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Research article Curtailment of Mercury ions using Punica Granatum Fruit Peel Sonam Kumari¹, Gaurav Sharma¹, Mudit Gupta²

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Abstract

Due to their poisonous nature and difficulty in biodegrading, heavy Chemical metals have developed into a significant cause of water Modification, contamination. Pomegranate Peel (PP) was employed in this study as **Bio-adsorbent** a bioadsorbent to remove heavy metals from tainted water. The fundamental issue with Pomegranate Peel bioadsorbent is that, compared to its commercial equivalent, biomass-derived activated carbon has a relatively poor adsorption capacity. A significant amount of leftover pomegranate peels (PP) also makes disposal more challenging. Thus, it is anticipated that the current effort, by converting pomegranate peel into bioadsorbent, will address the issues connected to pomegranate peel disposal. The purpose of this research is to create a bioadsorbent from fruit peels and evaluate how well it can absorb heavy metals. In order to evaluate the bio-capacity of the adsorbent to remove Hg ions, we also optimised the pH, solution temperature, adsorbent dosage, contact duration, and solidliquid ratio. It is strongly advised that low-cost raw materials be employed widely as bioadsorbent in wastewater operations due to their simplicity of processing, wide availability, and environmental friendliness.

Introduction:

In many industrial operations, such as metal finishing, electroplating, painting, dyeing, photography, surface treatment, and the creation of printed circuit boards, heavy metal ions are employed. When heavy metal ions are discharged into the environment untreated, they represent a risk to human health as well as serious harm to the aquatic ecology. The bulk of heavy metal ions have well-known carcinogenic toxic and tendencies (1-4). Heavy metals (HMs) are substances that have an increased atomic mass and density, such as cadmium (Cd), zinc (Zn), mercury (Hg), arsenic (As), silver (Ag), chromium (Cr), copper (Cu), iron (Fe), and platinum (Pt). Heavy metal water pollution is one of the most significant environmental problems affecting people, animals, and plants (5). Heavy metals are harmful even in tiny concentrations because they cannot be broken down by the body. (6)Metals and metalloid ions were divided into three groups. The first category contains metals that can be harmful even in small levels, such as lead, cadmium, and mercury. The second group of metals, which includes bismuth, indium, arsenic, thallium, and antimony, is less dangerous. The third group of metals, which includes essential elements like zinc, cobalt, copper, iron, and selenium that are required for a number of biochemical and chemical processes in the body but which are only toxic in excess amounts, is more dangerous. (7) Hyperaccumulator plants may retain hazardous metals in high amounts that might endanger human and animal health by poisoning the food chain.

Mercury is a heavy metal with a well-known toxicity profile that has been linked to public health catastrophes in Iraq and Minamata Bay, Japan (8-11). Smaller mercury exposures' effects on health are still debatable. Inorganic mercury, which comprises metallic mercury, mercury vapour, mercuric salts, and mercurous (Hg^{2++}) or mercuric (Hg⁺⁺) salts, is one of its many forms. Organic mercury, on the other hand, refers to compounds in which mercury is bound to a structure made of carbon atoms (methyl, ethyl, phenyl, or similar groups).

Chemical structure affects the biological activity, pharmacokinetics, and therapeutic importance of the many forms of mercury. In vivo, there is some interconversion between the several mercury forms. For instance, elemental mercury vapour inhaled into the lungs is easily absorbed and soon transforms into various forms (albeit not quickly enough to avoid significant deposits of elemental mercury in the brain). Although methyl mercury crosses the blood-brain barrier less effectively than elemental mercury, it accumulates in numerous tissues and is readily absorbed via the gut .Methyl mercury is gradually converted back to elemental mercury once it reaches the brain. In contrast, mercury salts often have an insoluble nature and are poorly absorbed.

The kind of mercury, the amount, and the pace of exposure all affect how dangerous it is to humans. The brain is the primary target organ for breathed mercury vapour . While methyl mercury is extensively disseminated throughout the body, mercuric and mercuric salts mostly harm the kidney and gastrointestinal lining. The severity of the toxicity depends on the dosage: significant acute exposures to elemental mercury vapour cause severe pneumonitis, which in severe cases can be deadly. Subtler symptoms and clinical findings are brought on by low-grade chronic exposure to elemental or other forms of mercury, as will be explained below.(12) There are several techniques documented for removing heavy metal ions, including membrane processes, solvent extraction, and precipitation reduction (13-15). However, techniques have a number of these drawbacks, including the creation of

hazardous sludge and insufficient metal

removal. Using bio-adsorbents, a new

technique for eliminating heavy metals,

represents a significant advance (16,17). The capacity of biological materials to absorb metals from wastewater heavy via metabolically mediated or physico-chemical absorption mechanisms is known as bioadsorption. Due to its potential to be more selective, more effective, simple to use, and cost-effective for the treatment of vast volumes of wastewaters with low pollutant concentrations, this phenomenon, known as bioadsorption, appears to be a promising alternative to the current approaches (18,19). Fruit peels are more advantageous from an economic and environmental standpoint when used as adsorbents since they are more readily available. accumulate less agricultural waste, can be recycled, and can be used to extract metals from other materials. The main residue is pomegranate peel (PP), which accounts for 30-40% of the weight of the fruit and is one of the most popular fruits in the world (20).

Pomegranate peel is a typical byproduct that is cellulose and mineral-rich. Large volumes of PP cause considerable resource waste and disposal problems. It is known that a number of functional groups, including carboxyl, hydroxyl, and amide groups, that are present on the PP surfaces play a crucial role in the biosorption processes. Additionally, it has carbon at 41.37%. (21). The disposal of PP can be done in an appealing way by recovering it as sorbent. Because of its porous structure and variety of surface groups, purified and treated PP has been used as a sorbent to remove dissolved heavy metals from wastewater (21). Due to its inadequate adsorption capacity, pure PP still has to be subjected to alkaline and acid chemical treatment. In this study, PP and pomegranate stalks were used by a number of research organisations for the removal of Hg,

Materials and methods Pomegranate peel adsorbent creation:

The PP was purchased at a neighbouring market and cleaned with distilled water two or three times to get rid of dirt and debris. It was then broken up into little pieces and given time to dry naturally outside in the sunlight. The peels were roasted at 150°C in an oven to get rid of dampness and facilitate grinding. Then, a crushing procedure and just one filter were used to eliminate the particles larger than 0.170 mm in diameter. The final solid mass was then kept until use in a desiccator.(25)

Bioadsorbent amendment using various chemicals:

The solid support is mixed with 1 N nitric acid solution in a weight-to-volume ratio of

a dangerous heavy metal ion, from aqueous solutions and industrial wastes (23,24).

In the current work, fresh PPs were used as raw materials, and they were then treated with acidic and basic solutions at different concentrations to develop efficient bioadsorbent. As a result, PP bioadsorbent was created, and its ability to remove heavy metals was tested. It has functional groups on its surface that contain oxygen.

1/2 in order to activate the bio-adsorbent, also known as (Acidic Pomegranate Peel) APP, so that it may go through a 24-hour thermochemical treatment at 120 °C. The excess acid was then filtered out of the mixture using distilled water, and the leftover acid was eliminated by soaking the mixture for an extended period of time in a 1% solution of NaHCO3. After that, the sturdy support was dried in an oven set at 105°C (25). Basic Pomegranate Peel, also known as BPP, is created by mixing 200 g of PP with 1 litre of 0.1 M caustic soda solution for the activation. After sitting for 24 hours, the BPP is then rinsed with distilled water until it reaches a neutral pH. (26,27). The material that had been treated with NaOH was then dried in an oven at 120 °C.

Preparation of Hg solution:

To create Hg(II) solution, HgCl₂, an analytical-grade chlorinated salt, was utilised. The aqueous solution of mercury chloride (HgCl₂) was made using distilled water. Test samples at different concentrations were made from this stock solution for tests on various parameters.

Experimental set-up and analysis:

The batch approach was used to conduct experiments for the adsorption of Hg from aqueous solution at room temperature. Initial Hg (II) concentrations were produced in the range of 5, 10, 20, 30, 40, 50, 70, 100, and 150 mg/L. Each adsorption experiment began with a 100 ml flask containing 100 ml of the required amount of Hg(II) aqueous solution. The pH of PP, APP, and BPP were adjusted by adding 0.1 N (or 1 N) HNO₃ and 0.1 N (or 1 N) NaOH. These 100 ml flasks were shaken at 200 rpm for one hour.

The effects of pH, the dosage and volume of the biosorbent, the initial concentration of Hg(II), the contact time, the rate of agitation, the temperature, and the pretreatment of peel were some of the experimental factors employed in the batch investigations. The effect of the adsorbent mass was investigated by varying the weight of the PP between 0.1 and 1 g. Additionally, different contact periods with Hg ions solutions were used to evaluate the PP's Hg adsorption capability (15 min, 30 min, 1 h, and 2 h). In all cases, the initial metal ion solution concentration of 50 mg/L, pH of 4, and temperature of 25 degrees Celsius were maintained.

% Removal and Metals Uptake Capacity Assessment

The concentration difference method was used to calculate the Hg ion absorption (24). The adsorption capacity, or q, is defined as the amount of metal ion (mg) that is absorbed per g (dry weight) of PP. The equilibrium concentration of metal ions in solution is Ce, the volume of the used metal ions solution is V(L), the starting concentration of metal ions is Ci (mg/L), and the weight of the adsorbent is W. (g).

The percentage metal absorption by the sorbent and adsorbent capacity at equilibrium qe (mg = g) were calculated using the following equations.

% Adsorption= $(C_i-C_e)*100/C_i$

Results and discussion: Effect of Biosorbent Particle Size:

Because they altered the total surface area accessible for the sorption of metal ions, the particle sizes of the biosorbent had a substantial effect on its sorption capacity. Smaller particles efficiently removed more Hg(II) than bigger ones, according to research on the effect of sorbent particle size on sorption capacity, q (mg=g).

As can be seen in the image below, finely ground biomass absorbed Hg (II) ions more fast. Equilibrium was reached more rapidly with smaller biosorbent particles than with larger ones. This was most likely brought on by the rise in surface area overall, which provided the metal ions with more sorption sites (29-31).



Figure 1: Effect of particle size on adsorption of Hg ions.

The impact of primary treatments:

To determine the effect of pretreatment on PP waste biomass, 25 mg/L of Hg(II) were shaken at 200 rpm for two hours with 0.1 g/L of pretreated bio-adsorbent with a size of 0.155 mm for Hg(II). The q values of untreated, physically and chemically changed PP waste biomass for Hg(II) sorption are

shown in the figure below. Because of the removal of mineral matter and the creation of new sorption sites on the biomass surface during the boiling process, the sorption capacity of the biomass has increased. Heat treatment of biomass decreased metal uptake because it inhibited intracellular absorption. (29).

The polymeric structure of biomass surfaces exhibits a negative charge due to the ionisation of organic and inorganic groups (30-32), and at a certain concentration, acids can increase the surface area and porosity of the initial sample, enhancing the absorption capacity of biomass. These two factors are crucial in determining the sorption capacity of a particular biomass following acidic pretreatment (28,33). Similar to how basic pretreatment has two factors that affected the bioadsorbent's propensity to absorb. It may harm autolytic enzymes in addition to eliminating lipids and proteins that cover up reactive sites and purifying the biomass (34-38). Additionally, the amount of protein amino groups that might take part in metallic ion binding drastically decreased beyond a certain alkali concentration. Deproteinization should theoretically reduce metal retention (38).

	% ADS
Native	61
HCI	69
Nitric Acid	82
Sulfuric Acid	92
NaOH	84
Ethanol	76
Methanol	82
Acetone	56



Figure 2: Effect of pre-treatment on adsorption of Hg ion

Result of pH:

Pomegranate peel that has been dried and used in an experiment to find out how pH influences Hg adsorption. To do this, combine 0.1 g of dry PP and 0.1 g of chemically changed PP powder with 100 ml of a 25 mg/L Hg ion solution (particle size: 0.160 mm). For pH adjustments between 3 and 10, samples were shaken for 8 hours at room temperature at 250 RPM using 1 N HNO3 or 1 N NaOH. Additionally, it influences the surface charge and functional group dissociation of the adsorbents (25, 26). The PP waste biomass's adsorption capability increased as the pH of the solution rose.

Due to competition between Hg ion and H+ for the same surface active sites as a result of the adsorbent's protonated surface active sites, Hg absorption was very low. When the biomass concentration was fixed (0.05 g/L) and the pH rose from 3-6, the Hg ion absorption increased shown in the figure below. As can be observed, the Hg ion's ability to bind to surfaces is lowest at pH-3 and increases to as high as 95% by pH-6. As pH rose further, the absorption of Hg ions decreased. Low adsorption percentage removal below pH-6 is anticipated and can be explained by a variety of mechanisms, including (a) repulsion between the positive charge of the sorbent and free Hg ions, (b) competition between free Hg ions and H+ for the sorbent's active sites, and (c) a decreased capacity to form complexes with metal ions as a result of protonation of surface functional groups.

рН	PP	BPP	APP
2	25	45	54
3	39	52	72
4	73	89	95
5	65	85	89
6	60	80	86
7	55	75	78
8	50	68	73





Effect of agitation speed:

The effect of agitation speed on the removal of Hg ions by PP, APP, and BPP is depicted in the figure below. The agitation speed was kept constant at 50–250 rpm. Other factors, such as pH (pH 4), contact time (20 min), and temperature (25 °C), were remained constant for PP, APP, and BPP. In terms of mg/g, more Hg ions were adsorbed as the agitation speed increased. The optimum outcomes for the removal of Hg ions were found at a speed of 250 rpm. For the remaining experiments, agitation was carried out at a speed of 250 rpm.

Speed	PP	BPP	APP
50	43	48	50
100	52	64	68
150	75	95	90
200	88	97	98



Figure 4: Effect of speed on adsorption of Hg ion.

Effect of adsorbent dose:

Figure above depicts the mass of modified and unmodified PP and how it affected the ejection of Hg ions from aqueous solution. Adsorbent was used in doses ranging from 0.1 to 1 g for 100 mL of a Hg ion solution. Other parameters, such as temperature (25) °C), contact time (20 min), and pH (pH 4), were maintained constant. The results reveal that while Hg ion removal % increases as adsorption adsorbent mass increases, capacity decreases, as seen in the figure below. This is due to the fact that increasing the dose of both modified and unmodified PP results in more active sites being accessible for interaction with metal ions. a rise in the percentage of metal ions eliminated from the aqueous solution as a result. Conversely, aggregation of modified and unmodified PP inside larger dosages of adsorbent may cause unsaturation of active sites, which may result in a decrease in adsorption capacity. The surface area of the adsorbent is decreased by this aggregation.





Effect of contact time:

In the figure below, the effect of contact time on copper removal by PP is shown. The curve has a traditional saturation curve shape. The three support ports activated by sulfuric acid and caustic soda (NaOH) reached saturation in the first five minutes or so as the Hg ion was quickly adsorbed. However, the natural support has a much slower rate of adsorption, and saturation takes around 10 minutes to reach. This is accounted for by the adsorption sites' initial emptiness, which makes it easy for metallic ions to quickly fill them and offer a high adsorption rate. The delayed adsorption beyond this first interval may be due to a slower diffusion of dissolved species via the material's pores.



Figure 6: Effect of contact time on adsorption of Hg ions.

Time	PP	BPP	APP
10	58	80	85
20	65	85	93
30	64	87	93
60	65	86	93
120	64	86	94
240	64	86	93
360	63	86	95
720	64	86	93
1440	63	87	93

Effect of temperature:

The figure below shows how temperature affects the adsorption of Hg(II) by pomegranate skin. At 35 °C, the greatest Hg(II) adsorption was achieved. The increase in solution temperature boosted ability to

adsorb, showing that the process was endothermic. Temperature rise decreased the thickness of the PP surface layer and accelerated the pace at which Hg(II) ions moved from the solution onto the open sites of PP.





Conclusion:

With the help of pomegranate waste, we have developed a bio-adsorbent that can take out Hg ions from aqueous solutions. We may change a variety of reaction parameters, including the solid-liquid ratio, pH, solution temperature, and amount of adsorbent used. It has been demonstrated that mercury removal from pomegranate peel is very efficient, economically viable, and affordable.

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