Application Of The The Pumpkin Seed Husk (*Cucurbita Maxima* Dutch) By The Removal Of Chromium (VI) In Solution

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***Abstract*—The pumpkin seed is the edible seed of the fruit of some species of the *Cucurbita* genus of the Cucurbitaceae family that has been dried. Native varieties and races of pumpkins are grown in practically all agricultural regions, accompanying corn and beans in what is known as milpa. The plant is native to northern Mexico, and southeastern and eastern United States. It can be used as an antioxidant, liver protector, anticancer and antiparasitic. Too, the potential use of their pumpkin seed husk, for accumulate heavy metals has been analyzed. So, the objective of this work was analyzing the Cr (VI) removal capacity in aqueous solution by the pumpkin seed husk by a colorimetric method.**

Biosorption at different pH was evaluated for 8 hours. We too studied the effect of temperature in the range of 28 to 60^{o}C, the removal at different initial concentrations of Cr (VI), biomass, and in contaminated niches. Therefore, the highest biosorption of the metal (50 mg/L) occurs within 8 hours, at pH of 1.0, 1.0 g of analyzed biomass, and 28°C. With respect to the incubation temperature, it does not influence the removal, since 100% of it is eliminated after 150 minutes at 40°C, 50°C, and 60°C, and the heavy metal concentration, has no effect on the removal of chromium (VI), although efficiency is greater at 40°C, If the biomass concentration increases, too increase the removal of the metal in solution. The metal can be desorbed, if the biomass is incubated in alkaline solutions, with similar desorption percentages in 0.1N and 0.5 N NaOH. Finally, this natural biomass removal efficiently the metal *in situ* (93% and 94.75% in soil and water contaminated, after 7 of incubation), with 5 g of biomass and 28^{o}C; so, it can be used to eliminate it from industrial wastewater.

Keywords—Chromium (VI), Removal, Pumpkin, Elimination

I. INTRODUCTION

In Mexico, we call pumpkin seeds pepitas that are collected from the ripe fruit and left to dry in the sun. Its consumption with or without shell dates back to pre-Hispanic times and with it they have been made since then: moles, pipianes, and typical Mexican sweets. In many places in the country, they can be found in small stands, where they sell toast (on the comal) with salt and with their white peel (or without it) to be enjoyed as a snack [1]. Many people generally consume them peeled because it is easier to taste the seed extract; However, what they don't know is that they are wasting a great source of nutrients [1]. This part that covers the seed offers a greater amount of proteins, fibers, vitamins and minerals than the pulp and leaves of the pumpkin itself, and according to a study carried out in the USA, this peel has substances called carotenoids, which prevent heart diseases [2]. They also have a protein that has antifungal effects, as they eliminate intestinal parasites and infections, likewise, it improves the functioning of the intestine and its unsaturated fats reduce blood cholesterol and balance blood pressure [3]

Pumkin seed is a nutritional food with high oil (50% w/w) and protein (35%) content that varies depending on the cultivar, Palmitic (≤15%), stearic (≤8%), oleic (≤47%) and linoleic (≤61%) fatty acids are the main components of the oil, while albumins and globulins constitute approximately 60% of the crude protein [4]. The oil and fatty acid content of pumpkin seed is comparable to that of soybean (*Glycine max*), sunflower (*Helianthus annuus*), safflower (*Cartamus tinctorius*), and watermelon (*Citrullus lanatus*) [4]. The high levels of unsaturated fatty acids (oleic and linoleic acids) in pumpkin seed oil provide health benefits that reduce the risk of arteriosclerosis and related heart problems [3]. Pumpkin seed contains significant levels of antioxidants (tocopherols and tocotrienols) that have been associated with reduced risks of gastric breast, lung, and colorectal cancers [5]. Additionally, phytosterols in pumpkin seeds play a key role in reducing cholesterol levels and treating enlarged prostates (benign prostatic hyperplasia) [3].

The usefulness of pumpkin seeds in different stages of human development is not new, although today, there is a comprehensive demand and modern to carry out the production process of this raw material in agroindustry, starting from the post-harvest process to final consumers, using mechanical and automated technology. Without owever, to achieve an effective process, it is advisable, know and interpret the physical and chemical characteristics and biological to guarantee better productivity, associated with a reduction in the use of resources employees [1].

Pumpkin seeds have become one of the most consumed snacks in the country. Although some people consume it raw, that is, freshly extracted from the fruit, they are also subjected to processes to modify their flavor and texture in such a way that they can be found in presentations such as toasted, cooked, baked or even caramelized with piloncillo [6]. Another use given to the seeds is the preparation of pastas such as moles popularly known as "pipianes". To do this, once extracted from the pumpkin, the seed must be toasted and go through another process that involves crushing it to use it as the main condiment [1, 6]. Various components such as proteins, fiber and vitamins A, K and B, as well as omega 3 and 6, can be found in the pumpkin seed. Likewise, there are minerals such as magnesium, manganese, iron and zinc. In addition, it has antioxidant properties that, due to its high level of alkalinity, make it an ideal food for people with conditions such as diabetes [3, 7].

On the other hand, the great industrial growth has produced a progressive increase in wastewater discharges from the same and, heavy metals are the main contaminants of aquifers due to its high toxicity, persistence, and mobility. Not being biodegradable, can become toxic to vertebrates and invertebrates, and they are directly related to the risks to the health of living beings, soil contamination, plant toxicity, and negative effects on the quality of natural resources and the environment. These risks are related to the specific toxicity of each metal, bioaccumulation, persistence and non-biodegradability, the greatest danger being its accumulation in plants and its transfer to humans and animals [8]. Their distribution in the different environments is highly complex and involves different factors, among which are: redox potential, pH, organic matter content, cation exchange capacity, groundwater level and its fluctuations, among others [9], and there are different investigations for carry out to determine the contamination of heavy metals in the environment, such as: cobalt, lead, mercury, chromium (VI), cadmium, and others, like: heavy metals potentially dangerous for human and environmental health, contamination of water sources. Others contaminants are of organic origin, such as hydrocarbons and pesticides, but, all are their importance and potential danger [10]. Some metals that are of great toxicological and ecotoxicological importance are: mercury, chromium, lead, cadmium, nickel and zinc, which, once released into the environment, accumulate and concentrate in the soil and sediments, where they can remain for hundreds of years affecting ecosystems. Therefore, it is more feasible to control the problem from the source and source of emission before they reach the environment [9]. In Mexico, agribusiness is one of the most important activities due to its growth in recent years, and it is the one that generates the most by-products that are not used [11], among which are: coffee bagasse, agave, maguey, sugar cane, straws from different crops, organic residues of fruits and vegetables [12]. In this regard, the use of different plant products with the ability to accumulate and/or bioadsorb heavy metals has been reported, which include pumpkin seed husk (*C. maxima*), for example: lead removal by activated carbon [13], the accumulation of some heavy metals in presence of citric acid, compost, and others [14, 15, and 16], the removal of synthetic dyes [17], and pesticides [18]. Therefore, the objective of this work was to analyze the removal capacity of chromium (VI) in solution, by the biomass of pumpkin seed husk (*C. maxima*),

II. EXPERIMENTAL

A. Biosorbent used

The pumpkin seed husk (*C. maxima*) biomass, was obtained from street sellers from the marketplace Republic, between the months of March to May in 2022, of the capital city of San Luis Potosí, S.L.P. México. To obtain the biomass, the peel was washed with EDTA 10% (p/v) for 24 hours, and after with trideionized water during 3 days at constant stirring, with water changes every 12 hours. Subsequently, it was boiling 1 hour to removal traces of the color and dust and were dry at 80°C for 72 hours in an oven, ground in blender and stored in amber vials until use.

B. Biosorption studies and determination of hexavalent chromium.

For these studies, was used 1 g of dried biomass mixed with 100 mL of trideionized water containing 50 mg/L of the metal, in an Erlenmeyer flask at the desired temperature and pH. The flasks were agitated on a shaking bath Yamato BT-25 model. Samples of 5 mL were taken at different times, and centrifuged at 3000 rpm for 5 min. The supernatant liquid was separated and analyzed for chromium ions. Hexavalent chromium was quantifying by a Spectrophotometric method with Diphenylcarbazide [19]. The information shown in the results section are the mean from three experiments carried out by triplicate.

III. RESULTS AND DISCUSSION

A. Effect of incubation time and pH

The optimum time and pH for Cr (VI) removal by pumkin seed husk biomass was 8 hours and pH 1.0, with 1 g/100 mL of biosorbent, and 50 mg/L of the heavy metal, at 28°C Figure 1). These results are very similar to what was reported 150 minutes with *Eucalyptus* leaf extract and for different natural biomasses [20, 21], 120 minutes for *Moringa stenopetala* seed powder [22], 24 hours for the biomass of palm leaf-derived biochar [23], 30 minutes for in natura and magnetic nanomodified hydroponic lettuce roots [24]. Changes in the cell permeability of unknown origin, could partly explain the differences founded in the incubation time, providing greater or lesser exposure of the functional groups of the cell wall of the biomass analyzed [10]. Adsorption efficiency of Cr (VI) was observe a maximum at pH 1.0 and 180 minutes with the biomass analyzed. The results showed with respect to the increase in pH resulted in decrease in the removal of the metal. It was reported an optimum pH 1.5 for adsorbents from agricultural waste material [20], a pH of 1.0 for in natura and magnetic nanomodified hydroponic lettuce roots [24]. Although other authors report an optimum pH 3.0 for adsorbents from agricultural waste material [20], pH of 2.0 for dry raw biomasses of *Dioscorea rotundata, Elaeis guineensis, Manihot esculenta, Theobroma cacao* and *Zea mays* [21], a pH value of 2.0 and 4.0, for the removal of chromium(VI) from wastewater using *M. stenopetala* seed powder and banana peel powder [22], too, a pH value of 2.0 using palm leaf-derived biochar, tea stalk biochar, and waste of *Musa acuminata* residue [23, 25, and 26]. This was due to the dominant species (CrO_4^{2-} and $Cr_2O_7^{2-}$) of Cr ions in solution, which were expected to interact more strongly with the ligands carrying positive charges [27].

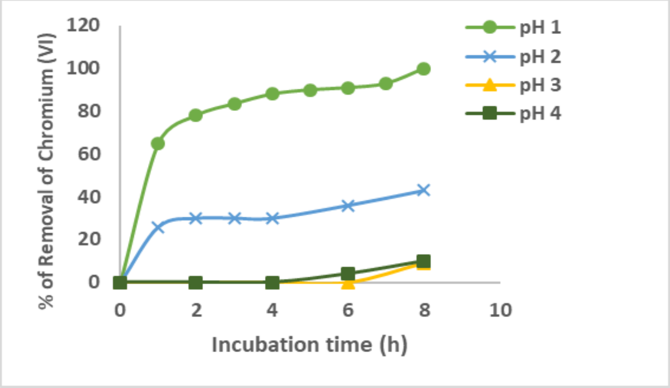

Figure 1. Effect of incubation time and pH on Chromium (VI) removal by pumpkin seed husk biomass. 50 mg/L Cr (VI), 100 rpm, 28°C. 1.0 g of biomass.

B. Effect of the temperature

On the other hand, the incubation temperature does not influence the removal of the metal, since at temperatures of 40°C, 50°C and 60°C, the removal is complete after 150 minutes, while at 28°C 90% of this contaminant is eliminated (Figure 2). To maintain constant the temperature in all experiments, we use a shaking bath Yamato BT-25 model. These results are coincident for the biomass of palm leaf-derived biochar, with the same temperature of removal [23], by a reusable chitosan-modified multi-walled carbon nanotube composite [28], by dried twigs of *Melaleuca diosmifolia* [29], for the removal of Cr (VI). But, they are different for removal of chromium (VI) from wastewater using *M. stenopeta* seed powder and banana peel powder, if increase this parameter, decrease the removal capacity of this biomasses [22], and for different natural biomasses, which exhibit higher adsorption efficiency at intermediate and low temperature values [21]. The increase in temperature increases the rate of removal of Cr (VI) and decrease the contact time required for complete removal of the metal, to increase the redox reaction rate [30].

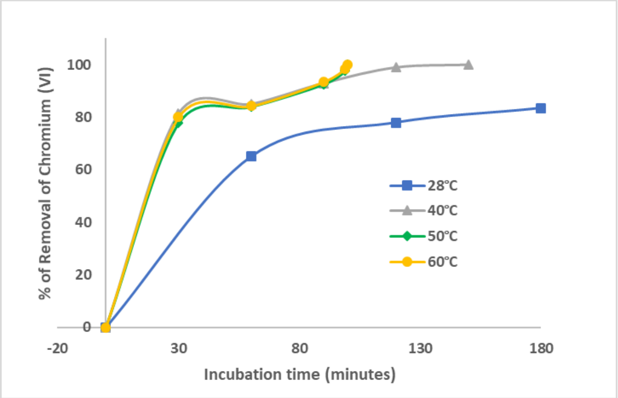

Figure 2. Effect of the temperature on Chromium (VI) removal by pumkin seed husk biomass. 50 mg/L Cr (VI), pH 1.0, 100 rpm. 1.0 g of biomass.

C. Effect of initial metal concentration

We observe that the removal of metal was 100% at 50 and 15 minutes, at 28°C, for 200 and 600 mg/L, 800 mg/L, and 1000 mg/L, respectively (Figure 3), while at 40°C, the removal is total after 65 minutes of incubation, at the chromium (VI) concentrations analyzed (Figure 4). These results are coincident for the removal of Cr (VI) by *Cucumis sativus* biomasses [31], but are different for the chromium removal using using *M. stenopetala* seed powder and banana peel powder, in which if increase the heavy metal concentration decrease the efficiency of removal and palm leaf-derived biochar [22, 23]. The increase in initial concentration of Cr (VI), results in the increased uptake capacity and decreased in the percentage of removal of the metal. This was due to the increase in the number of ions competing for the available functional groups on the surface of biomass [27, 28].

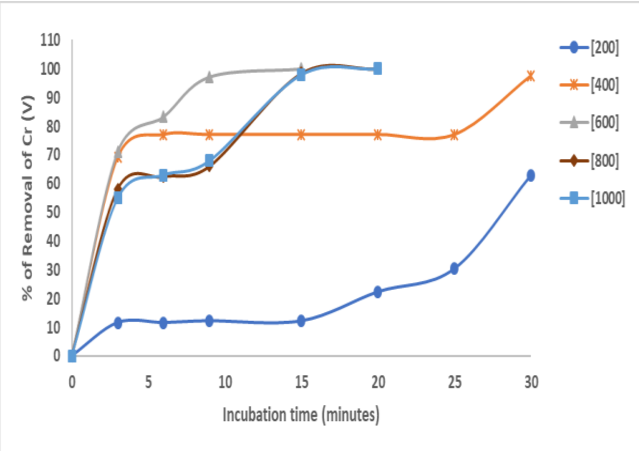

Figure 3.- Effect of initial metal concentration on Chromium (VI) removal by pumkin seed husk biomass. pH 1.0, 28°C. 100 rpm.

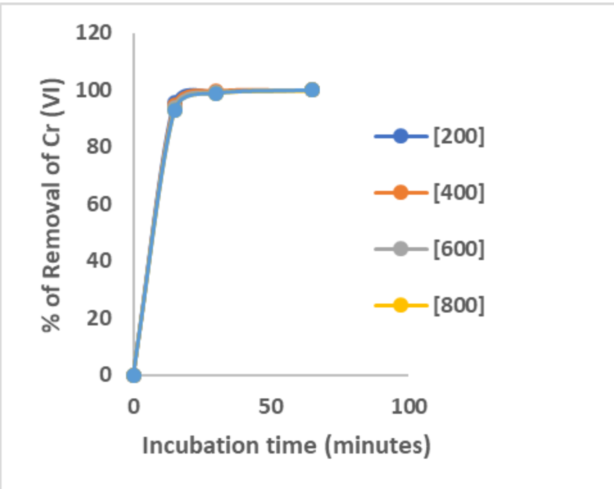

Figure 4. Effect of initial metal concentration on Chromium (VI) removal by pumkin seed husk biomass. pH 1.0, 100 rpm, 40°C.

D. Effect of biosorbent dose

The influence of biomass concentration on the removal capacity of Cr (VI) is depict in Figure 5. If we increase the amount of biomass, the removal of the metal in solution decreased significantly, because with 1 g of the analyzed biomass, 100% of the metal is removed after 480 minutes, while with 5 g, the removal is total after 25 minutes, although it has been reported what with more biosorption sites of the same, because the amount of added biosorbent determines the number of binding sites available for metal biosorption [36]. These results are similar for the removal of chromium (VI) from wastewater using *M. stenopetala* seed powder and banana peel powder, if increase the biomass concentration of 5 to 20 g/L [22], too for palm leaf-derived biochar [23], for the removal of chromium (VI) by *B. Cucumis sativus* biomasses [31]. Too, was reported a efficient removal of the metal if the biomass concentration was increased using modified Russian knapweed flower powder to initial concentrations of the heavy metal of 2, 10 and 15 mg/L with pH 2.0 [32].

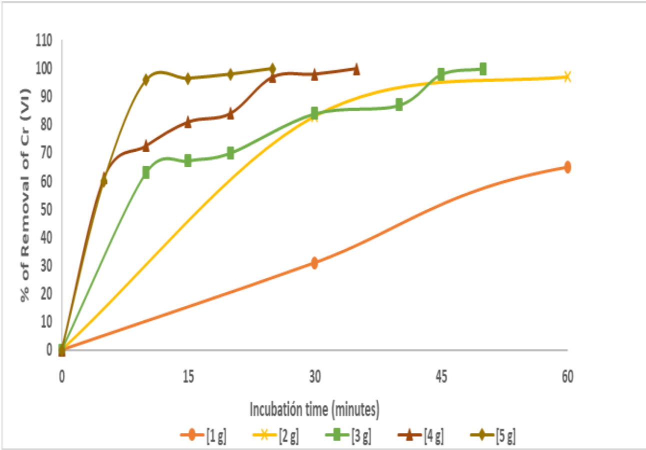

Figure 5. Effect of biomass concentration of pumkin seed husk biomass, on the removal of 50 mg/L Cr (VI), 28°C, pH 1.0, 100 rpm.

1 g, 2 g, 3 g, 4 g.

E. Desorption of Chromium (VI)

The recovery of heavy metals from metal-laden biomass has been approached by utilizing various desorption agents, including HCl, H_2SO_4, Na_2CO_3, NaOH, EDTA, and mercaptoethanol [33]. Among these approaches, decreasing and/or increasing the pH value using HCl and NaOH appears to have had the best desorption efficiency. To determine the optimal NaOH concentrations for metal desorption, the amount of metal released from the natural biomasses at different concentrations of NaOH was observed in Figure 6. Chromium (VI) can be recovered 59.3% and 61.3%, by washing twice with 0.1 M and 0,5M NaOH, respectively. and the results are similar for chromium (VI) desorption with NaOH, from agricultural waste material [20], for the biomass of palm leaf-derived biochar, with HCl treatment, and adjust pH to 7.0 with distilled water [23].

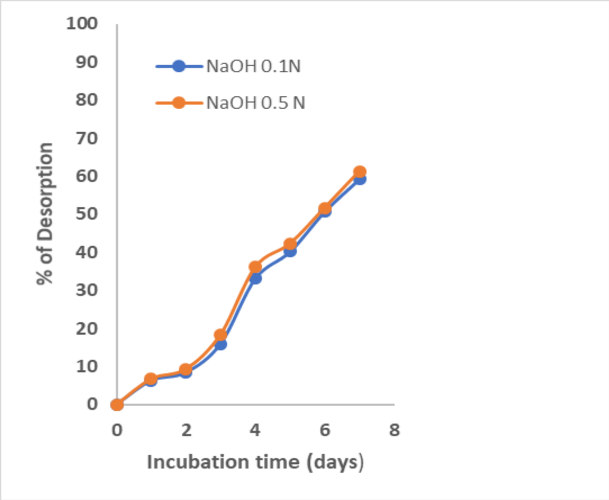

Figure 6. Effect of different concentration of NaOH on desorption of Chromium /VI) 50 mg/L of Cr (VI), 1g of fungal biomass, 100 rpm. 7 days. 28° C

F. *Removal of Cr (VI) in industrial wastes with B. vulgaris biomass.*

We adapted a water-phase bioremediation assay to explore possible usefulness of this biomass for eliminating Cr (VI) from industrial wastes. The biomass (5 g), was incubate with 10 g of non-sterilized contaminated earth with 100 mg/g, and wastewater containing 100 mg/L of Cr (VI) (adjusted), suspended in trideionized water to a final volume of 100 mL. It was observing that in 7 days of incubation, the Cr (VI) concentration of earth and water samples decrease 93% and 94.5% in both samples (Figure 7), and the decrease level occurred without change significant in total chromium content during the experiments (date not shown). In the experiment carried out without biomass, the Cr (VI) concentration of the earth samples decreased by about of 18% (date not shown); this might be caused by indigenous microflora and (or) reducing components present in the soil [34]. These results coincide with the literature reports for another natural biomass, such as for different natural biomasses [21, 35], for *Ginkgo biloba* leaves can effectively remove soil Chromium (VI) and reduce Chromium (VI) to Chromium (III) via quercetin in soil (36), removal of Chromium (VI) from wastewater using *M. stenopetala* seed powder and banana peel powder [22], for the biomass of palm leaf-derived biochar [23], the removal of chromium was found 95% from dilute tannery wastewater and 72% of chromium was extracted directly from raw tannery effluents by using different quantities of water hyacinth (37), for the phytoremediation of chromium-polluted waters in cold region (38), for waste of *M. acuminata* residue [26], and is more efficient that *Avena sativa* L. biomass, in which a lower uptake of chromium from soil in the Cr(VI)-contaminated, was observed.

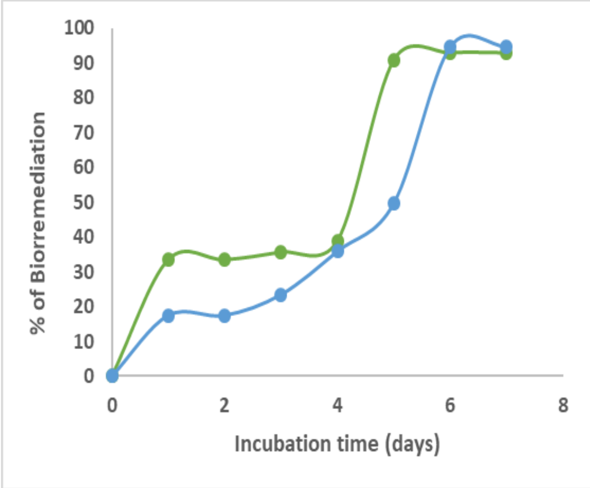

Figure 7. Removal of Cr (VI) in industrial wastes incubated with 5 g of pumkin seed husk biomass. 28°C, 100 rpm, 10 g of contaminated earth with 100 mg/g and 100 mL of contaminated water with 100 mg/L. contaminated water, (100 mg Cr (VI)/L (adjusted).
• Earth, • Water

G. Conclusions

The biomass analyzed, showed complete capacity of biosorption of 50 mg/L of Cr (VI) in solution at different time of incubation, at 28°C, 100 rpm with 1 g of natural biomass, besides this removal the metal *in situ* (7 days of incubation, with 5 g of biomass), in earth and water contaminated, respectively. These results suggest their potential applicability for the remediation of this metal from polluted soils in the fields.

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