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Notes

Chromium (VI) removal in aqueous solution by the modified biomass of rice grain (*Oriza sativa* L.)

Remoción de cromo (VI) en solución acuosa por la biomasa modificada del grano de arroz (*Oriza sativa* L.)

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Abstract

It was studied the removal capacity of Chromium (VI) in an aqueous solution by the modified biomass of rice grain, using the diphenylcarbazide colorimetric method, to evaluate the metal concentration. The biosorption at different pHs (1, 2, 3, and 4) was evaluated at different times. Too, the effect of temperature in the range of 28 to 60 °C and removal at different initial concentrations of Cr (VI) from 200 to 1 000 mg/l, were studied. The most removal of metal, was eight days, with 100 and 79 %, for both biomasses, modified and not modified, respectively, at 28 °C, pH 1.0, and 1 g of biomass. Concerning the incubation temperature, the highest removal was at 60 °C, with 100 % of removal after five days. Additionally, the biomass concentration does influence the metal removal, and efficiently removes the metal from contaminated earth and wastewater (71 and 73 %, respectively), after 10 days of incubation with 10 g of biomass.

Keywords: Chromium (VI), wastewater, rice, metals removal.

Resumen

Se estudió la capacidad de remoción de cromo (VI) en solución acuosa por la biomasa modificada del grano de arroz, utilizando el método colorimétrico de la difenilcarbazida, para evaluar la concentración del metal. Se evaluó la bioadsorción a diferentes pH´s (1, 2, 3 y 4) a diferentes tiempos. También se estudió el efecto de la temperatura en el intervalo de 28 a 60 °C y la remoción a diferentes concentraciones iniciales de Cr (VI) de 200 a 1 000 mg/l. La mayor remoción del metal fue a los ocho días, con un 100 y 79 %, para las biomasas modificada y no modificada, respectivamente, con un pH de 1.0, 28 °C y 1 g de biomasa. Con respecto a la temperatura de trabajo, la óptima fue de 60 °C, con un 100 % de remoción a los cinco días, mientras que a las concentraciones de cromo (VI) analizadas, la biomasa natural mostró una buena capacidad de remoción, eliminando 200 mg/l del metal a los 13 días y una temperatura de 28 °C. Además, a mayor concentración de la biomasa, es mayor la remoción del metal, y elimina eficientemente el metal de tierra y aguas residuales contaminadas (71 y 73 %, respectivamente), a los 10 días de incubación con 10 g de biomasa modificada.

Palabras clave: cromo (VI), aguas residuales, arroz, remoción de metales.

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Introduction

Rice is the seed of the plant *Oryza sativa*, which belongs to the family Poaceae (*graminaceae*). It is edible and constitutes the basis of almost half of the world's population's diet. It is a cereal considered a basic food in many cultures, especially in Asian cuisine, as well as for some parts of Latin America (Lema, 2018).

Until 1988, the Mexican Republic was self-sufficient in rice production, but in 2016 only 21.6% of the national consumption was produced. Currently, around 930 000 tons are imported per year (Sagarpa, 2017).

It has been reported that this is one of the three most consumed grains in the world, mainly as whole grain, and contributes in a very effective way to the daily energy requirement of the current human diet.

Rice is responsible for the caloric intake of one-fifth of the calories consumed by humans in the world (Babaso & Sharanagouda, 2017). On the other hand, rice has a low protein content (7-9% dry weight); the grain is the biggest protein source in the principal rice-consumer countries, and its protein quality is lower than oat and better than wheat and corn (Pinciroli, 2015). Its content percentage is the following: carbohydrates 64.3 (from which 80% is starch, such as amylose and amylopectin), 7.3 protein, 2.2 lipids, 1.4 ashes, and 0.8 crude fiber and vitamins (Pinciroli, 2015). Also, medicinal properties have been described, such as being hypoallergenic, anti-diarrheal, anti-inflammatory, laxative, lipid-lowering, useful for the treatment of gastritis, diarrhea, skin problems, muscular contractions, and trauma (Pinciroli, 2015; Chakraborty 2020). Even it has been reported that bran proteins have anticancer properties (Kawamura & Muramonto, 1993).

On the other hand, population growth, agriculture, and industrial development have increased the sources of environmental pollution and toxic waste. So many polluters like industrial effluents, pesticides and other chemical products used for diverse human activities reach the ground, rivers, lakes, and seas, causing irreversible damage to the environment. These damages translate into changes that affect the quality of life and health of the population, due to the disturbances induced in air, ground, and water, as well as in urban and rural environments (Kapahi & Sachdeva, 2019).

Among the main environmental pollutants, there are heavy metals, whose presence must be highly considered in food, both of animal origin and plant origin. These metals could accumulate in crops through the absorption by irrigation with polluted water, through the roots, and by deposition of airborne particles in the foliage (Sarabia-Meléndez, Berber-Mendoza, Reyes-Cárdenas, Sarabia-Meléndez, & Acosta-Rangel, 2018). Among these metals, chrome is precisely one of the most toxic and one of the hardest removals.

Currently, there is much research about the use of a wide variety of residual lignocellulosic biomasses for the removal of metal ions from polluted waters, which include tree barks, residues of wood, seeds, and leaves from different trees, dried fruit shells, residues of cereals, and citrus fruits. The residues of rice have been widely used for the elimination of many heavy metals, sometimes naturally and other times pre-treated chemically to greatly increase their removal capacity (Quiñones, Tejada, Arcia, & Ruiz, 2013; Llanos-Páez, Ríos-Navarro, Jaramillo-Páez, & Rodríguez- Herrera, 2016). For example, we have the removal of phenol by activated carbon of rice husks treated with sodium hydroxide (Khan *et*

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al., 2017); the percentage of elimination of chromium (III) from tannery effluents using modified silica obtained from rice husks was 70% for the compound SRH-NH_2, and 90 % for the compound SRH-triamine (Gutiérrez-Valtierra *et al.*, 2019); the absorption of methylene blue with rice husk modified with phosphoric acid (Rodríguez, Campos-Rosario, & Pérez-Flores, 2019); the bioadsorption of cadmium (II) by chemically treated rice husk (Hoyos-Sánchez, Córdoba-Pacheco, Rodríguez-Herrera, & Uribe-Kaffure, 2017); the removal of chromium by rice husk treated with formaldehyde (Tariq *et al.*, 2019); the elimination of lead and cadmium by the same biomass modified with nanoparticles of silver (Abbas, Nady, Abd-El-Rahman, & Ali, 2018), and the adsorption of lead by activated carbon of rice husk treated with sodium carbonate (Hanum, Bani, & Izdiharo, 2017).

The great capacity of adsorption of rice husk is attributed to the nature of its components, mainly cellulose, hemicellulose, lignin, and some proteins (Chuah, Jumasiah, Azni, Katayon, & Thomas-Choong, 2005), and to the presence of high contents of lignin (Bansal, Garg, Singh, & Garg, 2009). The objective of this work is to assess the capacity of adsorption of rice grains modified with sodium hydroxide (seed, caryopsis) for the removal of chromium (VI) in solution.

Materials and methods

Bioadsorbent

We obtained biomaterial from commercial rice grain from different supermarkets in San Luis Potosí, S. L. P., Mexico. For the obtention of the biomass, we washed the grain for 24 hours with EDTA at 10% (w/v in triple deionized water), and after 72 hours with triple deionized water stirring constantly, changing the water every 12 hours. Subsequently, it was in contact with sodium hydroxide 1 N for 24 hours (this treatment causes changes in the structure of the grain, improving the properties of accessibility of the biomass); it was rinsed three times with the same water, and then dried at 37 °C for 24 hours in a bacteriological oven, ground in a blender and stored in amber-colored jars until its use.

Solutions of Cr (VI)

We worked with 100 ml of a solution with 50 mg/l of Cr (VI) concentration obtained by the dilution of a model solution of 1.0 g/l prepared in triple deionized water from K_2CrO_4. We adjusted the pH of the dilution to be analyzed with HNO_3 1 M and/or NaOH 1 M, before adding it to the biomaterial.

Removal studies

We mixed 1 g of the biomass of rice grain (previously sterilized at 15 lb and 120 °C, in Erlenmeyer flasks of 250 ml) with 100 ml of a solution of 50 mg/l of Cr (VI) (at 1.0, 2.0, 3.0, and 4.0 pH values; temperatures of 28, 40, 50, and 60 °C; and metal concentrations of 200, 400, 600, 800, and 1 000 mg/l) and we maintained a temperature of 28 °C and 100 rpm, taking at different times aliquots of 5 ml each, which were centrifugated at 3 000 rpm (5 min). To the respective supernatant, we determined its Cr (VI) concentration in solution, using the diphenylcarbazide colorimetric method (development of pink-violet coloration) (Greenberg, Clesceri, & Eaton, 1998). All the experiments were performed 3 times minimum and in duplicate.

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Trials of remediation of polluted soil and water

We mixed into Erlenmeyer flasks of 500 ml, 180 ml of triple deionized water and 10 g of biomass treated with no sterile soil, contaminated with 100 mg of chromium (VI)/g from soil (adjusted), and 100 ml of contaminated water with 100 mg of Cr (VI) (adjusted), resuspending the soil in triple deionized water at 28 °C and 100 rpm. For the other experiment, we added to the mixture 190 ml of water contaminated with 100 mg/l of Cr (VI), obtained from a rinse tank from a chrome plant in Celaya, Gto., Mexico, and we maintained them at 28 °C, stirring constantly (100 rpm); and at different intervals of time, we determined the Cr (VI) concentration in the supernatant. All the experiments were performed 3 times and in duplicate.

Results and discussion

We analyzed the adsorption of 50 mg/l of chromium (VI), at different times of contact (0-10 days), and different values of pH (1, 2, 3, and 4), adjusting the experiments to a unifactorial fixed-effects model. We observed that pH values of 1.0, 2.0, 3.0, and 4.0 it is removed 100, 92.3, 76.3, and 56.2 % of the metal, respectively, at day eight with the modified biomass; while with the non-modified biomass it is eliminated 79, 50.3, 34.4, and 20 % in the same time of contact, at 28 °C and pH values of 1.0, 2.0, 3.0 and 4.0, respectively. Figure 2 shows the other data of adsorption with different pH values, with 1.0 g of biomass (Figure 1 and Figure 2), which indicates a higher removal of the metal with the modified biomass. It has been reported a maximum efficiency of removal of 95% for 3 hours, for the removal of chromium by the rice husk treated with formaldehyde (Tariq *et al*., 2019); removal of Cr (VI) higher than 94% at pH of 1.0, at 12 hours of incubation with rice husk (Doria-Herrera, Hormaza-Anaguano, & Gallego-Suarez, 2011); the removal of Cr (VI) by rice residues at pH of 1.5 and 2.0, at 5 hours of contact with controlled temperature (Kumar-Naiya, Singha, & Kumar-Das, 2011); removal efficiencies of 49.2, 54.5 and 72.8 %, from industrial effluents for the rice husk ash biomasses, rice husk activated with H_3PO_4 and rice husk ashes activated with NaOH, respectively, with a pH range between 0 and 5.0, at 24 hours of development (Rodríguez, Salinas, Ríos, & Vargas, 2012); percentages of remotion of 71% and 76.5% for rice husk treated with heat and formaldehyde, respectively, at a pH value of 2.0 and 3 hours of development (Bansal *et al.*, 2009); elimination of 99 % of chromium (III) from tannery effluents using modified silica obtained from rice husk, in a pH range between 2.3 and 4.0 in 20 minutes (Gutiérrez-Valtierra *et al*., 2019). Chromium (VI) is found as $HCrO_4^-$, $Cr_2O_7^{2-}$, CrO_4^{2-}, $Cr_4O_{13}^{2-}$, $Cr_3O_{10}^{2-}$ (Rollison, 1973).

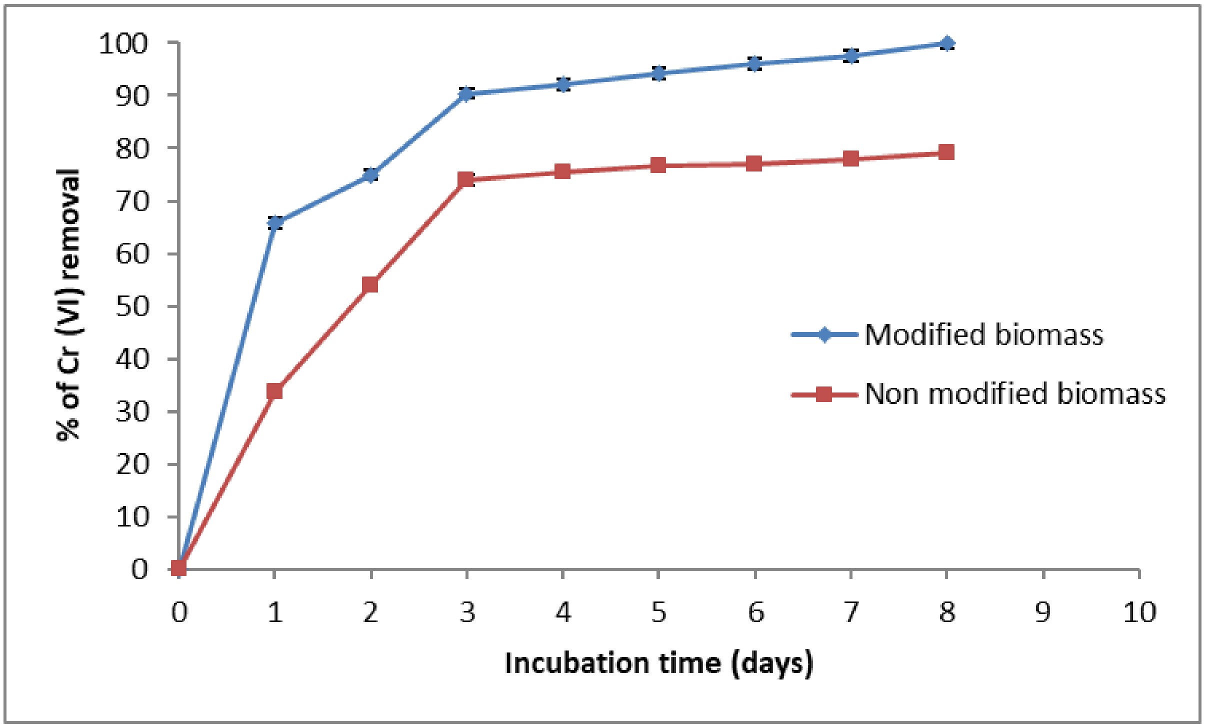

Figure 1. Effect of development time over the removal of chromium (VI) by the modified and non-modified biomass of rice grain (1 g of biomass, 50 mg/l Cr (VI), 28 °C, 100 rpm).

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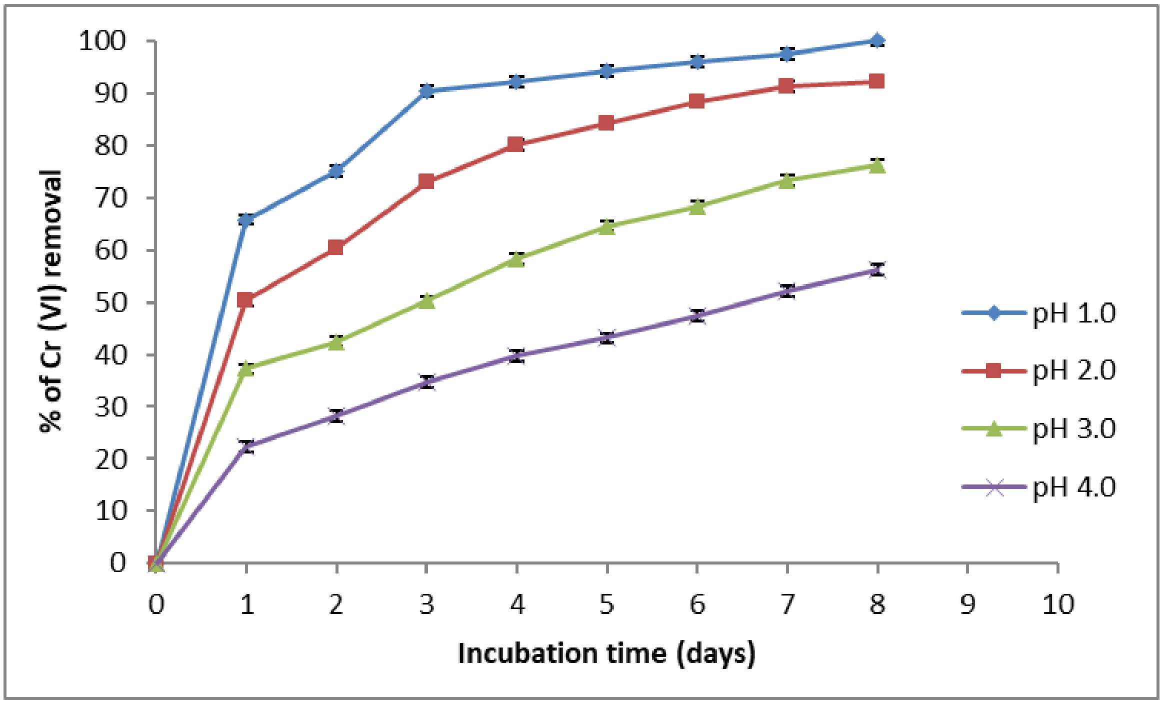

Figure 2. Effect of development time and the initial pH over the removal of chromium (VI) by the modified biomass of rice grain (1 g of biomass, 50 mg/l Cr (VI), 28 °C, 100 rpm).

A drop in pH causes the protonation of the adsorbent's surface, which induces a strong attraction to Cr (VI) ions of the solution charged negatively, so the adsorption increases as the acidity of the solution increase. Nevertheless, when the pH increases, the concentration of OH- ions increases, inducing changes on the adsorbent's surface, impeding the absorption of the chromium (VI) ions charged negatively, which decreases the absorption of the metal at these pH values (Gutiérrez-Corona, Romo-

Rodríguez, Santos-Escobar, Espino-Saldaña, & Hernández-Escoto, 2016).

Also, we observed that at higher temperatures it is higher the adsorption of the metal at 60 °C removes 100% at 5 days of development, while at 28 °C the total is removed until day eight (Figure 3). It has been reported that an increase in temperature from 25 °C to 45 °C increases the Cr (VI) removal, since it reduces the time of elimination from 91 to 21 hours, with guava seed modified with H_2SO_4 1 N (Ortiz-Gutiérrez, 2014); also, the removal of cobalt, nickel, and pyridine derivatives increases when the temperature increases, using rice husk and its ashes as adsorbent, respectively (Swelam, Awad, Salem, & El-Feky, 2016; Lin & Wang, 2011; Lataye, Mishrab, & Mall, 2009). The increase in temperature increases the velocity of Cr (VI) removal and decreases the time of contact required for the entire removal of the metal, by increasing the velocity of the redox reaction (Wittbrodt & Palmer, 1996).

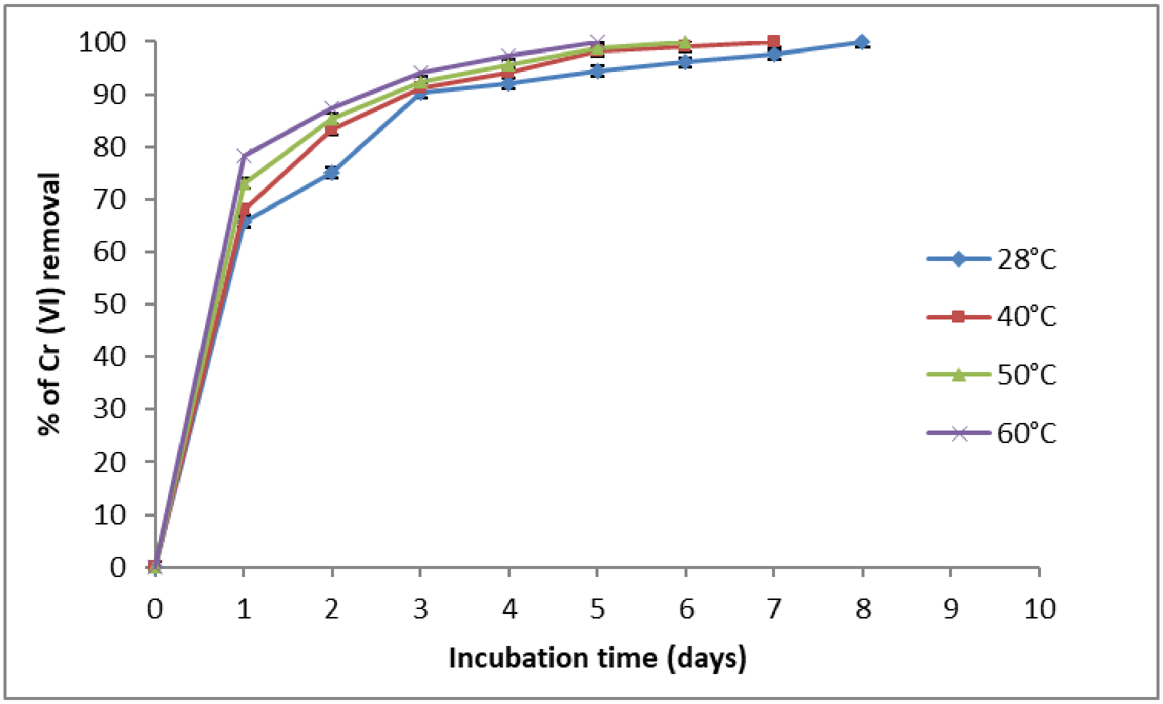

Figure 3. Effect of temperature of development over the removal of chromium (VI) by the modified biomass of rice grain (1 g of biomass, 50 mg/l Cr (VI), pH = 1.0, 100 rpm).

About the effect of different concentrations of chromium (VI) in solution, at pH values of 1.0 +/- 0.2, with 1 g of biomass, at 28 °C and 100 rpm, we observed that at higher concentrations of the metal, it is lower the removal since, at day 13 of development, they are fully removed the 200 mg/l. In this case, with 1 000 mg/l, it is removed only 55% of the metal in the solution at day 15 (Figure 4a); while at 60 °C, on day nine, 200 mg/l is removed. With 1 000 mg/l, 65.4% is removed at day 15 of development (Figure 4b). These results are similar to those reported for the removal of 10 to 70 mg/l of Cr (VI) by rice husk treated with heat and formaldehyde (Bansal *et al.*, 2009); for a high concentration of the metal (3 mg/l) the percentage of removal is very low (22 %) at 180 minutes with rice husk (Doria- Herrera *et al*., 2011). On the other hand, some reports (Batagarawa & Ajibola, 2019) indicate that the quantity of nickel, chromium, and manganese eliminated by some biomasses from different sources, such as crude rice, carbonized rice, millet, and maize leaves as low-cost adsorbents, increases in direct proportion with the increase of the concentration of the metal in solution. Also, if the concentration of cobalt (II) increases from 117.86 to 825.05 mg/l, the removal of this metal with rice husk increases (Swelam *et al*., 2016); likely, it occurs for the removal of 100 to 500 mg/l of nickel (II), zinc (II), and lead (II) at 120 minutes, by activated carbon of rice husk (Taha, Shuib, Shaharun, & Borhan, 2014); it happens the same when using rice husk ashes for the removal of nickel (Lin & Wang, 2011), and for the elimination of derivates of pyridine, using rice husk and its ashes as adsorbent (Lataye *et al*., 2009).

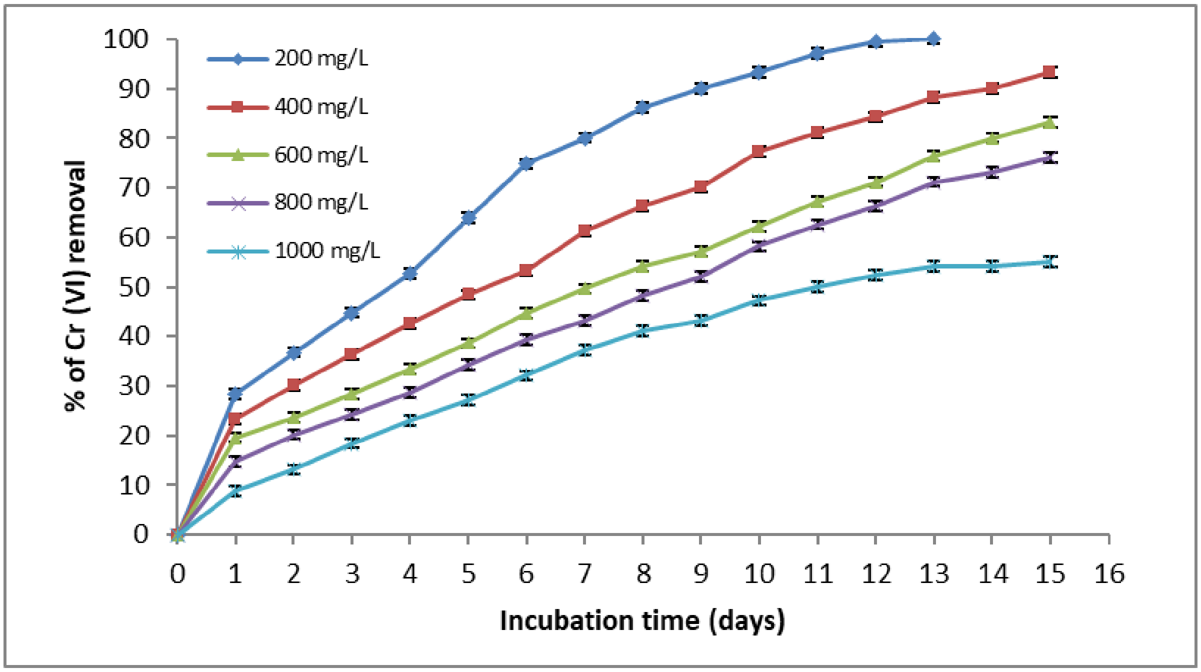

Figure 4a. Effect of the concentration of chromium (VI) over the removal of the metal (1 g of biomass, 28 °C, pH = 1.0, 100 rpm).

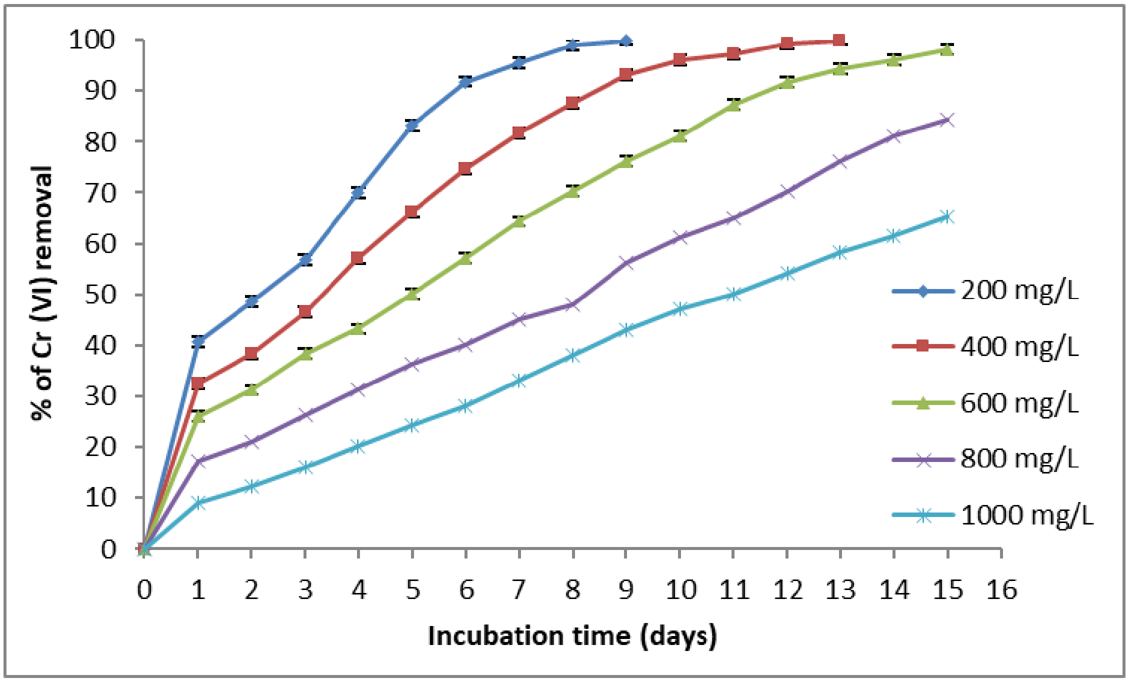

Figure 4b. Effect of the concentration of chromium (VI) over the removal of the metal (1 g of biomass, 60 °C, pH = 1.0, 100 rpm).

On the other hand, at higher concentrations of the biomass, there is higher removal of the metal in solution (Figure 5), since there are more adsorption sites of the metal; this is because the quantity of added adsorbent determines the number of binding sites available for the adsorption of the metal (Cervantes *et al*., 2001). Similar results have been reported for the removal of Cr (VI) with the biomass of rice husk treated with heat and formaldehyde (Bansal *et al*., 2009); rice husk (Doria-Herrera *et al*., 2011), for the elimination of chromium (III) from tannery effluents using modified silica obtained from rice husk (Gutiérrez-

Valtierra *et al*., 2019); for the removal of nickel, chromium, and manganese, by biomasses like crude rice, carbonized rice, millet, and maize leaves (Batagarawa & Ajibola, 2019); for the removal of cobalt (II) with rice husk (Swelam *et al*., 2016); and the elimination of pyridine derivatives using rice husk and its ashes as adsorbent (Lataye *et al*., 2009), but these results are different to those reported for the removal of nickel using rice husk ash, where it is shown that at higher concentration of the biomass, the removal is lower (Lin & Wang, 2011).

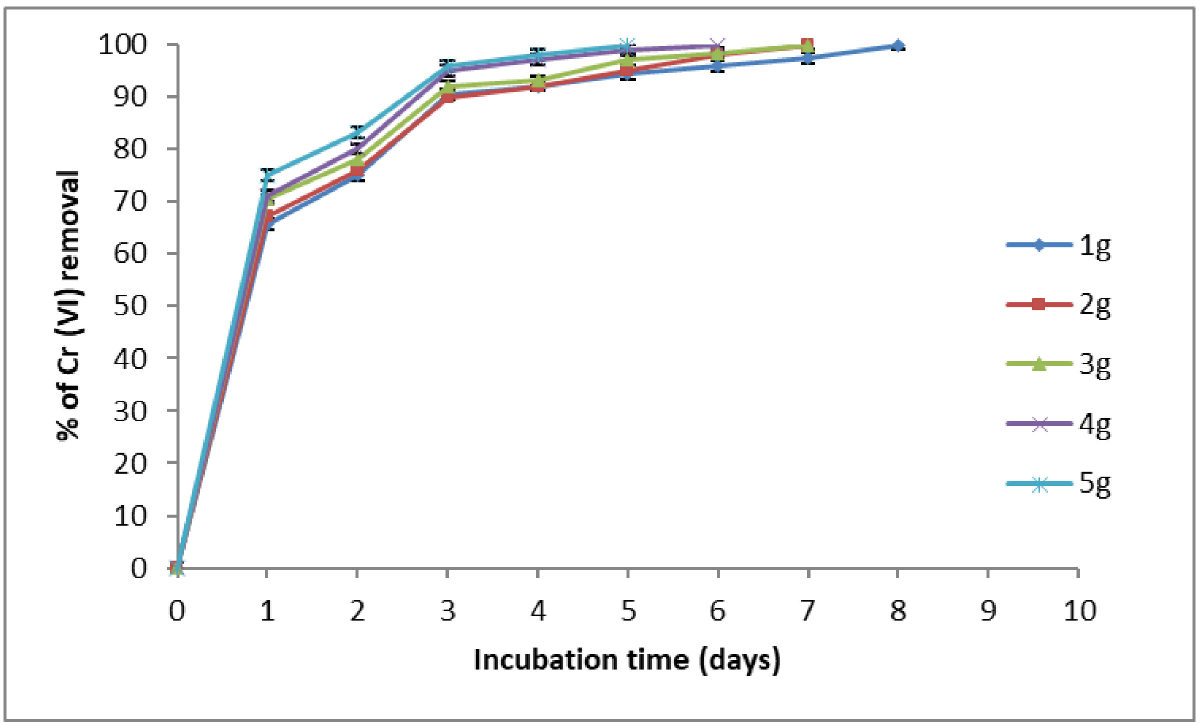

Figure 5. Effect of different concentrations of the biomass over the removal of chromium (VI) (28 °C, pH = 1.0, 100 rpm).

However, the reports about applications of natural biomasses and microorganisms for studies of remediation of polluted waters and soils with chromium (VI) are scarce. For example, it has been reported the elimination of copper, nickel, and chromium (VI) in polluted waters, using *Moringa oleífera* seeds as adsorbents (Landázuri, Cahuasquí, & Lagos, 2019). Also, there are reports about the removal of chromium (VI) from wastewater using residues of coffee pulp (Gómez-Aguilar, Rodríguez-Miranda, Esteban-Muñoz, & Betancur, 2019), ground coffee (Krishna-Mohjan, Naga-Babu, Kalpana, & Ravindhranath, 2019), and corn straw (Ma *et al*., 2019); and the removal of phenol and chromium (VI) with materials derived from algae biomass (Cui, Masud, Aich, & Atkinson, 2019). Therefore, to study the possible use of the rice husk biomass to eliminate chromium (VI) from industrial waste, we adapted a trial of remediation in aqueous solution, incubating 10 g of biomass treated with no sterile soil, contaminated with 100 mg of chromium (VI)/g of soil (adjusted), and 100 ml of water contaminated with 100 mg of Cr (VI) (adjusted), resuspending the soil in triple deionized water at 28 °C and 100 rpm, observing that after 10 days of experimentation, it is removed 71 and 73 % of the metal from the samples of contaminated soil and water, showing no significant changes in the total content of chromium (Figure 6), which agrees with the literature about different biomasses and microorganisms such as the removal of chromium (VI) from synthetic wastewater under different experimental conditions by using the biomass of rice husk treated with heat and formaldehyde (Bansal *et al*., 2009); the same metal from tannery wastewater simulated at laboratory conditions with rice husk (Doria-Herrera *et al*., 2011); removal of 70% of chromium (III) from tannery wastes from León, Guanajuato, México, with modified silica with amino and polyamino groups, obtained from rice husk (Gutiérrez-Valtierra *et al*., 2019); the removal of chromium, lead, and nickel from wastewater with rice husk treated with formaldehyde (Tariq *et al*., 2019); the removal of lead from wastewater with rice husk (Sovattei, Bacani, Promentilla, Hinode, & Seingheng, 2013); the significative decrease of the concentration of heavy metals and arsenic from water from formal minning at Cerro de Pasco, Sierra Central, Lima, Peru, using soap with no excipients and rice husk (Alcántara, 2020); and the elimination of lead from car accumulators with activated carbon of rice husk treated with Na_2CO_3 (Hanum *et al.*, 2017).

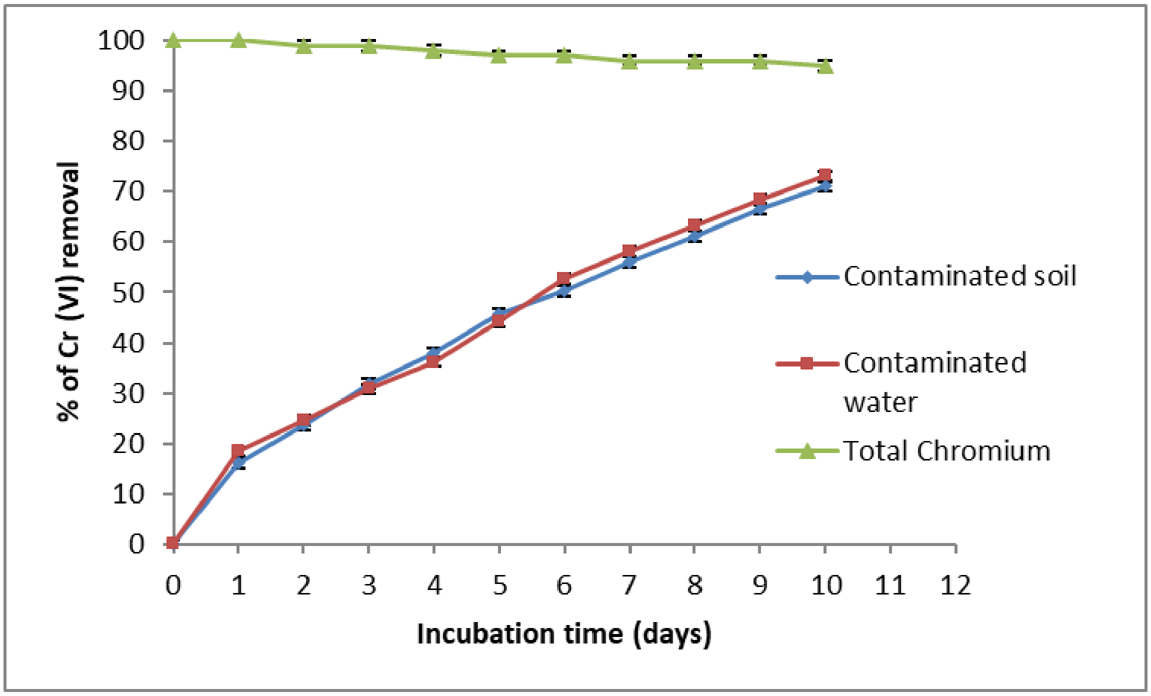

Figure 6. Remediation of 100 mg of chromium (VI)/g of contaminated soil and 100 mg/l water, by the modified biomass of rice husk (10 g of biomass, 10 g of soil, 28 °C, 100 rpm).

Conclusions

The rice husk biomass showed an excellent capacity of adsorption of 50 mg/l of Cr (VI) in solution, after eight days of development, at 28 °C, 100 rpm, and 1 g of biomass modified with sodium hydroxide 1 N; also, the metal can be efficiently removed in situ (71 and 73 % of removal, with 10 days of experimentation, 10 g of biomass, in soil and water contaminated with this metal). These results suggest the potential applicability of this biomass for the remediation of polluted places with Cr (VI).

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