Journal of Environmental Sciences Studies

Journal home page: www.jess.ir

Effect of pH in Biosorption of Nickel from Aqueous Solutions by Using of Microbial and Biomass of Derived from Plants

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Received: (24-08-2020)

Accepted: (25-12-2020)

Abstract

Pollution of water by heavy metals is a major problem such as nickel. In the previous interpretations, it is compared the processes of removal and recycling of nickel metal from industrial wastewater. Research question is that which method with which PH is more effective for removal of heavy metals? Results show that biosorption, as an environmentally friendly method, has a brilliant performance and is a low-cost internal method for wastewater treatment. The biological treatment of effluents is carried out by microbial and biomass of derived from plants to examine the conversion of effluent into a harmless state. In a certain range of pH due to the operating conditions and used adsorbent are caused the reduction of H^+. Because most of the functional groups such as carboxyl and phosphate have a negative charge, they encounter nickel ions that have a positive charge, and this leads to a Electrostatic attraction is absorbed between (Ni^{2+}) cations and negatively charged coupling sites. Under alkaline conditions, adsorption decreases due to the interaction between hydroxide (OH^-) and nickel (Ni^{2+}) anions and sediment formation. Therefore, determining the optimal pH from low pH values until the formation of sediment is of great importance for operational processes.

Keywords: Biosorption; nickel; heavy metals; aqueous solutions; pH.

Introduction

With the advancement of technology and the development of economic activities, large volumes of waste containing heavy metals enter the natural environment. Heavy metals are a serious threat to the environment and public health due to their toxicity, accumulation in the food chain, and their indestructibility and stability in nature [1]. Pollution of water resources by non-degradable wastewater of heavy metals has caused great concern in recent decades [2]. The highest concerns about toxic heavy metals in industrial wastes based on WHO include nickel, cadmium, chromium, cobalt, copper, lead, mercury and zinc [1]. These metals enter as a solution in water and soil and while polluting surface water, groundwater and soil, they disrupt the ecosystems that enter it. These metals can enter the human food chain after entering the soil. In addition, they can be easily absorbed by marine animals and thus enter the human body system, which will pose a high risk to consumer health [3]. Nickel is an important and indestructible heavy metal and its excessive concentration can be dangerous for humans and other living organisms, although its presence is necessary for some enzymatic reactions and metabolic activities of the human body [2]. Nickel is a hard white-silver metal that is an abundant element in nature and can be easily combined with metals such as copper, zinc, chromium and iron to form an alloy [4]. This metal is resistant to corrosion by air, water and alkali and therefore its use has a special place in various industries [5]. Its applications include the production of stainless steel, coins, metal alloys, superalloys, non-ferrous metals, batteries, plating, tanning, paper pulp processing, pigments and mineral processing [5, 6 and 7].Excessive consumption of nickel metal causes many problems for the human immune and nervous systems, allergic skin diseases, lung problems, fibrosis and gastrointestinal disorders [2, 8]. According to the Iranian Institute of Standards and Industrial Research (ISIRI 1053), the maximum allowable amount of nickel in drinking water is 0.07 mg /liter. Therefore, due to the mentioned destructive effects, its removal from wastewater and industrial waste before discharge and discharge into the environment is vital. A number of technologies have been developed over the years to remove heavy metals from aqueous solutions. Conventional methods include ion exchange, solvent extraction, chemical precipitation, membrane filtration, electrochemical methods, reverse osmosis, adsorption, evaporation and flocculation [8].Conventional methods have low efficiency in low concentrations of metal ions, high operating cost, high energy consumption, sensitive operating conditions, incomplete metal removal, high chemical sludge production and secondary contamination [9]. These constraints, along with the environmental requirements for heavy metals, further highlight the need to develop new techniques. The development of technically and economically attractive simple methods for industrial wastewater treatment is one of the most important priorities of the 21st century. Biosorption as a method based on high efficiency of absorption, low investment and operating costs, ease of management and operation and low secondary pollution, has attracted much attention [7]. Biosorption is a low-cost, environmentally friendly technology for the separation of heavy metals using the process of attaching metal ions to the biosorbent, the efficiency of which is directly related to the adsorbent conditions. An ideal adsorbent should be economical, environmentally friendly, highly efficient, abundant and renewable. Agricultural wastes, which are mainly composed of lignin and cellulose, have been proposed as a suitable option for wastewater treatment with heavy metals[9]. Use of living and dead microorganisms and biological materials Since 1980, has been used to eliminate heavy metal ion contaminants[10]. During the biosorption process, metal ions in aqueous solution are adsorbed on the surface of the non-living adsorbent. Compared to living organisms, non-living biomasses show more advantages such as high efficiency, no need for additional growth medium and nutrients, low waste sludge [8]. Recent research has focused on adsorbents with selective metal removal, which usually have a large surface area with different functional groups as well as kinetics of rapid and suitable reaction for the removal of heavy metals. In order to improve the performance of the adsorbent, chemical modification of the surface and in-cell biological modification are used when necessary [11].In this paper, while introducing the types of microbial and plant-derived biomass along with the preparation methods required for greater effectiveness, the optimal conditions of PH for maximum biosorption of nickel ions are described based on dozens of experimental process studies.

1. Biosorption:

Biosorption is a special feature of a variety of inactive microbial biomass and plant-based biomass for the binding to biosorbent and removal of heavy metals from very dilute aqueous solutions. This process is a rapid phenomenon and the binding capacity of certain biomasses is comparable to that of synthetic cation exchange resins. Biomass with this property acts like an ion-exchanging chemical, albeit of biological origin. The basis of work in the process of biosorption is the interaction between metal ions with biomass and stabilization on it. These interactions include surface adsorption, ion exchange reactions with functional groups at the biomass surface, and surface complex reactions. Binding sites for metal ions located on the biomass surface include various groups including carboxyl, carbonyl, hydroxyl, phosphate, amine, and sulfate of fats, proteins, and polysaccharides located on the biomass surface [6, 7, 12]. It is known that the structure of the biomass cell wall is the cause of this adsorption property. In general, the biosorption process should focus on three issues: biosorbent (selection of adsorbent with high adsorption capacity, easy accessibility and low cost), adsorption mechanism and large-scale testing [7]. The advantages and disadvantages of biosorption by non-living biomass are as follows:

1.1) Advantages:

Growth-independent non-living biomass is not subject to cell contamination restrictions. This biomass does not need any nutrients for cell growth in the feed solution, so it is economically viable, so the problem of excretion of excess nutrients or metabolic products will not be raised.

- Biomass can be obtained from existing fermentation industries, which are in fact post-fermentation wastes.

This process does not involve the physiological limitations of living microbial cells.

This process is very fast and takes between a few minutes to a few hours because the non-living biomass acts as an ion exchanger. Binding of metal to biomass is often so great that it leads to highly efficient metal uptake.

- Since cells are non-living, the process conditions are not limited to cell growth. In other words, a wide range of operating conditions such as pH, temperature, metal concentration, etc. are used, while no aseptic conditions are required for this process.

- The metal can be easily disposed of and if the amount of recycled metal is significant, it can be recovered.

- Use of cheap biosorbent.

1.2) Disadvantages:

Premature saturation can be a problem. For example, when metal interaction sites are occupied, metal disposal is necessary regardless of the amount of metal before further use.

- The potential for improvement in biological processes (for example, through genetic engineering) is limited because cells are not metabolizing. Because adsorbent production occurs before the growth stage, there is no biological control over the adsorbent characteristics. This is especially true when using biomass from fermentation waste [13].

2. Biosorbent:

Bacteria, fungi, yeasts, algae, fruit skins, crop residues, activated sludge and biopolymers are various biosorbents. Researchers classify adsorbents extracted from biological sources as biosorbents. Conventional bio-sorbents must meet the following criteria:

1. High adsorption capacity and suitable kinetics [14].

2. Appropriate size, appearance and physical characteristics [15].

3. Separation of biosorbent from solution should be cheap, fast and high efficiency [16].

4. High mechanical strength, temperature stability and optimal chemical resistance [14, 17].

5. Availability of biosorbent and efficient preparation methods [18].

6. Rehabilitation and reuse [19].

Table (1) provides a list of nickel biosorbents with a summary of the preparation steps and specific properties. According to this table, the general steps of preparation of adsorbents are as follows:

1. Collection of primary biosorbents 2. Washing with distilled water and filtration 3. Drying 4. Improving the properties of biosorbents and operating groups 5. Optimizing the geometry and dimensions of adsorbents such as crushing and placing on a substrate 6. Storage in suitable conditions [10].

2. Materials and Methods

Bio-removal of nickel by inanimate and inactive microbial or plant-derived biomass is considered as an alternative and innovative technology for removing this contamination from aqueous solutions, which while solving the problems of conventional methods , includes easy availability of biosorbent with high renewability and high biosorption capacity. In this study, with the aim of introducing various types of microbial and plant-derived biosorbents in order to remove nickel from aqueous solution, the articles found between 2001 and 2020 have been used.

What is the effect of pH of solution in the use of inanimate and inactive microbial or plant-derived biomass on the biological removal of nickel?

The pH of the solution plays a very important role in the process of biosorption. The acidity of the solution, which is directly related to the pH, affects the behavior of metal ions in the aqueous solution. Changes in pH cause changes in the adsorbent surface properties in terms of functional groups, changes in the adsorbent surface charge, metallic properties and, consequently, their interaction. The pH of the solution is effective through hydrolysis, complex formation and oxidation and reduction on separation because the pH determines the adsorbent surface charge, the degree of ionization and the type of adsorption. PH can isolate interconnected sites and also plays an important role in microbial growth and enzymatic activity [10].

Table 1. Different biosorbents of nickel metal, preparation steps and their special properties

Preparation steps of Biosorbent	Advantages	Ref.
Peanut husk powder: collected→ washed, water→ dried, 80°C, 12 h→ milled→ washed, diluted HCl and NaOH→ washed, water→ dried, 80°C, 24 h.	Peanut husk is a cheap and affordable biosorbent..	**[20]**
Coconut husk: harvested→ washed→ air-dried, 24 h→ dried 70C, 7 h→ ground 100μm.	The order of priority for biosorption of ions on the Coconut Husk is based on various parameters, such as electronegativity value, hardness/softness value, ionic radii, hydration energy and electron affinity etc. of the metal ions. Low cost.	**[21]**
Myriophyllum spicatum: harvested→ exposed to air→ dried, couple days, room temperature→ dried at 60°C, 6 h→ crushed 0.2 mm.	-	**[22]**
Corn silk: powdered 150–180 μm , 0.5 g + 15 ml HNO3 0.5 M, ultrasonication, 4 h→ centrifuged→ washed→ dried 70°C, 4 h	This biosorbent was tested for synthetic waste water treatment and results showed that corn silk can be used several times after regenerating with good efficiency.	**[23]**
Dicerocaryum eriocarpum leaves: dried→ boiled water, 24 h→ dehydrate, 25°C→ mucilagesolution filtration→ + KCl or NaCl→ filtration→ dried.	Both modified and raw biosorbents provide successful results. High biosorption efficiency in mucilage may not be highly dependent on the viscous nature of the mucilage, but rather the increase in the ionic strength of the mucilage. Dicerocaryum eriocarpum mucilage is suitable for households in rural	**[24]**

	community, because it's there at a low cost easily accessible.	
Trichoderma: isolated, soil, modified Trichoderma-specific medium 14, dilution plate method.15→ collected→ air-dried→ powder.	Of the fourteen Trichoderma isolated, threecases were resistant to high levels of cadmium and three cases of nickel, the results showed that they can be used to treat agricultural soils contaminated with nickel and cadmium.	**[25]**
Pseudomonas: isolated→ All strains were stored −80 °C, sterile glycerol→ grown→ Agar Nutrient, meat extract, peptone, agar→ + 10 µM Hg→ grown aerobically, 30°C, 24 h→ kept at 4°C.	-	**[26]**
Alginate-extraction residue: brown seaweed Sargassum filipendula→ collected→ washed→ dried 60°C, 24 h→ 15 g + formaldehyde, 500 mL, 30 min→ rinsed water→ + hydrochloric acid 0.1 M, 500 mL, 1-2 h, shaking→ + sodium carbonate 20 g L-1, 60C, 100 rpm, 5 h→ alginate extraction→ vacuum filtered.	The kinetics of the process is controlled by the internal mass transfer resistance.	**[27]**
Nocardiopsis sp. and *Nocardia* sp.: Actinomycetes, isolated→ incubated, 7 d, 28°C→ identified→ 69 Actinomycete isolates→ MORSY1948 and MORSY2014, selected.	Dead biomass has a higher efficiency than living cells, also if use 4% more of optimum amount of dosage, the biosorption will be 100%.	**[28]**
Immobilization of dried staphylococcus aureus: dried bacterial cells powdered 4 g + magnetic Fe3O4-phthalate nanoparticles 10 g + 5 mL water, grinding, 15 min→ mixture, dried 60°C.	The amount of biosorption capacity on the biomass for heavy metal ions was as follows: Pb (II)> Ni (II)> Cu (II). The interaction of Ca(II) and Mg(II) in the reaction medium reduces the amount of biosorption capacity.	**[29]**
Free and immobilized marine algae *Sargassum* sp.: *Sargassum* sp. powdered + sodium alginate solution→ mixture, dropped, CaCl2.2H2O→ beads, kept, 4 h→ washed→ stored room temperature.	Calcium alginate matrix, by immobilizing biosorbent, increases the biosorption capacity also immobilization makes the packing easy but porosity is low. Immobilized biosorbents are god alternative for fix bed columns	**[30]**
Magnetic biosorbent based on oil palm empty fruit bunch fibers, cellulose and Ceiba pentandra: oil palm empty fruit bunch + cellulose→ homogenized, mill, 0.005-0.02 mm. Ceiba pentandra→ dried 70°C→ milled, 30 min, 450 rpm, 0.1–0.002 mm. 50 mg Fe_2O_3 + 100 mL of materials (5 mg mL-1), sonicated, 5 min→ washed→ stored at room temperature.	Magnetic nanoparticles are well spread on the surface of the base material, this adsorbent with regeneration ability to use up to 5 times.	**[31]**
Lemon peel: (1.25-2 mm) Grinding then drying at 60. Then rinse with distilled water and then Collecting. 4 grams of chopped lemon peel+ 100 mlit (H3PO4, HCL, HNO3, NaOH, Cacl2, NH3) Rinse with distilled water and store at 30 ° C	Improvement of adsorption correction with NH3 and NaOH alkalis was observed to be about 2.5 times unmodified - cheap adsorbent	**[32]**
Non-living streptomyces roseorabens : Inoculation of microorganisms in liquid culture medium Collection of biomass mixed in shaker at 28 ° C at 120 rpm for 48 hours Crushing dry in an oven at 100 ° C for 6 hours, rinse with deionized water	Very high absorption capacity and cost-effective	**[9]**

Screening		
Aspergillus niger Aspergillus niger frozen and dried then Culture medium sterilized at a pressure of 1.5 bar at 121 for 20 minutes Incubation inside the shaker at 200 rpm at 30 for 5 days Rinse with autoclave filtration water at 121 and 15 minutes Crush to 100 mesh Dry at 50 ° C for 24 hours Hold at 0.5 ° C boiling for 20 minutes Drying in 50 for 24 hours Rinse with centrifugal water at 4000rpm Powdering	-	**[7]**
Dry activated sludge Growth of microorganisms in liquid culture medium under agitated and aerated conditions at 150 rpm and temperature 25 ℃ Dry at 60 ° C for 24 hours. Rinse with distilled water 5 grams of dried biomass Homogenize by homogenizer at 8000rpm for 20 minutes with distilled water	-	**[6]**
Mango shell Powder Drying Wash the mango skin with distilled water Drying at a temperature of 150 + 150 ° C for 24 hours mixing with sulfuric acid Immerse in sodium bicarbonate for 24 hours, rinse with distilled water Washing sieve	A simple process and as a suitable alternative to its expensive-abundant adsorbents in the tropics and the peel content of 60% of the total fruit	**[33]**
Brown seaweed Dry at 60 ° C Wash with distilled brown algae water Crushing to the size of 1.5 mm	High metal absorption, low cost, renewability and abundance	**[5]**
Rice husk combined with humic acid Drying at 50 ° C for 12 hours Rice husk collected from the	Increasing the absorption rate due to the additional correction method	**[34]**

mill Stir for 1 hour at 70 ° C and then cool, adding the soda Filtration Add rinsing hypochlorite with soluble filter water Convert to IR drying powder Filtration, adding humic acid and keeping the mixture overnight in a rotating shaker Dry to a moisture content of less than 0.1%. Rinse with baking soda and then with water		
Palm oil waste and Al2o3 nanoparticles Rinse with water and dry for 24 hours, shredding the waste into smaller pieces	Removal of 81% Nickel - Adding Al2o3 also increases nickel absorption by 5.9%	**[35]**
Add dimethyl sulfoxide to biomass at 120 rpm for 24 hours Add tetraethyl silicate and nanoparticle powder to suspension and stir at 120 rpm for 12 hours Dry at room temperature with ethanol and distilled water	-	**[36]**
Rotten white fungus (P.chrysosporium PTCC5270)(Culture in PDA culture medium at 30 ° C for 24 hours Shake at 34 rpm and add biomass + PDA liquid Sterilize with autoclave at 121 for 20 minutes	89.48 %. efficiency of nickel removal, low cost and efficient adsorbent	**[8]**
89.48 %. efficiency of nickel removal, low cost and efficient adsorbent of olive oil factory waste Dry at room temperature Wash biomass with distilled water Sieve the size of 0.25-0.15 mm then mill	Eco-friendly, affordable and affordable	**[3]**
:Bakery yeast Mix with Hcl 0.1N for 24 hours Rinse several times with distilled water Dry in the oven at 6060 for 24 hours	-	**[37]**

Curtobacterium SP: Microbial culture in lactose broth medium with sugar incubator at 30 ° C at 180 rpm for 24 hours Centrifuge at 5000 rpm for 10 minutes Dry wash with distilled water (3 times)	High nickel absorption	**[22]**
Sugarcane pulp Dry at room temperature with distilled water Mill size less than 1mm	Abundance, high availability and low cost Appropriate absorption capacity	**[38]**
Pseudomonas aeruginosa : Culture in acetamide liquid culture medium at 37 ° C for 48 hours		**[39]**
Bacillas laterosporus: Culture in liquid culture medium under aerobic conditions and stirring at 30 ℃ for 2 days Centrifuge at 4 و and 8000 rpm for 2 minutes Dry at 60 ° C for 24 hours. Rinse with distilled water Store powders	Bacilli are a good choice compared to other microorganisms due to their high metal adsorption potential, high adsorption rate and reuse potential.	**[40]**
Brown algae Rinse with distilled water drying in the sun Screening to a size of 1-1.25 mm Dry at 60 ℃ overnight	-	**[1]**
Grapefruit peel Rinse with distilled water crush into small pieces collect the skin of the fruit Screening less than 355 μm Drying at 70-80 ° C for 24 hours	Fast adsorption rate, recovery of metal ions from the adsorbent, reusability of the adsorbent several times, low cost	**[2]**
: Seaweed Drying in the sun Wash with distilled water Convert size 0.767 mm	Usability Re-adsorption, low cost, high biomass efficiency	**[41]**
: Straw barley Sift crushing drying in the sun	High absorption capacity, abundance-available	**[42]**
Brown algae Dry in the sun for 48 hours, rinse with water Rinse with double distilled water, sieve to a size of 0.5-1 mm, crush	Abundance and low cost	**[43]**
Drying at 80℃ for 24 hours		
Modified algae Dry in the sun for 3 days at 343k for 40 minutes rinse with water Mix with 0.1m hydrochloric acid solution with stirrer at 200rpm for 8 hours at room temperature	Effective adsorbent for nickel absorption due to economic efficiency	**[44]**
(Enteromorpha prolifera) brown algae Rinse twice with water Dry in the sun for 1-2 days and then dry at 105℃ for 24 hours in the oven Pour in distilled water and use a blender to increase the surface area	Abundance and available in nature	**[45]**

3. Results & Discussion

1) Heavy metals including nickel due to toxicity, accumulation in the human body and other living organisms, stability and non-degradability is a serious threat to the environment and public health.

2) Conventional methods of separating heavy metals from wastewater and aqueous solutions at low concentrations of metal ions are expensive and inefficient.

3) Biosorption with non-living microorganisms and biomaterials has more benefits than living biosorbent.

4) The abundance and low value of biosorbents justifies their use economically.

5) The use of physical and chemical methods to modify the surface of the biosorbent and the internal modification of the cell and the combination of biomass with other materials, has a significant effect on increasing the efficiency of adsorption.

Table 2 shows the optimal pH values associated with different biosorbents for nickel metal in previous studies. The amount will change according to the operating conditions. Nickel is generally in the form of (Ni^{2+}) cation, its adsorption efficiency at low pH is clearly negligible.

Table 2: Optimal pH(between 3-11) in some studies for nickel uptake

Biosorbent	PH	Ref.
modified lemon peel	5	**[9]**
Non-living Streptomyces roseorubens Sy	4	**[7]**
Mangosteen shell activated carbon	5	**[5]**
Oil Palm residual biomass and AL_2O_3	6	**[36]**
Aspergillus niger	6.25	**[6]**
dried activated sludge	4.5	**[33]**
white-rot fungi	6	**[8]**
brown algae Sargassum sp	6	**[31]**
waste pomace of olive oil factory	4	**[3]**
Baker's yeast	6.75	**[37]**
Thermophilic Geobacillus stearothermophilus	6	**[4]**
rice bran	6	**[12]**
Curtobacterium sp	5	**[22]**
sugarcane bagasse	5	**[38]**
Pseudomonas aeruginosa Asu 6a	7	**[39]**
Bacillus laterosporus	7	**[40]**
brown algae	6	**[1]**
grapefruit peel	5	**[2]**
Marine algae	4	**[41]**
Aspergillus sp	4	**[46]**
barley straw	4.85 ± 0.1	**[47]**
chemically modified brown algae	6	**[43]**
treated alga (Oedogonium hatei)	5	**[44]**
chemically modified brown macroalgae	4	**[48]**
Algea (Chlorella Vulgaris)	4.5	**[33]**
(Enteromorpha prolifera green algae)	4.3	**[45]**
Coconut husk	6	**[21]**
Arthrospira Platensis	5	**[49]**
(Corn Silk)	6	**[24]**
Dicerocaryum eriocarpum leaves	4	**[25]**
Trichoderma	5	**[26]**
Alginate-extraction residue	4	**[28]**

4. Conclusions

The presence of toxic metals in the environment has detrimental effects on human and animal health and disrupts the balance and order of the ecosystem. Therefore, it is necessary to study the ways to eliminate these pollutants. The aim of this study was to compare nickel metal uptake by biological uptake methods with the help of microbial and plant derived biomass. Based on the previous interpretations, It is compared the processes of removal and recycling of nickel metal from industrial wastewater. Research question is that which method with which pH is more effective for removal of heavy metals? Obtaining the optimum pH of the solution is very important in practical processes. Table 2 shows the optimal pH values associated with different biosorbents for nickel metal in previous studies. The amount will change according to the operating conditions. Nickel is generally in the form of (Ni^{2+}) cation, its biosorption efficiency at low pH is clearly negligible. At low pH which leads to an increase in the concentration of positive etching ions the active sites on the biosorbent surface are significantly occupied by H^+ ions. In this case, due to the competition between H^+ and (Ni^{2+}) ions, the biosorption of nickel by the biosorbent is limited and as a result decreases. On the other hand, due to the positive charge of both ions, the repulsive force is also involved in reducing the binding of nickel to active sites and the rate of adsorption. In a certain range of pH due to the operating conditions and biosorbent used and the reduction of (H^+), because most of the functional groups such as carboxyl and phosphate have a negative charge, they encounter nickel ions that have a positive charge, and this leads to a Electrostatic attraction between (Ni^{2+}) cations and negatively charged sites and biosorption capacity increase. Under alkaline conditions, biosorption decreases due to the interaction between hydroxide (OH^-) and nickel (Ni^{2+}) anions and sediment formation. Therefore, determining the optimal pH from low pH values until the formation of sediment is of great importance for operational processes. The results clearly show that the range of low (very acidic) pH and alkaline pH are not favorable for the reasons stated that lead to a decrease in adsorption capacity.

Acknowledgements

The authors of the article would like to thank Tonekabon Branch, Islamic Azad University for supporting this research.

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