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QUINCE *(CYDONIA OBLONGA)* PEEL AS A NATURAL BIOSORBENT FOR CHROMIUM (VI) REMOVAL IN BATCH SYSTEMS

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ABSTRACT

This study investigates the potential of quince (Cydonia oblonga) peels as a natural biosorbent for removing hexavalent chromium (Cr(VI)), a highly toxic heavy metal, from aqueous solutions. Experiments were conducted in batch systems to evaluate both the reduction of Cr(VI) to Cr(III) and its biosorption onto the quince peels. Results demonstrated that quince peels effectively facilitated both processes, with reduction efficiency maximized at 40°C due to increased molecular kinetic energy. Lower initial Cr(VI) concentrations resulted in higher reduction and removal efficiencies, attributed to greater availability of active binding sites on the biosorbent. Acidic conditions (pH 2) significantly enhanced both reduction and biosorption, likely due to protonation of the active sites on the peels, achieving a maximum biosorption capacity of 1.5 mg/g. The study further explored the influence of biosorbent dosage, revealing high reduction efficiency across various doses, with slightly higher

efficiency observed at low to intermediate doses. These findings highlight the potential of quince peels, an abundant agricultural waste product, as a cost-effective and eco-friendly alternative for Cr(VI) remediation, offering dual functionality in both reducing and removing this hazardous pollutant from water.

Keywords: Biosorption, Quince, Chromium, Reduction.

1. INTRODUCTION

The problem of water pollution by heavy metals represents a global environmental and public health concern. Heavy metals such as lead, cadmium, mercury, and metalloids like arsenic are non-biodegradable and can accumulate in water bodies due to various anthropogenic activities, including industrial discharges, mining, and agricultural runoff. These metals are toxic even at low concentrations and can cause severe health problems, including neurological disorders, organ damage, and cancer. Heavy metals' persistence and bio accumulative nature make them particularly hazardous, as they can enter the food chain and magnify through trophic levels, affecting aquatic life and humans. Industrial processes, mining, and agricultural practices are primary sources of heavy metal pollution. Effluents from these activities often contain high concentrations of metals like lead, cadmium, and mercury, which are discharged into water bodies [1]. Natural phenomena such as volcanic eruptions and weathering of rocks can also contribute to heavy metal presence in water, although the impact is generally less significant compared to human activities [2].

Among these pollutants, hexavalent chromium (Cr(VI)) stands out for its high toxicity, carcinogenicity, and ability to cause genetic mutations [3]. This metal is commonly found in industrial effluents from activities such as electroplating, leather tanning, and pigment manufacturing, and its presence in water bodies poses serious risks to ecosystems and human communities. Exposure to chromium VI (Cr(VI)) is linked to a heightened risk of several health issues, including cardiovascular disease, respiratory problems, and skin irritation. The United States Environmental Protection Agency (USEPA) has classified Cr(VI) as a human carcinogen. Industrial workers and communities near industrial areas are especially vulnerable to exposure to this harmful substance [4, 5]. Thus, developing and implementing effective remediation strategies to mitigate its impact is essential.

Conventional methods for removing heavy metals, such as chemical precipitation, ion exchange, and chemical reduction, have demonstrated significant limitations. These include high operating costs, low efficiency in treating large volumes of water with low metal concentrations, and the generation of toxic byproducts that require additional treatment. In this context, biosorption emerges as an attractive alternative, taking advantage of the ability of certain biological materials to adsorb metals through mechanisms such as ion exchange, physical adsorption, and chemical complexation [6]. This process uses biological materials like agricultural waste, fungi, algae, and other biomass, which are readily available and cost-effective. This makes them suitable for large-scale applications in removing heavy metals from

water. The advantages of biosorption over traditional methods are numerous, making it an attractive option for addressing the challenges posed by Cr(VI) contamination [6]. Biosorption is an eco-friendly process that minimizes harmful chemicals and energy-intensive procedures typically associated with conventional methods like chemical precipitation and ion exchange. The process is sustainable, using biodegradable natural materials and not introducing secondary pollutants into the environment [7]. Biosorbents have shown high efficiency in removing Cr(VI) from aqueous solutions, with removal rates often exceeding 90% under optimal conditions. For instance, Aspergillus flavus achieved a removal efficiency of 99.96% at a specific pH level [8]. Also, acid-modified guava seeds showed high efficiencies for Cr(VI) reduction and removal [9].

Quince (*Cydonia oblonga*) peel may offer several advantages for the removal of hexavalent chromium (Cr(VI)) from contaminated aquatic environments, primarily due to its rich polyphenolic content [10]. These compounds, particularly flavonoids and phenolic acids, have been shown to possess significant antioxidant properties, which can facilitate the reduction of Cr(VI) to the less toxic Cr(III) form. This process detoxifies the chromium and reduces its mobility and bioavailability in the environment. The polyphenolic profile of quince includes hydroxycinnamic acids, hydroxybenzoic acids, and flavonoids, which are effective in binding and reducing heavy metals [10]. The presence of these compounds in quince extracts may enhance the reduction of Cr(VI) through complexation and redox reactions, as well as its removal from contaminated waters. Quince (*Cydonia oblonga*) plays a multifaceted role in ecosystems and biodiversity worldwide, primarily through its nutritional and medicinal properties, its role in agriculture, and its historical and cultural significance. As a fruit with a rich composition of bioactive compounds, quince contributes to both human health and agricultural biodiversity. Its cultivation and use have implications for sustainable agriculture and the preservation of genetic diversity. Quince is cultivated globally, with significant production in regions such as the Mediterranean, Central Asia, and parts of Europe and America. It serves as a rootstock for pear cultivars, contributing to agricultural biodiversity [11, 12]. The fruit's hardiness and adaptability to different climates make it a valuable crop for sustainable agriculture. Its cultivation supports the circular economy by utilizing by-products as functional ingredients in various industries [13]. Thus, the use of its residues can be a potential source of raw material for producing an effective and efficient biosorbent for Cr(VI) removal from water.

Based on the above, the main objective of this study is to evaluate the ability of quince peels to reduce and adsorb Cr(VI) in batch systems. Specific objectives include (1) determining the kinetic constants and equilibrium parameters of the biosorption process and (2) evaluating the influence of operational factors such as pH, biosorbent dosage, temperature, and initial Cr (VI) concentration on treatment efficiency.

2. MATERIALS AND METHODS

2.1 Biosorbent preparation

The biomass (quince peels), provided by a traditional candy factory, was washed with plenty of water to remove pulp residues and dried in an oven at 60 °C. Once dried, the seeds were separated from the peel. They were then crushed into small pieces and washed several times with distilled water to remove various

impurities. Then, they were dried in an oven at a temperature of 60 °C and then crushed to a particle size of approximately 1 mm. This material was labeled as QP and used for reduction and biosorption tests without further modification.

2.2 Chromium reduction and biosorption experiments

Experiments were conducted with QP and a $K_2Cr_2O_7$ solution at different contact times to determine the reduction kinetics of Cr (VI) to Cr(III) and chromium biosorption. This procedure was done to establish the time at which the process reached equilibrium. To carry out the above, 5 g/L of QP was placed in a beaker with 200 mL of solution, varying the initial chromium concentrations (1, 10, and 20 mg/L of $K_2Cr_2O_7$). They were left in contact at different exposure times (from 1 to 8 hours), working with the operating conditions shown in Table 1, according to a factorial design 3^2. Then, the biosorbent was separated once the contact time was over, and the supernatant was saved for later chromium analysis. Cr(VI) reduction analysis was performed on the supernatants resulting from these experiments using a UV-Vis spectrophotometer, and chromium biosorption analysis was performed using Atomic Absorption Spectroscopy (AA). Each experiment was performed in triplicate to determine the reproducibility.

Table 1: Factorial Design (3^2) for experimental analysis and its levels.

	-1	25 °C
A: Temperature	0	35 °C
	1	40 °C
	-1	2
B: pH	0	7
	1	9

In order to evaluate the influence of biosorbent dose and concentration on chromium reduction and removal, experiments were performed varying these parameters and making contacts in batch-type systems, as previously described. For the biosorbent dose, different masses of QP were put in contact with chromium solutions at a single concentration, keeping constant the pH at 7, particle size < 1 mm, Cr (VI) concentration at 10 mg/L, temperature (T) at 25 °C and equilibrium time of 60 minutes; and for the concentration, a concentration range between 1 to 300 mg/L of $K_2Cr_2O_7$ was considered at a biosorbent dose of 5 g/L and T = 25 °C. In these experiments, the experimental conditions of the factorial design were also considered (Table 1).

3. RESULTS AND DISCUSSION

3.1 Chromium reduction kinetics

Figure 1 shows the reduction kinetics of Cr(VI) to Cr(III) using QP at different temperatures. It can be observed that at 25 °C, the reduction kinetics are slow at the beginning. In the first two hours, the Cr(VI) reduction reaches about 80% and stabilizes to around 90% after 4 hours but does not reach 100%. On the other hand, at 35 °C there is an intermediate kinetics compared to the other temperatures, since in 2 hours

the reduction reaches approximately 90%, exceeding the temperature of 25 °C; it stabilizes near 95% reduction after 4 hours but does not reach 100% either. Finally, at 40 °C, faster and more efficient kinetics is observed, since in only 1 hour, it reaches 90% reduction, much faster than the other temperatures. After 3 hours, a 100% reduction is achieved, indicating that this temperature is the most effective for reducing Cr(VI) using QP as a biosorbent.
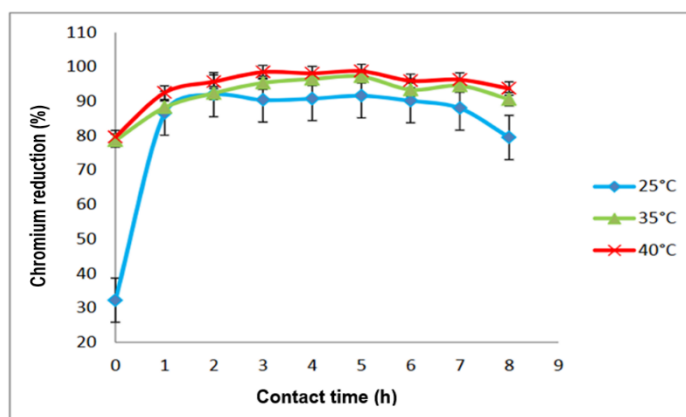

Figure 1: Reduction of Cr(VI to Cr(III) as a function of time using QP as a biosorbent at different temperatures.

Regarding the initial reduction rate, at 40°C, the process is noticeably faster, reaching 90% in the first hour, while at 35°C and 25°C, it takes 2 and 4 hours, respectively, to reach a similar percentage. This behavior reflects a clear dependence of the process's kinetics on temperature, probably due to the increase in the molecules' kinetic energy and the acceleration of the chemical reactions involved. The temperature of 40°C is the only one that achieves a 100% reduction in the time evaluated, while at 35°C, the maximum reduction is approximately 95%, and at 25°C, it is ~90%. This shows that, although all temperatures allow a significant reduction of Cr(VI), only at 40°C is a complete reduction achieved under optimal conditions. A similar behavior has been reported using Biochar for Cr(VI) reduction [14]. Moreover, At 25°C and 35°C, after 4 hours, the curves show a slight decrease in the reduction percentage. This could be due to saturation of the biosorbent's active sites or a partial reversal of the redox process. On the contrary, at 40°C, the reduction remains stable at 100%, indicating that the higher temperature guarantees both the speed and stability of the process. While higher temperatures generally enhance the kinetics and efficiency of Cr(V) reduction to Cr(II), the optimal temperature can vary depending on the specific biosorbent and its characteristics.

On the other hand, the reduction of Cr(VI) to Cr(III) is a temperature-dependent process. At higher temperatures, the greater kinetic energy of the molecules favors both the diffusion of Cr(VI) ions towards the biosorbent and the chemical reactions at the active sites. The greater efficiency at 40°C suggests the process is endothermic, i.e., favored by increasing temperature. The observed behavior at 25°C and 35°C indicates that the process is viable at lower temperatures, although at a lower rate and efficiency. Although 40°C is the optimal temperature for complete reduction, evaluating the energy costs and environmental

impact associated with heating the system is important. At ambient temperatures (~25°C), the process remains effective (~90%), which can be helpful in applications where temperature control is limited. Treatment systems should ideally operate at temperatures close to 40°C To maximize Cr(VI) removal efficiency. In applications where heating is not feasible, ambient temperatures can be used, but longer contact time should be considered. The comparative analysis reinforces the importance of temperature as a critical variable in biosorbent-based treatment processes.

Figure 2 shows the reduction kinetics of Cr(VI) using QP at different initial chromium concentrations. At a low concentration (1 mg/L), the highest reduction efficiency is presented, reaching 90% in the first 2 hours and remaining stable at ~90% for 5 hours, but after 6 hours, it begins to decrease slightly. At the end of 8 hours, the reduction efficiency drops to approximately 80%. For a medium concentration (10 mg/L), an intermediate behavior is shown in terms of kinetics and efficiency: It reaches a 90% reduction in 3 hours but begins to decline gradually after 5 hours. At the end of 8 hours, the reduction efficiency is lower than that observed with 1 mg/L, standing at around 70%. However, for a high initial chromium concentration (20 mg/L), the curve with the lowest reduction efficiency is observed in all tests since it reaches a maximum reduction (~70%) in the first 2 hours but quickly stabilizes without reaching higher values in addition, it does not show a significant decrease. However, it remains around 70% throughout the contact time.
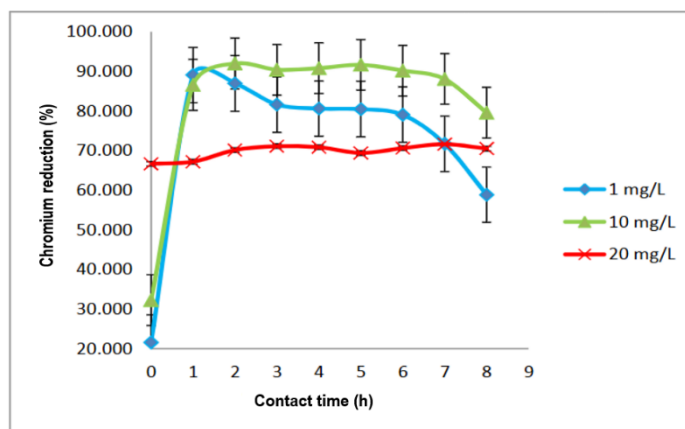

Figure 2: Reduction of Cr(VI) to Cr(III) as a function of time using QP as a biosorbent at different initial chromium concentrations.

Comparatively, the initial reduction rate is higher for lower concentrations (1 mg/L and 10 mg/L), suggesting that there is a greater availability of active sites to adsorb and reduce Cr(VI) ions under these conditions.

The initial rate is lower at 20 mg/L, probably due to the early saturation of the biosorbent's active sites [15]. Furthermore, at a lower initial concentration (1 mg/L), the highest reduction efficiency (~90%) is achieved in the first hours, while for higher concentrations (10 mg/L and 20 mg/L), the maximum efficiency is lower (~70% for 20 mg/L). This behavior indicates that the capacity of quince peel to reduce

Cr(VI) is limited by the saturation of its active sites [15]. Additionally, at 1 mg/L and 10 mg/L, after 5 hours, the curves show a decrease in reduction efficiency, which can be attributed to the saturation of the active sites and the

Competition between Cr(VI) ions and the products generated during the reaction, such as Cr(III), could inhibit efficiency. In contrast, at 20 mg/L, the curve remains constant at ~70%, suggesting that the system has already reached its capacity limit and has no additional active sites to continue reducing Cr(VI). As the initial concentration increases, the molar ratio between Cr(VI) ions and the active sites of the biosorbent decreases, explaining the lower efficiency at higher concentrations. This suggests that the system is more suitable for treating waters with low Cr(VI) concentrations. At low concentrations, the active sites are readily available for Cr(VI) ions, facilitating both adsorption and reduction to Cr(III). At higher concentrations, in addition to saturation, the accumulation of reaction products such as Cr(III) could block the active sites, reducing the overall efficiency of the process.

This analysis suggests that quince peels are more effective for treating water with low Cr(VI) concentrations. For higher concentrations, it might be necessary to Increase the amount of biosorbent, Implement a pretreatment to reduce the initial Cr(VI) concentrations or Operate in a continuous flow system where the active sites are renewed. Finally, treatment with quince peels is more efficient at low initial Cr(VI) concentrations. For practical applications, adjusting the biosorbent dosage based on the pollutant loading is critical to maximize reduction efficiency.

On the other hand, Figure 3 shows the influence of pH on the reduction of Cr(VI) to Cr(III) over the contact time, evaluating three pH values: 2, 7, and 9. It can be observed that at a pH of 2, the reduction starts at an initial value of around 30% and quickly reaches almost 100% in the first hour, remaining stable until the end of the experiment (8 hours). This curve indicates that the reduction process is extremely efficient and rapid under acidic conditions, which may be due to the greater availability of protons (H^+), which favor the reduction of Cr(VI) to Cr(III) [16]. The subsequent stability suggests that there is no reoxidation or significant loss of the reduced product. For a neutral pH (pH 7), the reduction percentage starts at around 30% and reaches approximately 90% after the first hour, stabilizing at this value until the end of the experiment. At this pH, although the reduction is high, it does not become complete as at pH 2. This could be due to a lower availability of protons and the equilibrium of the Cr(VI) species, which is less favorable for reduction at this pH. A slight decrease is observed towards the end of the experiment, possibly due to secondary reactions or reoxidation processes. On the other hand, under alkaline conditions (pH 9), the initial reduction is 30%, similar to the other curves, but it reaches a maximum of 80% in the first hour, followed by a progressive decrease over time, reaching around 60% at the end. Under alkaline conditions, the reduction process is less efficient and less stable. The low availability of protons (H^+) and the possible formation of more stable Cr(VI) species under these conditions (such as CrO_4^{2-}) make the conversion to Cr(III) difficult [17]. The constant decrease in the reduction percentage suggests reoxidation or loss of stability of the Cr(III) produced. While acidic conditions are ideal, their application must consider factors such as material corrosion and safety. Neutral systems could be a less aggressive
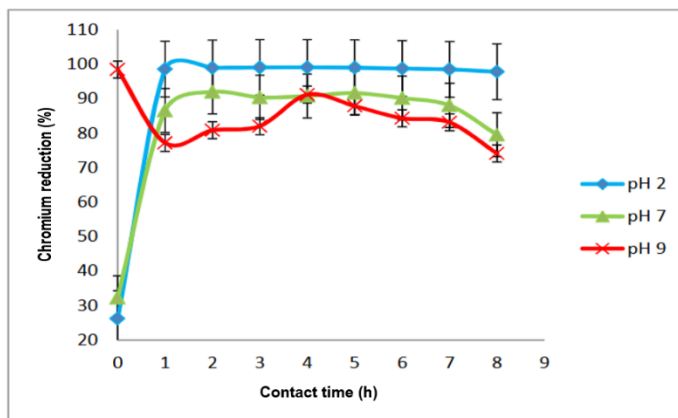
alternative, although with slightly lower efficiency.

Figure 3: Reduction of Cr(VI to Cr(III) as a function of time using QP as a biosorbent at different pH values.

3.2 Effect of biosorbent dosage on Cr(V) reduction

Figure 4 shows the corm reduction as a function of QP dosage. The reduction efficiency is close to 100% at low doses, indicating high efficiency in converting Cr(VI) to Cr(III). The efficiency stabilizes at high levels with an intermediate dose, while at higher doses, efficiency improves slightly but remains high. At higher doses, efficiency recovers and remains close to 95%. However, efficiency decreases slightly at higher doses (8, 10, and 12 g/L), remaining around 90%. Comparative analysis shows high efficiency at low doses (0.1 and 0.5 g/L), stabilization at intermediate doses (1 and 2 g/L), efficiency recovery at higher doses (4 to 6 g/L), and slight decrease at high doses (8, 10, and 12 g/L). In conclusion, the efficiency of Cr(VI) reduction to Cr(III) using quince peels as biosorbent is high at all doses tested. Low and intermediate doses show a slightly higher efficiency compared to the highest doses, suggesting a high effectiveness of quince peels in Cr(VI) reduction.
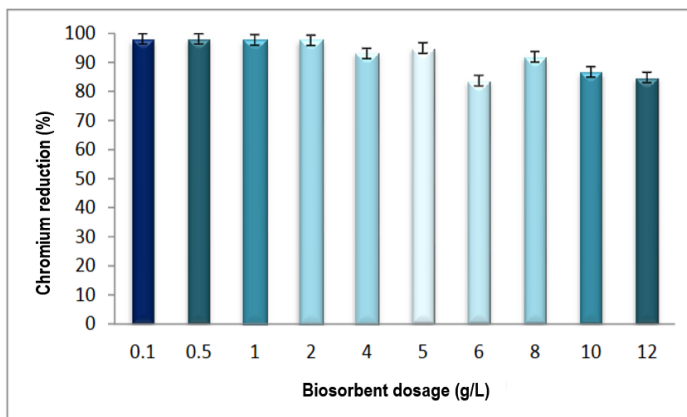

Figure 4: Reduction of Cr(VI) to Cr(III) as a function of QP dosage.

3.3   Chromium biosorption kinetics

Chromium biosorption was evaluated using QP as a biosorbent. This evaluation was performed by determining total chromium concentrations by AA in the supernatants from contact experiments between QP samples and chromium solutions. The results of chromium biosorption kinetics using QP as a biosorbent can be seen in Figure 5. At 25 °C, the curve shows a gradual increase in the percentage of biosorption over time. It starts at 0% and reaches approximately 45% after 8 hours. At this temperature, chromium biosorption is slower compared to the other temperatures. The curve shows a steady increase but does not reach the levels of biosorption observed at higher temperatures. This suggests that the lower temperature limits the ability of quince peels to adsorb chromium. At 35°C, the curve shows a faster increase in the percentage of biosorption, reaching approximately 55% after 8 hours; biosorption is more efficient than at 25°C. The curve shows a rapid increase in the first hours and then levels off. This indicates that a moderate temperature improves the adsorption capacity of quince peels, possibly due to higher molecular activity and better interaction between chromium and the peels. The red curve (40 °C) shows a similar behavior to that at 35°C, reaching approximately 50% biosorption after 8 hours. At 40°C, biosorption is slightly less efficient than at 35°C, although it is still better than at 25°C; the curve shows a rapid initial increase followed by a stabilization. The slight decrease in efficiency at 40°C compared to 35°C could be due to the denaturation of active components in quince peels at higher temperatures. The temperature of 35°C is found to be the most efficient for chromium biosorption, reaching the highest percentage of biosorption in the shortest time. The temperature of 25°C is the least efficient, while 40°C shows an intermediate efficiency. The curves at 35°C and 40°C show a faster increase in biosorption in the first hours compared to 25°C. This suggests that higher temperatures accelerate the biosorption process. Higher temperatures increase the kinetic energy of molecules, leading to more frequent and effective collisions between chromium ions and biosorbent sites, thus accelerating the biosorption process [18, 19]. All curves tend to stabilize after approximately 5 hours, indicating that an equilibrium in biosorption is reached.
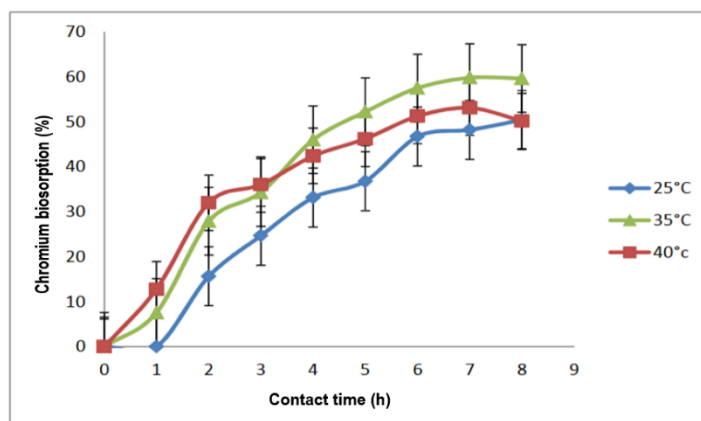

Figure 5: Chromium biosorption as a function of time using QP as biosorbent.

The biosorption kinetics of chromium presented behaviors similar to those of its reduction (described above), so the graphs of the effect of pH and initial Cr concentration are not presented.

3.4 Chromium biosorption isotherms

Figure 6 shows the total chromium biosorption isotherms using QP at different pH values: 2, 7, and 9. At pH 2, the isotherm shows a high biosorption capacity (~1.5 mg/g), with qt increasing significantly with increasing Ce. At this acidic pH, biosorption is more efficient due to the higher protonation of the active sites of quince peels, which facilitates the attraction of chromium ions. The curve for pH 7 shows an intermediate biosorption capacity, with qt reaching approximately 0.8 mg/g. At neutral pH (7), the biosorption efficiency is lower than at acidic pH due to partial neutralization of active sites, reducing electrostatic interaction with chromium ions. The curve for pH 9 indicates the lowest biosorption capacity, with qt reaching a maximum of about 0.6 mg/g. At an alkaline pH, the biosorption efficiency decreases considerably due to the deprotonation of active sites and the formation of less adsorbable chromium species. The maximum efficiency at pH 2 is observed, suggesting that acidic conditions are more favorable for chromium adsorption by quince peels. At pH 7, biosorption is moderately efficient, consistent with the theory that partial neutralization of active sites reduces adsorption capacity. At pH 9, the lowest efficiency is observed, suggesting that alkaline conditions are less favorable for chromium adsorption due to the deprotonation of active sites and the formation of less adsorbable chromium species. These results are comparable to other biosorbents, like the biomass of the Fabaceae Plant, which reached a maximum adsorption capacity for chromium of 5.3 mg/g [20].
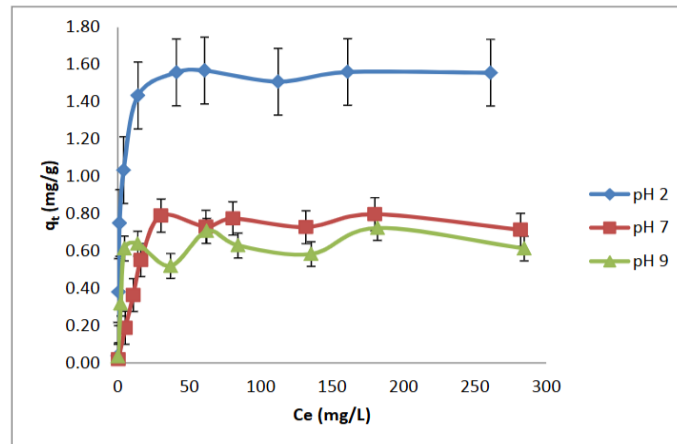

Figure 5: Chromium biosorption as a function of time using QP as biosorbent.

Moreover, the isotherm at pH 2 shows a sigmoidal curve, indicating multiple adsorption sites with different affinities for chromium [21]. This results in a rapid initial adsorption followed by a gradual saturation. The isotherm at pH 7 has an initial linear shape followed by a saturation curve. The L-type isotherm (pH 2) indicates adsorption at specific sites with maximum capacity, as observed in the saturation curve. The isotherm at pH 9 shows a linear shape with a low slope, indicating weak adsorption without saturation (type H isotherm). The sigmoidal isotherm indicates more complex adsorption with multiple sites, typical in acidic conditions where active sites are more protonated. The L-type isotherm (pH 7)

shows more specific and saturable adsorption, which is common in neutral conditions with balanced interactions between biosorbent and chromium. The H-type isotherm indicates weak and linear adsorption, typical in alkaline conditions where deprotonation of active sites reduces adsorption capacity [21].

3.5 Factorial Design Evaluation

From the results obtained from the experimental design, in which the temperature and pH parameters were varied, it can be observed that the worst scenario is obtained in the combinations with pH 2, which fall within the range of 60%, while the best percentages were obtained with pH 9 above 98% reduced. From the combination of both parameters, we have a percentage of 62.70 for pH 2 with a temperature of 35 °C and 99.18% (Table 2). The study reveals that chromium removal from quince peels is highly efficient under acidic conditions (98.34% - 99.18%), suggesting that higher temperature improves adsorption efficiency. The same results were observed under neutral pH (98.43% - 98.66%) and slightly better at 40°C (98.66%). However, under alkaline conditions (70.11% - 70.69%), the efficiency decreases due to the deprotonation of adsorption active sites and the formation of less adsorbable chromium species. The combined effect of temperature and pH is more pronounced at pH 2 and less so at pH 9. Acidic pH (2) showed the highest removal efficiency, followed by neutral pH (7) and alkaline pH 9%, indicating that protonation of active sites plays a crucial role in effective chromium adsorption. The findings can guide the optimization of operating conditions to maximize biosorption efficiency in practical applications.

Table 1: Factorial Design (3^2) analysis of results.

Experiment number	A	B	T (°C)	pH	Chromium removal (%)
1	-1	-1	25	2	98.58
2	0	-1	35	2	98.34
3	1	-1	40	2	99.18
4	-1	0	25	7	98.44
5	0	0	35	7	98.43
6	1	0	40	7	98.66
7	-1	1	25	9	70.69
8	0	1	35	9	70.11
9	1	1	40	9	70.17

4. CONCLUSIONS

This study conclusively demonstrates the potential of quince (*Cydonia oblonga*) peels as an effective and eco-friendly biosorbent for the removal and reduction of hexavalent chromium (Cr(VI)) from aqueous solutions. The biosorbent exhibited high Cr(VI) reduction efficiency, particularly at 40°C and lower initial Cr(VI) concentrations, attributed to increased reaction kinetics and greater availability of active sites. Acidic pH conditions (pH 2) significantly enhanced both reduction and biosorption processes, achieving a maximum biosorption capacity of 1.5 mg/g. The influence of biosorbent dosage revealed high reduction

efficiency across various concentrations, with optimal performance observed at low to intermediate doses. These findings suggest that quince peels, a readily available agricultural byproduct, offer a promising, sustainable, and cost-effective alternative to conventional Cr(VI) remediation methods, showcasing dual functionality in reducing the toxicity of Cr(VI) and removing it from contaminated water.

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