strategy due to its low cost and relatively simple design. To date, many such absorbents have been studied, including chitosan, zeolites, microorganisms, clay, and waste products from industrial operations such as fly ash, coal and oxides (Babel and Kurniawam, 2003).

Bivalve mollusk shells are available and very common by-products during sea food processing, and are very common around the coasts of many coastal countries. Therefore they can be brought free from the local markets and industries. The research application of mollusca shells as a biosorbant for removing contaminants from the environment is judged to be very helpful in preserving eco-systems, preventing the damage of natural landscape and solving health problem. Therefore, new applications for utilizing these wasted Mollusca shells are expected to contribute towards recycling consciousness within the society [Asaoka et al., 2009].

II. Importance of Utilizing Mollusca Shells for Waste Water Management

Pertinence of mollusk in both live form and waste shell is very crucial for the waste water management practices due to many reasons. First one is their propensity to remove contaminants through bio- filtration process by live specimens and bio-sorption processes by the shells of mollusks. In here we only consider about the ability of Mollusca shell as a waste or byproduct to remove wastes by bio-sorption process that are generated in industrial processes.

Mollusks include many of the most important seafood such as abalone, clams, mussels, oysters, octopus, squids, scallops and terrestrial snails. From many of these animals only mussel part is consumed as a food. Therefore the shell or skeletal part is removed as wastes or byproduct which is subjected to generate ecological and social problems. For instance, a large amount of oyster shells is a general waste that fishermen should take care of but it seems difficult to handle it effectively due to the problems of securing of landfill sites and collection/transportation of oyster shells [Jung, 2005, 2007]. This waste bulks at coastal areas generate many environmental problems including pollution of coastal fisheries, damage to natural landscape, management of public water surface and health hazards. According to Kim, 2007 generation of oyster shell is estimated on the average at 270,000tons/year and more than 50-70% of them are dumped into waters bodies and reclaimed lands. This pave the way to cause a disagreeable fishy odour by virtue of decomposition of fresh remnant attached to oysters (Kim, 2007; Yoon et al., 2003).

Generally shellfish wastes is classified into category-3 animal byproducts and must be handled and treated to the same standards as other animal byproducts. This means that it should only be treated or disposed of through facilities that are licensed specially to take that category product according to the requirements of Animal Byproduct Regulations (ABPR). Thereupon reusing and recycling has emerge as forthcoming and inevitable issue in mariculture industry. Recycling and reusing of Mollusca shells has various applications including as a construction material (Yoon et al., 2003), as a fertilizer, larval farming (Nippon, 1993), eutrophication control (Kwon et al., 2004), soil conditioner (Lee et al., 2008), sludge conditioner (Lee et al., 2001), catalysts (Nakatani et al., 2009), and desulphurization sorbents (Jung et al., 2000). Apart from these applications, the utilization of Mollusca shell for waste treatment is an upcoming expectation for future because not only it's availability but also its composition.

The Mollusca shell is an organo-mineral composite, where the dominant mineral is aragonite, or calcite, or in particular cases, vaterite. It is internally associated to an organic matrix, which accounts only for 0.1–5% of the shell weight. This matrix represents amalgamate of proteins, glycoproteins, chitin and acidic polysaccharides, secreted by the calcifying tissues during skeletogenesis. This organo- mineral mixture is consequently sealed within the skeleton during their growth. At macroscopic level, the adjunction of organic components to a mineralized structure enhances the mechanical properties to the whole organo-mineral assembly. At molecular level, the matrix plays a key role in the mineralization process (Marin etal, 2008). Metal absorbance is related to elimination of Ca^{2+}on shell surface and exposure of more functional groups that can bind contaminants specially metal ions to the shell surface.

E. Sorption potential estimation

A sorption experiment was conducted using 100ml sorption systems that contained 1g of bivalve shell powder and different initial concentrations of metal solutions (Ex; ranging from 100mg/L to 1400 mg/L). The resulting supernatant was then analyzed for the presence of residual copper by atomic absorption.

III GENERAL METHODOLOGY

A. Pretreatment of the Mollusca shells

Collected shells were washed using water and dried in sun for about 8-10 hours and crushed them using mortar and pestle to form small pieces and these pieces were powdered using mixer. Prior to use as an absorbent, the powered Mollusca shell were washed for 10 minutes at 100rpm with distilled deionized water and after that they were collected by centrifugation at 2000rpm for 5 min. Then the precipitate of Mollusca shell was dried at room temperature.

B. Acid treatment

An aliquot of the dried Mollusca shell precipitate was acid-treated for 60 minutes at 100rpm by soaking the solution in 1M H_2SO_4, after which the resulting shell powder was collected by centrifugation. Another aliquot of the dried shell powder precipitate, as a control, was parallel-treated with distilled deionized water

C. Base treatment

An aliquot of the dried Mollusca shell precipitate was base-treated for 60 minutes at 100rpm by soaking the solution in 10% of 250mL NaOH solution, then remaining shell precipitate was collected by centrifugation. Same as the acid treatment another aliquot of dried shell powder precipitate was parallel treated with distilled deionized water as a control.

D. Sorption experimental procedure for heavy metals

Sorption experiments were conducted in batches in Erlenmeyer flasks (generally 250mL is used) that contained 100mL of solution containing the metal salt(s) or 100mL of the actual wastewater. The required amount of powdered Mollusca shell was added to each flask and sealed with a cap and subsequently shaken for the required time at 200rpm at the indicated temperatures. Then immediately flasks were centrifuged at 4800rpm for 5 minutes in which supernatant was collected which was directed to atomic adsorption analysis. The removal efficiency of specific heavy metal was calculated using following formula,

$Q = C_0 - C_1/C_0$

In where,

Q= The removal efficiency of specific metal (%)

C_0= Initial concentration of specific metal (mg/L)

C_1=Residual concentration of specific metal after sorption (mg/L)

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IV RESEARCH FINDINGS AND FUTURE PROSPECTS

Jatto etal, 2010 conducted a research to investigate the suitability of snail shell (Achatina achatina) as an adsorbent or coagulant in waste water treatment collected water from food industry located Edo state which is specialized in the production of corn flakes

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methods for waste water and effluent analysis (Ademoroti, 1996).

Parameter	Results before treatment	Results after treatment
pH	5.42	6.42
Colour	Dark brown	Light brown
Turbidity	332 NTU	133 NTU
Temperature	20 ^oC	20 ^oC
Conductivity	0.294 $Mscm^{-1}$	0.164 $Mscm^{-1}$
Total Suspended Solids	0.814 mg/L	0.184 mg/L
Total Dissolved Solids	15600 mg/L	15200 mg/L
Total Solids	15600.8 mg/L	15200.2 mg/L
Ammonium Nitrogen	0.085 mg/L	0.015 mg/L
Nitrate	41.08 mg/L	13.32 mg/L
Sulphate	58.11 mg/L	15.46 mg/L
Phosphate	0.173 mg/L	0.00 mg/L
DO	1.14 mg/L	2.16 mg/L
BOD	29.27 mg/L	19.77 mg/L
COD	872 mg/L	215 mg/L

The increment of pH value after treatment indicates slightly alkaline condition due to presence of Calcium Carbonate in the shell (Aboua, 1995). According to the stability studies conducted shows that snail shell powder was stable for wide range of pH values and according to optimum dosage measurements done by using turbidity and COD as an index shows that 3.3g-3.4g per 100mL of shell had ability to coagulate or treat waste water effectively (Jatto etal, 2010).

Phosphate was completely removed from the treated water sample. This is a result of the presence of calcium ion in the shell. Calcium has the ability to react with phosphate ion resulting in the formation of calcium phosphate and calcium hydrogen phosphate which can be removed by filtration (Jatto etal, 2010).

Many researches were conducted to investigate the efficiency of removing heavy metals and contaminants by using Mollusca shell powder as a bio sorbent. Removing of heavy metals like mercury, Cadmium, Arsenic, Lead, Nickel by using raw materials available in nature like crab shell powder, oyster shell powder and bivalve shell powder as redundant and bio- sorbent was contacted by Naik, 2016. Bivalve shell powder and Oyster shell powder showed 98% to 99% removal for all metals except arsenic for which Bivalve powder showed 91% removal and Oyster powder showed 85% removal. In life cycle assessment

Bivalve powder and Oyster powder showed 100% removal for Cadmium and Lead for all 10 cycles that they conduct the experiment. The adsorption of metals followed the trend: Mercury> Lead> Nickel> Fluoride> Cadmium> Arsenic. Mercury and Lead removal was found to be 100% followed by Lead which showed 99% removal. Nickel and Cadmium showed almost 98%-99% removal for all materials tested (Naik, 2016). Adsorption capacity of metals by adsorbent on acid digestion has dramatically reduced in both Bivalve shell powder and Oyster shell powder for Nickel and Mercury and overall capacity for all metals has declined after acid digestion. Adsorption Capacity of Metals by adsorbents on base digestion has increased for all metals (Naik, 2016).

It has been reported that pretreatments that were conducted for absorbants by physic-chemical processes have ability to ameliorate the propensity of heavy metal removal (Wang and Chen,2006).for instance acid pretreatment with Sulfuric acid can improve the copper sorption capacity of bivalve molluscan shell powder (Yang etal, 2009).

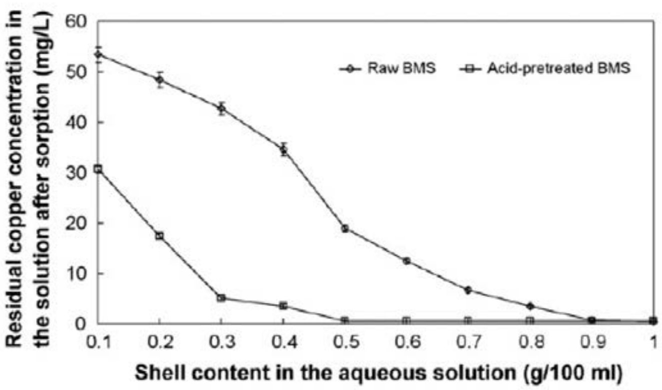

Figure 4.1: Variation in copper removal with bivalve mollusca shell powder dosage in the sorption system

Regardless the nature of the material (raw or acid treated) the copper removal efficiency has increased with the amount of bivalve shell powder increased. Especially copper removal efficiency has increased in expeditious way until a dose of 0.5g/100mL and when dose reach more than 0.5g/100mL the removal efficiency performed by raw bivalve shell powder continued to increased rapidly while acid treated bivalve shell powder become low in its efficiency (Yang etal, 2009). Depending on the absorbent and process employed in most cases alkali and acid treatment of absorbent enhance the metal sorption capacity, whereas acid treatment has almost has no influence on metal sorption (Vianna etal, 2000). For the raw bivalve shell powder, good copper removal efficiencies were observed when the $CuSO_4 \cdot 5H_2O$ concentrations were less than 400mg/L; however, the removal efficiencies declined sharply at higher concentrations (Yang etal, 2009).

It is crucial to optimize the contact time for heavy metal removal in waste water management practices and industry. Both raw and acid treated bivalve shell powder demonstrated high copper removal efficiencies when the contact time is less than 90 minutes and their efficiencies after 90 minutes contact were 99.33% and 99.94%, for raw and acid-pretreated bivalve shell respectively Yang etal, 2009), indicating a rapid copper sorption similar to the biosorption of fungal biomass (Wang and Chen,2006)

Physico- chemical parameters like pH and temperature are very important factors of sorption ability of the sorbent.

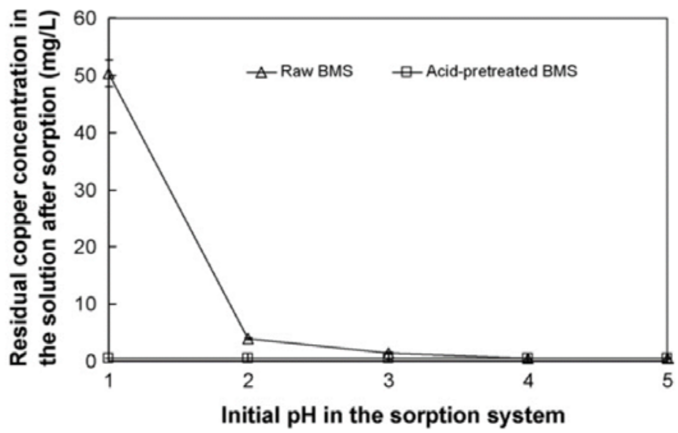

Figure 4.2: Effect of initial pH of the solution on copper removal.

The effect of pH on the removal of heavy metals can usually be explained by strong influences on site dissociation at the surface of the absorbents as well as by the solution chemistry of the heavy metals (Wang and Chen, 2006) and the pH is one of the most important environmental factors. The optimal pH for the sorption of metal ions varies among systems. For example, the optimal pH value for the removal of copper by *Sacchromyces cerevisiae* is 5–9(Volesky. B, 1990)], whereas it is 1.5–9.0 for removal by industrial by-products such as iron/steel slag (Kuniawan etal,2006) Copper removal efficiency of raw bivalve shell powder was lowest when the pH value was 1 and while increasing the pH value removal efficiency has increased upto 99.5%. Throughout the entire pH range acid treated bivalve shell powder showed high removal efficiencies (Figure 4.2) involved in the sorption of heavy metal ions (Yang etal, 2009).

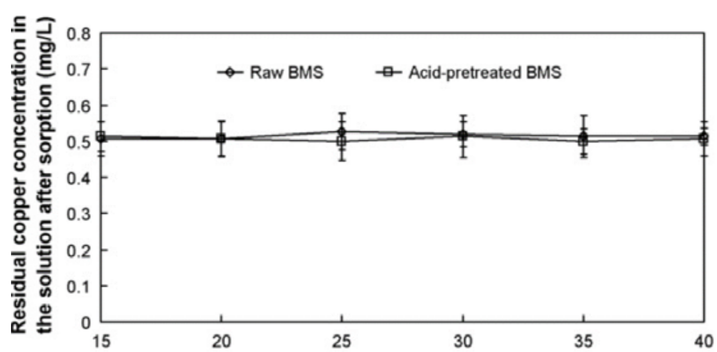

Figure 4.3: Effect of temperature on copper removal

It is believes that sorption reactions are generally exothermic in where bio-sorption capacity increases as temperature decreases (Kapoor and Viraragharan, 1997). At the same time this decrease in bio-sorption capacity at higher temperatures likely occurs due to damage to the active binding sites in the biomass (Ozar and Ozar, 2003). This differences are mainly depend on the nature of the adsorbent and /or species of metal. According to Yang etal,2009 both raw and acid treated bivalve shell powder demonstrated higher and stable copper removal efficiencies in the range of 15^{0}C- 40^{0}C reaching the 99.5% efficiency approximately (Figure 4.3).

Not only as a single bio-sorbent but also Mollusca shell was utilized in combinations with other naturally available materials to obtain higher efficiency. The study was conducted to examine the effectiveness of 4.0–4.75mm crushed shells and *Sphagnum* peat moss as low-cost natural adsorbent filter materials for the removal of cadmium and nickel ions from binary aqueous solutions (Li and Champagne, 2009). Cadmium and nickel were selected as representative heavy metals commonly found in metal rich wastewater such as landfill leachate (El-Gendy etal, 2006). Crushed mollusk shells were found to be a better filter medium for cadmium removal, while the *Sphagnum* peat moss was noted to be the better filter medium for nickel removal. The highest overall cadmium and nickel removal efficiencies were noted to be 47.9% and 42.7%, respectively, in the 10cm diameter fixed-bed column packed with a 15cm depth of mollusk shells, hydraulically loaded with a nickel and cadmium binary solution at a rate of 1.5 mL/min (Li and Champagne, 2009).

As a waste material which is generated in food industry mainly Mollusca shell has high propensity to use for the applications of bioremediation. Though many researches were conducted by using Mollusca shell (mainly bivalve shells) as a bio-sorbent for removing contaminants in water very few number of species were selected for the experiments. Owing to the composition and abundance of shells, Mollusca shell can be tested for various contaminants not only for heavy metals but also for fluorides, dyes and eutrophication responsible nutrients like nitrate and phosphates. By utilizing the combinations of naturally available materials, the absorbance efficiency of contaminants can be increased while considering the specific characteristics of the materials. Limited scope and researches were conducted for air pollutant removal by using Mollusca shell. After pretreating processes low reactive calcium component (in the form of calcium carbonate) can be converted into calcium oxide and calcium hydroxide that readily react with acid gasses.

The application of many kinds of waste shells, which have been dried, crushed and calcined to sorption of contaminants and heavy metals in water is not only economically valuable but also very important in the aspect of waste recycling. This manner of water treatment is highly economically feasible due to not having sludge generation unlike other techniques and these adsorbents, being biological, are easy to dispose of without concerns of degradation. This is one of the green technology where there is an almost insignificant amount of energy consumption at the initial stage for the preparation of bio-adsorbents from raw materials; apart from this, the entire process is passive. The water obtained after purification can be used for various domestic and agricultural purposes, and with further treatment steps can be obtained potable quality wate

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