

## Article

# Effect of Ionic Strength and Coexisting Ions on the Biosorption of Divalent Nickel by the Acorn Shell of the Oak *Quercus crassipes* Humb. & Bonpl.

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**Abstract:** This study investigated the effect of ionic strength and background electrolytes on the biosorption of  $\text{Ni}^{2+}$  from aqueous solutions by the acorn shell of *Quercus crassipes* Humb. & Bonpl. (QCS). A NaCl ionic strength of 0.2 mM was established to have no effect on the  $\text{Ni}^{2+}$  biosorption and the biosorption capacity of the heavy metal decreased as the ionic strength increased from 2 to 2000 mM. The background electrolytes (KCl,  $\text{NaNO}_3$ ,  $\text{Na}_2\text{SO}_4$ ,  $\text{CaCl}_2$ ,  $\text{MgSO}_4$ , and  $\text{MgCl}_2$ ) had no adverse effects on the biosorption of  $\text{Ni}^{2+}$  at a concentration of 0.2 mM. However, at background electrolyte concentrations of 2 and 20 mM, divalent cations ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) had greater negative effects on the biosorption of  $\text{Ni}^{2+}$  compared to the monovalent cations ( $\text{Na}^+$  and  $\text{K}^+$ ). Additionally, the  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  anions affected the biosorption of  $\text{Ni}^{2+}$ . The fractional power, Elovich, and pseudo-second order models represented the kinetic processes of the biosorption of  $\text{Ni}^{2+}$  adequately. The results show that QCS can be a promising and low-cost biosorbent for removing  $\text{Ni}^{2+}$  ions from aqueous solutions containing various types of impurities with different concentrations.

**Keywords:** biosorbent;  $\text{Ni}^{2+}$ ; background electrolytes; kinetic modeling

## 1. Introduction

Currently, environmental pollution by toxic heavy metals is one of the most alarming problems of modern society [1–3]. Since heavy metals are non-biodegradable and highly toxic, their presence in water resources poses a great risk to the balance of the natural environment and the health of living beings [4,5].

The divalent  $\text{Ni}^{2+}$  is one of the most toxic heavy metals found in wastewater discharges owing to various anthropogenic activities, such as the manufacture of metal alloys, stainless steel, super-alloys, accumulators, batteries, electrical and electronic products and components, pigments, paints, coins, and ceramics, mineral processing, steel casting, nickel mining and refining, metallurgy, electroplating, leather tanning, and porcelain enameling [3,6,7]. Notably, it is evident that  $\text{Ni}^{2+}$  is widely used in several industrial sectors, including transportation, construction, electronics, aeronautics, automotive, and telecommunications [8].

Exposure to high levels of  $\text{Ni}^{2+}$  causes a range of harmful effects on human health, such as endocrine disorders, gastrointestinal distress, allergies, headache, anemia, dizziness, chest tightness, pulmonary fibrosis, cyanosis, rapid breathing, and encephalopathy, as well as damage to the kidneys, central nervous system, and lungs [1,4,9–11]. Moreover,  $\text{Ni}^{2+}$  exhibits carcinogenic, embryotoxic, and teratogenic properties [9,10]. Therefore, to protect the public health from the harmful effects of

$\text{Ni}^{2+}$ , the World Health Organization (WHO) established a reference value of 0.07 mg/L to control the concentration of nickel in drinking water [12].

The conventional methods used to remediate industrial wastewater contaminated with  $\text{Ni}^{2+}$ , such as chemical coagulation and precipitation, adsorption onto activated carbon, ion exchange, and various electrochemical and membrane technologies [8,13] have several disadvantages. These disadvantages include high cost, inefficient or ineffective treatment of wastewater with low  $\text{Ni}^{2+}$  concentrations, production of toxic chemical sludge that requires additional treatment, and/or they are highly sensitive to the operating parameters [9,14]. These disadvantages together with the increasing implementation of stricter environmental regulations have prompted the search for new treatment technologies [13]. Biosorption is a cost-effective, flexible, and efficient technology for removing heavy metals from aqueous solutions, which uses plant, animal, and microbial biomass or their derived products as biosorbents [15–17]. Agricultural and forestry residues and by-products, which are mainly composed of cellulose, hemicellulose, and lignin, are abundant in nature, renewable, economical, and environmental friendly. Additionally, they are highly efficient and effective for removing organic and inorganic contaminants from aqueous solutions via biosorption. Therefore, they are a viable option for bioremediation of industrial effluents contaminated with heavy metals [2,8,13,18,19].

Our previous studies established that the acorn shell of *Quercus crassipes* Humb. and Bonpl. (QCS) is a versatile and effective novel biosorbent for removing anionic and cationic heavy metals from aqueous solutions. QCS has a remarkable ability to remove hexavalent chromium (anionic heavy metal in aqueous solution) and to biosorb total chromium from aqueous solutions, both in batch [20,21] and continuous [22] systems.

Furthermore, so far, QCS is one of the best biosorbents reported for the biosorption of  $\text{Ni}^{2+}$  (cationic heavy metal) from aqueous solutions. Therefore, it was established that the QCS performance in the biosorption of  $\text{Ni}^{2+}$  ions is affected by the contact time, pH of the solution, initial  $\text{Ni}^{2+}$  concentration, and temperature. The optimal pH for the biosorption of  $\text{Ni}^{2+}$  by QCS is 8.0, whereas its point of zero charge is 5.4. The kinetic and equilibrium biosorption processes of  $\text{Ni}^{2+}$  are significantly represented using the pseudo-second order and Freundlich models, respectively. Moreover, it was established that the biosorption of  $\text{Ni}^{2+}$  by QCS is an endothermic process, non-spontaneous, and of chemical nature, in which the carboxyl, carbonyl, and hydroxyl functional groups play a major role in the removal of the heavy metal [9].

One of the critical parameters to be considered in the scaling up and large-scale application of biosorption processes is the presence of co-ions in the wastewater to be treated [20,23]. Therefore, it is important to note that most studies on biosorption of toxic heavy metals have been carried out using synthetic solutions that contain the metal of interest only. However, real industrial effluents are usually complex mixtures containing different types of background electrolytes, such as monovalent and divalent cations and anions at different concentrations [24]. The background electrolytes and their concentrations can affect the biosorption of the heavy metal of interest since they can: (1) compete with the heavy metal of interest for the available biosorption active sites, (2) decrease the specificity of the biosorbent by binding to sites to which the metal ion of interest does not bind, and/or (3) form chemical complexes or precipitates with the heavy metal of interest [20,23,24].

In spite of their great importance and relevance for the biosorption processes of toxic heavy metals, there is practically no information in the specialized literature about the effects of ionic strength and competing ions on the biosorptive removal of heavy metals from aqueous solutions [25]. This information is crucial for analyzing, interpreting, understanding and designing biosorption processes for heavy metal removal from aqueous solutions, meaning there is a clear need for investigation concerning the inhibitory effects of ionic strength and competing ions on the biosorption of the heavy metal of interest.

Therefore, the aims of the current investigation are to assess the influences of background cations, background anions, and NaCl ionic strength, on both the biosorption of  $\text{Ni}^{2+}$  ions onto QCS in aqueous solution, and the kinetic modeling of the  $\text{Ni}^{2+}$  biosorption process.

## 2. Materials and Methods

### 2.1. Biosorbent

The acorns of the oak *Quercus crassipes* Humb. & Bonpl. were collected from the town of El Durazno de Cuauhtémoc, located in the municipality of Jilotepec de Molina Enríquez, in the state of Mexico, Mexico. The acorns were washed under running water, rinsed with distilled deionized water, and then dried in an oven at 60 °C for 24 h. Thereafter, the shells were separated from the acorns and grounded in a Glen Creston® laboratory mill (Glen Creston, Ltd., London, England, UK). The resulting powder was sieved using American Society for Testing and Materials (ASTM) sieves (ASTM International, West Conshohocken, PA, USA), and the fraction with particle sizes ranging between 180–212 µm was used in all the experiments carried out in this study.

### 2.2. Stock and Test Solutions

Stock solutions of NiSO<sub>4</sub>, KCl, NaNO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub>, CaCl<sub>2</sub>, MgSO<sub>4</sub>, and MgCl<sub>2</sub> with a concentration of 20 mM and 2000 mM NaCl were prepared by dissolving a precisely weighed amount of chemical compounds in 1 L distilled deionized water. All reagents were of analytical grade (JT Baker®, Monterrey, Mexico). The test solutions were prepared by diluting the stock solutions with distilled deionized water.

### 2.3. Biosorption Experiments

The kinetic experiments were carried out using batch systems to assess the influence of ionic strength and background electrolytes on the biosorption of Ni<sup>2+</sup> from aqueous solutions by the acorn shell of *Quercus crassipes* Humb. & Bonpl. (QCS). The biosorption studies were performed in 500 mL Erlenmeyer flasks containing 110 mL Ni<sup>2+</sup> solution at an initial concentration of 1.97 mM and a QCS biomass at a concentration of 1 g/L. The flasks were shaken at a constant speed of 120 rpm in a Cole-Parmer® linear shaking water bath (Cole-Parmer®, Vernon Hills, IL, USA) for 120 h at 25 ± 1 °C. The pH of each test solution was measured regularly over the course of the experiments and then adjusted to 8.0 ± 0.1 using 0.1 M NaOH and/or HCl solutions when necessary.

Ni<sup>2+</sup> solutions containing some of the anions and cations that have been frequently found in industrial effluents were used to assess the influence of ionic strength and background electrolytes on the biosorption of Ni<sup>2+</sup> by QCS [24]. The effect of ionic strength was tested using NaCl as the background electrolyte at concentrations ranging from 0.2 to 2000 mM. To assess the effect of background electrolytes, chemical compounds (KCl, NaNO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub>, CaCl<sub>2</sub>, MgSO<sub>4</sub>, and MgCl<sub>2</sub>) consisting of monovalent and divalent anions and cations were used at concentrations of 0.2, 2, and 20 mM. Control experiments that only contained QCS biomass at a concentration of 1 g/L and Ni<sup>2+</sup> solution at an initial concentration of 1.97 mM with no background electrolytes were conducted simultaneously.

Additionally, control experiments with no QCS biomass were performed using the same operating conditions of the Ni<sup>2+</sup> biosorption experiments to determine whether there was loss of Ni<sup>2+</sup> because of precipitation or adsorption onto the glass. Statistically, there were no significant differences between the Ni<sup>2+</sup> concentrations in the control experiments with no QCS biomass at different experimental times. Thus, the decrease in the Ni<sup>2+</sup> concentration observed in the experiments with QCS biomass was caused by QCS.

Samples were taken at different experimentation times, and then they were filtered through a Whatman® grade 42 filter paper (Whatman®, St. Louis, MI, USA). The collected filtrates were analyzed to determine their residual Ni<sup>2+</sup> concentration.

The Ni<sup>2+</sup> biosorption capacity ( $q_t$ , mmol/g) at any time  $t$  was calculated using Equation (1):

$$q_t = \frac{(C_i - C_t)}{C_b} \quad (1)$$

where  $C_i$  (mmol/L) is the initial concentration of  $Ni^{2+}$  at time  $t = 0$  h,  $C_t$  (mmol/L) is the residual concentration of  $Ni^{2+}$  at time  $t = t$ , and  $C_b$  is the concentration of the QCS biosorbent (g/L).

The effect of background electrolytes and ionic strength on the biosorption of  $Ni^{2+}$  by QCS was quantitatively assessed using the global performance index of the  $Ni^{2+}$  biosorption ( $\xi$ , %), which was calculated using Equation (2) [26]:

$$\xi = 100 \frac{\left( \int_{t=0}^{t=t_f} q_t dt \right)_{\text{problem}} - \left( \int_{t=0}^{t=t_f} q_t dt \right)_{\text{control}}}{\left( \int_{t=0}^{t=t_f} q_t dt \right)_{\text{control}}} \quad (2)$$

where  $(q_t dt)_{\text{problem}}$  and  $(q_t dt)_{\text{control}}$  are the time courses of the  $Ni^{2+}$  biosorption capacity in the experiments carried out with (test experiments) and without (control experiments) background electrolytes, respectively, and  $t_f$  is the total contact time between the  $Ni^{2+}$  solution and the QCS biomass (120 h).

Integration of the  $(q_t dt)_{\text{problem}}$  and  $(q_t dt)_{\text{control}}$  functions from  $t = 0$  to  $t = t$  was performed using Mathematica version 7.0 (Wolfram Research, Champaign, IL, USA). The biosorption of  $Ni^{2+}$  by QCS is not affected by the ionic strength or background electrolytes if  $\xi = 0\%$ . There is an improvement in the biosorption of  $Ni^{2+}$  compared to the control if  $\xi > 0\%$ , thus, the ionic strength or background electrolytes have a positive or synergistic effect on the metal biosorption. Finally, if  $\xi < 0\%$ , the biosorption of  $Ni^{2+}$  is decreased compared to the control, thus, the ionic strength or background electrolytes have a negative or antagonistic effect on the metal biosorption [26].

#### 2.4. Determination of the $Ni^{2+}$ Concentration

The  $Ni^{2+}$  concentration in the liquid phase was determined using the dimethylglyoxime method [27]. The absorbance of the chemical complex with a color ranging from red wine to brown that is formed during the reaction of  $Ni^{2+}$  ions with dimethylglyoxime was measured in a Thermo Scientific™ Evolution 201 spectrophotometer (Thermo Fisher Scientific, Waltham, MA, USA) at a wavelength of 465 nm.

#### 2.5. Biosorption Kinetic Modeling

The biosorption kinetics describes the rate of biosorption of the adsorbate and, therefore, controls the time required to reach dynamic equilibrium [28]. This study used the pseudo-first order, pseudo-second order, Elovich, intraparticle diffusion, and fractional power models to analyze the kinetics of the  $Ni^{2+}$  biosorption process.

The pseudo-first order and pseudo-second order models can be described using Equations (3) and (4), respectively [29]:

$$q_t = q_{e1} (1 - e^{-k_1 t}) \quad (3)$$

$$q_t = \frac{t}{\frac{1}{k_2 q_{e2}^2} + \frac{t}{q_{e2}}} \quad (4)$$

where  $q_{e1}$  and  $q_{e2}$  are the equilibrium biosorption capacities (mmol/g) predicted by the pseudo-first order and pseudo-second order models, respectively,  $q_t$  is the biosorption capacity (mmol/g) at time  $t = t$  (h), and  $k_1$  (1/h) and  $k_2$  (g/mmol·h) are the rate constants of the pseudo-first order and pseudo-second order models, respectively.

The Elovich kinetic model is given by Equation (5) [30]:

$$q_t = \frac{1}{B_e} \ln(1 + A_e B_e t) \quad (5)$$

where  $A_e$  (mmol/g·h) and  $B_e$  (g/mmol) are the initial biosorption rate and the desorption constant, respectively.

The fractional power kinetic model is given by Equation (6) [31]:

$$q_t = k_{fp} t^v \quad (6)$$

where  $k_{fp}$  (mmol/g) is the fractional power model constant and  $v$  (1/h) is the rate constant of the fractional power model. The product of these constants ( $k_{fp} \cdot v$ , mmol/g·h) is known as the specific biosorption rate at unit time, that is, when  $t = 1$ .

The intraparticle diffusion model can be described by Equation (7) [32]:

$$q_t = k_{id} t^{0.5} + c \quad (7)$$

where  $c$  (mmol/g) is the intercept of the model that is related to the thickness of the boundary layer and  $k_{id}$  (mmol/g·h<sup>0.5</sup>) is the intraparticle diffusion rate constant.

## 2.6. Determination of the Parameters of the Kinetic Models and Statistical Analysis

All the experiments of the study were carried out at least twice and the results reported herein are the average values  $\pm$  mean standard deviation. Statistical analysis of the Ni<sup>2+</sup> biosorption data and estimation of the parameters of the tested kinetic models were performed using the GraphPad Prism version 8.0 (GraphPad Software, Inc., San Diego, CA, USA).

The analysis of variance (ANOVA) was performed using the Tukey's test for group comparisons to determine whether there were statistically significant differences in the Ni<sup>2+</sup> biosorption data. A significance level ( $\alpha$ ) of 0.05 was used. Probability values ( $p$ ) lower than  $\alpha$  indicate that the evaluated groups differ significantly.

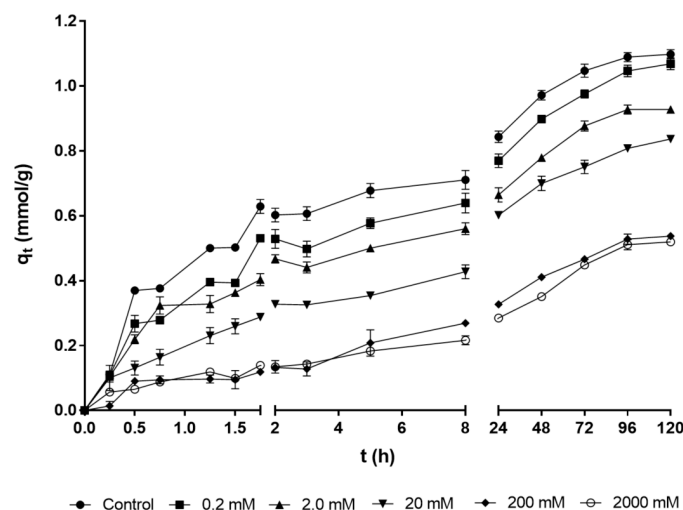
The kinetic models parameters were obtained via non-linear regression analysis of the experimental data. Various error functions, such as root mean squared error (RMSE), sum of squared error (SSE), Akaike information criterion (AIC), coefficient of determination ( $r^2$ ), and 95% confidence intervals were used to determine the accuracy and adequacy of the tested kinetic models fit. Small RMSE, SSE, and AIC values,  $r^2$  values close to one, and narrow 95% confidence intervals indicate that the models describe the experimental Ni<sup>2+</sup> biosorption data more accurately [22].

## 3. Results and Discussion

### 3.1. Influence of Ionic Strength on the Biosorption of Ni<sup>2+</sup> by Acorn Shell of *Quercus Crassipes* Humb. & Bonpl. (QCS)

The ionic strength of an aqueous solution is an environmental parameter that significantly affects the biosorption of heavy metal ions at the interface between the solid biosorbent and the liquid phase [24]. Thus, this study investigated its effect on the kinetics of biosorption of Ni<sup>2+</sup> by QCS.

Figure 1 shows the variation in Ni<sup>2+</sup> biosorption capacity as a function of the biosorption time at NaCl ionic strengths ranging from 0.2 to 2000 mM. The kinetic profile of the Ni<sup>2+</sup> biosorption at an ionic strength of 0.2 mM is similar to that of the control experiment (0 mM NaCl). There were no significant differences between the Ni<sup>2+</sup> biosorption capacities ( $p > 0.05$ ) for the control experiment and at an ionic strength of 0.2 mM at different experimental times, which indicates that the Ni<sup>2+</sup> biosorption is not affected by a NaCl ionic strength of 0.2 mM.



**Figure 1.** Effect of the NaCl ionic strength on the biosorption of  $\text{Ni}^{2+}$  by acorn shell of *Quercus crassipes* Humb. & Bonpl. (QCS).

In contrary, the  $\text{Ni}^{2+}$  biosorption capacity decreased gradually as the ionic strength increased from 0.2 to 200 mM ( $p < 0.05$ ). However, the  $\text{Ni}^{2+}$  biosorption capacities were very similar and there were no significant differences between them ( $p > 0.05$ ) at different biosorption times and at very high ionic strengths of 200 and 2000 mM. These results indicate that the biosorption of  $\text{Ni}^{2+}$  is negatively affected by NaCl ionic strengths equal to or greater than 2 mM.

The global performance indexes for the biosorption of  $\text{Ni}^{2+}$  at the different ionic strengths are summarized in Table 1.

**Table 1.** Effect of the NaCl ionic strength on the global performance index of the biosorption of  $\text{Ni}^{2+}$  by acorn shell of *Quercus crassipes* (QCS).

| Ionic Strength (mM) | $\xi$ (%) |
|---------------------|-----------|
| 0.2                 | −4        |
| 2                   | −17       |
| 20                  | −26       |
| 200                 | −55       |
| 2000                | −58       |

The global performance index revealed that a NaCl ionic strength of 0.2 mM had a negligible negative effect on the biosorption of the metal ( $\xi = -4\%$ ). However, as the ionic strength increased from 2 to 200 mM, the global performance index decreased from  $-17$  to  $-55\%$ , which indicates that the adverse effect of the ionic strength on the biosorption of  $\text{Ni}^{2+}$  increases with an increase in ionic strength ( $p < 0.05$ ). Moreover, the global performance index for the biosorption of  $\text{Ni}^{2+}$  at NaCl ionic strengths of 200 and 2000 mM were similar, thus, the adverse effects on the biosorption of the metal were similar under these conditions ( $p > 0.05$ ). These results are in agreement with the negative effects observed in the kinetic studies of the  $\text{Ni}^{2+}$  biosorption.

The decrease in  $\text{Ni}^{2+}$  biosorption with increasing NaCl ionic strengths from 0.2 to 2000 mM can be attributed to the fact that the aqueous solution contain more positively charged  $\text{Na}^+$  ions, which competed with the  $\text{Ni}^{2+}$  cations for the available biosorption active sites on the QCS surface [24,33]. Moreover, changes in the ionic strength of a solution can make the reactive functional groups on the surface of the biosorbent less accessible to  $\text{Ni}^{2+}$  ions [20,34]. Furthermore,  $\text{Na}^+$  ions can reduce the concentration of other electrostatically bound counterions that balance the negative charge of the biomass, thus,  $\text{Na}^+$  ions affect the intraparticle ion concentration and the binding of other ions, such as the  $\text{Ni}^{2+}$  ions [33]. Additionally, it has been reported that high  $\text{Cl}^-$  ion concentrations (present in NaCl)



favor the formation of the nickel chloride ( $\text{NiCl}^-$ ) complex, thus decreasing the number of free  $\text{Ni}^{2+}$  ions in the aqueous solution, which results in a lower number of interactions between  $\text{Ni}^{2+}$  ions and biosorption active sites and consequently, a decrease in the heavy metal biosorption capacity [35].

Additionally, it has been reported that the NaCl ionic strength negatively affects the biosorption of  $\text{Ni}^{2+}$  by grape stalks wastes [35], filamentous fungi such as *Rhizopus* sp., *Mucor* sp., and *Penicillium* sp. [36], and barley straw [37]. Moreover, the NaCl ionic strength decreased the sorption capacity of other heavy metals, such as  $\text{Cu}^{2+}$  by  $\text{HNO}_3$ -pretreated newspaper scraps,  $\text{HNO}_3$ -pretreated-maize spatha [38], and by an exopolysaccharide of *Wangia profunda* [39]; also  $\text{Cd}^{2+}$  by an exopolysaccharide of *Wangia profunda* [39] and by magnetic graphene oxide-supported sulfanilic acid [24],  $\text{Pb}^{2+}$  by *Sargassum filipendula* [40], and  $\text{Cr}^{6+}$  by lignin [28]. Likewise, the increase of NaCl ionic strength inhibited the biosorption of organic adsorbates, such as Methylene Blue and Rhodamine B dyes by *Phellinus igniarius* and *Fomes fomentarius* [41], and benzene and toluene by *Macrocystis pyrifera* [42].

Analysis of the kinetic model for biosorption study is crucial for understanding dynamics, mechanism, and reaction pathway of the biosorption process. It also helps in determining mass transfer, rate-controlling steps, and physicochemical interaction in the biosorption process. Additionally, knowledge about kinetics of metal biosorption is useful for determining optimum conditions for biosorption processes [43]. In the present work, the experimental kinetic data of the biosorption of  $\text{Ni}^{2+}$  by QCS at different ionic strengths were analyzed using the pseudo-first order, pseudo-second order, Elovich, fractional power, and intraparticle diffusion models. These models have been used previously to analyze and understand biosorption kinetics of  $\text{Ni}^{2+}$  ions in single metal systems by different biosorbents [7,11,26,44] but to the best knowledge of the authors, the kinetic process of  $\text{Ni}^{2+}$  biosorption from aqueous solutions containing NaCl or other background electrolytes has not been mathematically modeled. The parameters of the kinetic models are presented in Table 2.

The highest  $r^2$  values and the lowest SSE, RMSE, and AIC values were obtained using the pseudo-second order, Elovich, and fractional power models. Therefore, these models represent the kinetic profiles of the biosorption of  $\text{Ni}^{2+}$  by QCS most adequately under the studied conditions.

The equilibrium  $\text{Ni}^{2+}$  biosorption capacities obtained using the pseudo-second order model ( $q_{e2}$ ) were similar to those obtained experimentally ( $q_{e \text{ exp}}$ ). Furthermore, as the NaCl ionic strength increased from 0 to 2000 mM, the rate constant of the pseudo-second order model ( $k_2$ ) decreased from 0.166 to 0.133 g/mmol·h. A similar behavior was observed in the studies of the biosorption of total chromium by QCS at different ionic strengths [20]. A previous study that analyzed the kinetics of the biosorption of  $\text{Ni}^{2+}$  by QCS at different pH conditions, initial metal concentrations, and temperatures using the pseudo-first order and pseudo-second order models established that the pseudo-second order model described the biosorption kinetics of the heavy metal more satisfactorily than the pseudo-first order model [9]. The present study demonstrates that the pseudo-second order model can also describe the kinetics of the biosorption of  $\text{Ni}^{2+}$  by QCS at different ionic strengths.

Furthermore, it was observed that higher ionic strengths lead to higher rate constants of the fractional power model ( $v$ ) and lower initial biosorption rates ( $A_e$ ), as predicted by the Elovich model, as well as lower constants ( $k_{\text{FP}}$ ) and specific biosorption rates ( $k_{\text{FP}} \cdot v$ ) of the fractional power model. The decrease in  $k_2$ ,  $A_e$ , and  $k_{\text{FP}} \cdot v$  with increasing ionic strengths indicates that the interactions between the QCS biomass and the  $\text{Ni}^{2+}$  cations decrease with increasing NaCl ionic strengths, which prevents the binding of  $\text{Ni}^{2+}$  ions to biosorption active sites and decreases the heavy metal biosorption capacity.

Notably, even though NaCl ionic strengths equal to and greater than 200 mM significantly affected the biosorption of  $\text{Ni}^{2+}$  by QCS, this can be ignored in the present study since the ionic strength of industrial wastewater is lower than 100 mM [45].

**Table 2.** Effect of the NaCl ionic strength on the parameters of different kinetic models for the biosorption of Ni<sup>2+</sup> by QCS.

| Pseudo-First Order Model |                             |                         |                               |                |        |        |        | Pseudo-Second Order Model                  |                          |                |        |        |        |
|--------------------------|-----------------------------|-------------------------|-------------------------------|----------------|--------|--------|--------|--------------------------------------------|--------------------------|----------------|--------|--------|--------|
| Ionic Strength (mM)      | q <sub>e</sub> exp (mmol/g) | k <sub>1</sub> (1/h)    | q <sub>e1</sub> (mmol/g)      | r <sup>2</sup> | SSE    | RMSE   | AIC    | k <sub>2</sub> (g/mmol·h)                  | q <sub>e2</sub> (mmol/g) | r <sup>2</sup> | SSE    | RMSE   | AIC    |
| Control                  | 1.078                       | 0.127 ± 0.041           | 1.029 ± 0.057                 | 0.9601         | 0.0710 | 0.0769 | −65.6  | 0.166 ± 0.054                              | 1.118 ± 0.048            | 0.9872         | 0.0234 | 0.0441 | −81.1  |
| 0.2                      | 1.030                       | 0.127 ± 0.035           | 0.998 ± 0.049                 | 0.9771         | 0.0383 | 0.0619 | −60.0  | 0.158 ± 0.055                              | 1.079 ± 0.048            | 0.9892         | 0.0181 | 0.0426 | −69.0  |
| 2                        | 0.910                       | 0.102 ± 0.038           | 0.863 ± 0.061                 | 0.9422         | 0.0751 | 0.0791 | −64.8  | 0.148 ± 0.061                              | 0.953 ± 0.059            | 0.9775         | 0.0292 | 0.0494 | −78.0  |
| 20                       | 0.798                       | 0.103 ± 0.025           | 0.776 ± 0.038                 | 0.9772         | 0.0249 | 0.0476 | −72.7  | 0.142 ± 0.035                              | 0.860 ± 0.034            | 0.9916         | 0.0092 | 0.0289 | −85.6  |
| 200                      | 0.510                       | 0.044 ± 0.021           | 0.512 ± 0.059                 | 0.9229         | 0.0372 | 0.0557 | −74.6  | 0.138 ± 0.104                              | 0.565 ± 0.073            | 0.9521         | 0.0231 | 0.0439 | −81.3  |
| 2000                     | 0.491                       | 0.070 ± 0.037           | 0.467 ± 0.047                 | 0.9211         | 0.0339 | 0.0556 | −68.7  | 0.133 ± 0.084                              | 0.547 ± 0.063            | 0.9275         | 0.0183 | 0.0408 | −76.7  |
| Elovich                  |                             |                         |                               |                |        |        |        | Intraparticle Diffusion                    |                          |                |        |        |        |
|                          | A <sub>e</sub> (mmol/g·h)   | B <sub>e</sub> (g/mmol) | r <sup>2</sup>                | SSE            | RMSE   | AIC    |        | k <sub>id</sub> (mmol/g·h <sup>0.5</sup> ) | c (mmol/g)               | r <sup>2</sup> | SSE    | RMSE   | AIC    |
| Control                  | 1.885 ± 1.095               | 6.577 ± 0.707           | 0.9964                        | 0.0066         | 0.0234 | −98.85 |        | 0.090 ± 0.026                              | 0.259 ± 0.190            | 0.8227         | 0.3231 | 0.1641 | −44.37 |
| 0.2                      | 1.003 ± 0.531               | 6.216 ± 0.676           | 0.9969                        | 0.0052         | 0.0229 | −83.86 |        | 0.090 ± 0.023                              | 0.184 ± 0.173            | 0.8857         | 0.1910 | 0.1382 | −40.68 |
| 2                        | 0.671 ± 0.396               | 6.728 ± 0.892           | 0.9931                        | 0.0090         | 0.0273 | −94.56 |        | 0.078 ± 0.019                              | 0.188 ± 0.139            | 0.8679         | 0.1714 | 0.1195 | −53.24 |
| 20                       | 0.274 ± 0.072               | 6.391 ± 0.470           | 0.9972                        | 0.0031         | 0.0167 | −99.94 |        | 0.072 ± 0.014                              | 0.127 ± 0.099            | 0.9248         | 0.0824 | 0.0866 | −57.13 |
| 200                      | 0.142 ± 0.088               | 9.761 ± 1.769           | 0.9841                        | 0.0076         | 0.0253 | −96.77 |        | 0.047 ± 0.006                              | 0.061 ± 0.047            | 0.9549         | 0.0217 | 0.0426 | −82.15 |
| 2000                     | 0.071 ± 0.040               | 8.420 ± 1.729           | 0.9795                        | 0.0088         | 0.0283 | −86.19 |        | 0.046 ± 0.005                              | 0.046 ± 0.039            | 0.9697         | 0.0130 | 0.0344 | −81.10 |
| Fractional Power         |                             |                         |                               |                |        |        |        |                                            |                          |                |        |        |        |
|                          | k <sub>FP</sub> (mmol/g)    | v (1/h)                 | k <sub>FP</sub> ·v (mmol/g·h) | r <sup>2</sup> | SSE    | RMSE   | AIC    |                                            |                          |                |        |        |        |
| Control                  | 0.503 ± 0.040               | 0.168 ± 0.019           | 0.0842                        | 0.9963         | 0.0067 | 0.0237 | −98.58 |                                            |                          |                |        |        |        |
| 0.2                      | 0.427 ± 0.038               | 0.193 ± 0.021           | 0.0826                        | 0.9975         | 0.0042 | 0.0206 | −86.41 |                                            |                          |                |        |        |        |
| 2                        | 0.360 ± 0.036               | 0.202 ± 0.024           | 0.0729                        | 0.9948         | 0.0068 | 0.0238 | −98.39 |                                            |                          |                |        |        |        |
| 20                       | 0.251 ± 0.028               | 0.256 ± 0.027           | 0.0641                        | 0.9951         | 0.0054 | 0.0222 | −92.55 |                                            |                          |                |        |        |        |
| 200                      | 0.131 ± 0.020               | 0.299 ± 0.035           | 0.0390                        | 0.9926         | 0.0036 | 0.0172 | −107.5 |                                            |                          |                |        |        |        |
| 2000                     | 0.102 ± 0.022               | 0.342 ± 0.049           | 0.0350                        | 0.9890         | 0.0048 | 0.0208 | −94.22 |                                            |                          |                |        |        |        |



### 3.2. Influence of Coexisting Ionic Species on the Biosorption of $\text{Ni}^{2+}$ by Acorn Shell of *Quercus Crassipes* Humb. & Bonpl. (QCS)

Generally, there are various types of background electrolytes in industrial wastewater at varying concentrations, which can affect the biosorption of the heavy metal of interest. This study investigated the effect of coexisting ionic species on the biosorption of  $\text{Ni}^{2+}$  by QCS using different cations and anions at three different concentrations, namely 0.2, 2.0, and 20 mM.

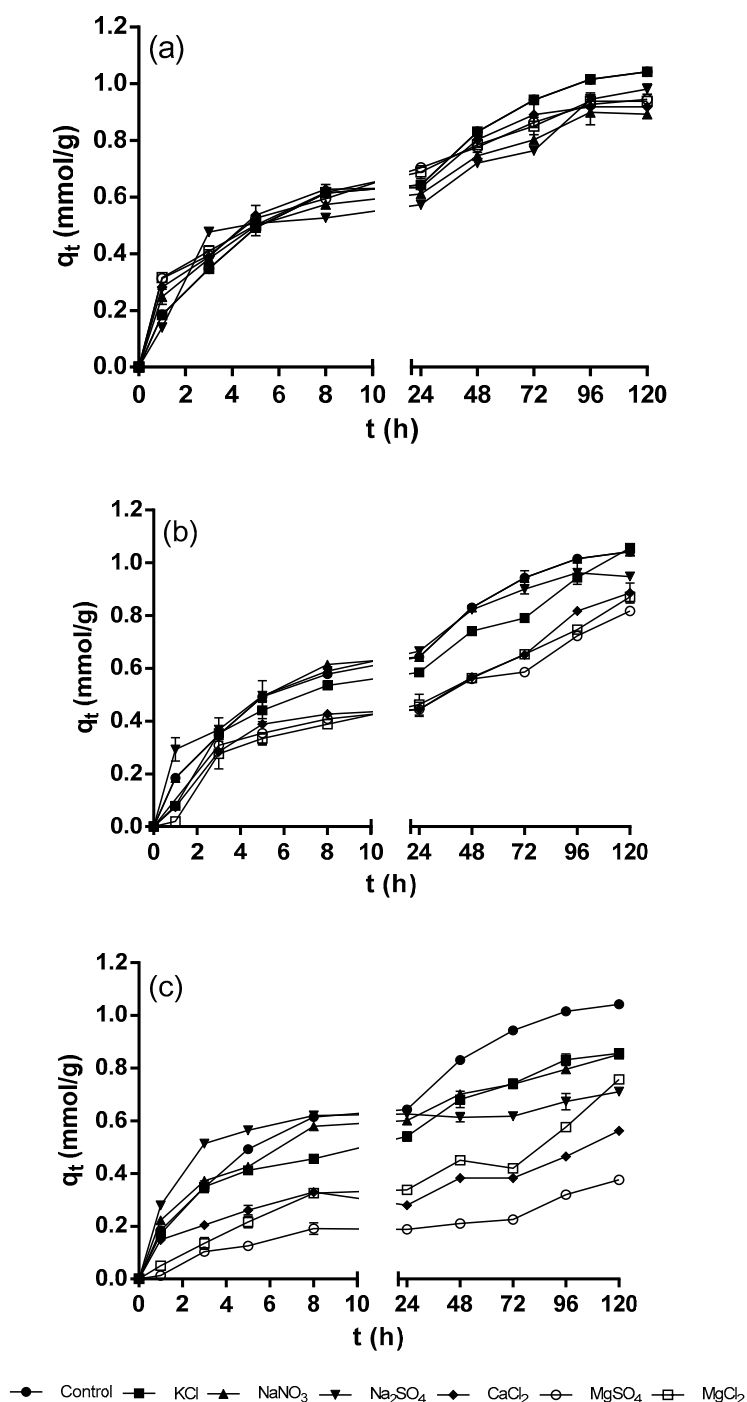
Figure 2 shows the kinetic profiles of the biosorption of  $\text{Ni}^{2+}$  by QCS at the different concentrations of background electrolytes. There was a small variation in the  $\text{Ni}^{2+}$  biosorption capacity over the course of the experiments for background electrolyte concentration of 0.2 mM compared to the control experiment (with no background electrolytes) (Figure 2a). However, there were no statistically significant differences in all cases ( $p > 0.05$ ), thus, it is concluded that a background electrolyte concentration of 0.2 mM has no negative effect on the biosorption of  $\text{Ni}^{2+}$ . Moreover, the global performance indexes for the biosorption of  $\text{Ni}^{2+}$  at a background electrolyte concentration of 0.2 mM were small and ranged from  $-7$  to  $-5\%$  (Table 3), which confirms that background electrolytes at a concentration of 0.2 mM have a negligible effect on the biosorption of  $\text{Ni}^{2+}$ .

**Table 3.** Effect of background electrolytes at concentrations of 0.2, 2, and 20 mM on the global performance index of the biosorption of  $\text{Ni}^{2+}$  by QCS.

| Background Electrolyte   | $\xi$ (%) |      |       |
|--------------------------|-----------|------|-------|
|                          | 0.2 mM    | 2 mM | 20 mM |
| KCl                      | −6        | −8   | −21   |
| $\text{NaNO}_3$          | −6        | −12  | −20   |
| $\text{Na}_2\text{SO}_4$ | −5        | −10  | −27   |
| $\text{CaCl}_2$          | −6        | −20  | −54   |
| $\text{MgSO}_4$          | −7        | −23  | −77   |
| $\text{MgCl}_2$          | −7        | −24  | −42   |

Increasing the concentration of background electrolytes to 2 mM led to a further decrease in the  $\text{Ni}^{2+}$  biosorption capacity. However, as shown in Figure 2b, the different background electrolytes influenced the biosorption of  $\text{Ni}^{2+}$  at varying degrees. The decreases in  $\text{Ni}^{2+}$  biosorption capacity were more significant in the presence of salts with divalent cations ( $\text{CaCl}_2$ ,  $\text{MgSO}_4$ , and  $\text{MgCl}_2$ ) and, to a lesser extent, with monovalent cations (KCl,  $\text{NaNO}_3$ , and  $\text{Na}_2\text{SO}_4$ ). The statistical analysis revealed that there were significant differences in  $\text{Ni}^{2+}$  biosorption capacities between the control and the experiments carried out in the presence of compounds with divalent cations ( $p < 0.05$ ). However, there were no statistically significant differences in the maximum  $\text{Ni}^{2+}$  biosorption capacities between the control and the experiments performed with monovalent cation salts (KCl,  $\text{NaNO}_3$ , and  $\text{Na}_2\text{SO}_4$ ) ( $p > 0.05$ ). The global performance indexes corroborated that the compounds containing divalent cations ( $\text{CaCl}_2$ ,  $\text{MgSO}_4$  and  $\text{MgCl}_2$ ) affected the biosorption of  $\text{Ni}^{2+}$  more negatively (from  $-24\%$  to  $-20\%$ ) compared to the compounds containing monovalent cations (KCl,  $\text{NaNO}_3$  and  $\text{Na}_2\text{SO}_4$ ) (from  $-12\%$  to  $-8\%$ ).

The background electrolytes at a concentration of 20 mM caused an even greater decrease in the  $\text{Ni}^{2+}$  biosorption capacity compared to that obtained at 2 mM (Figure 2c). The statistical analysis revealed that the differences between the  $\text{Ni}^{2+}$  biosorption capacities of the control and the experiments carried out with all the background electrolytes were statistically significant ( $p < 0.05$ ). For all the experiments, the lowest  $\text{Ni}^{2+}$  biosorption capacities were obtained when  $\text{MgSO}_4$ ,  $\text{CaCl}_2$ , and  $\text{MgCl}_2$  salts were present in the aqueous solutions (Figure 2c). Additionally, the global performance indexes showed that the biosorption of  $\text{Ni}^{2+}$  was more negatively affected at the highest concentration (20 mM) of background electrolytes and that the negative effect of the background electrolytes followed the order:  $\text{MgSO}_4 > \text{CaCl}_2 > \text{MgCl}_2 > \text{Na}_2\text{SO}_4 > \text{NaNO}_3 \approx \text{KCl}$ .



**Figure 2.** Effect of background electrolytes at a concentration of (a) 0.2 mM, (b) 2 mM, and (c) 20 mM on the biosorption of  $\text{Ni}^{2+}$  by QCS.

These results clearly show that the  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  cations have a more pronounced effect on the biosorption of  $\text{Ni}^{2+}$  from aqueous solutions by QCS compared to the  $\text{Na}^+$  and  $\text{K}^+$  cations. This could be attributed to the following reasons: First, the divalent  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  cations competed more efficiently with the  $\text{Ni}^{2+}$  ions for the biosorption active sites present on the QCS surface compared to the monovalent  $\text{Na}^+$  and  $\text{K}^+$  cations. Second, the divalent  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  cations biosorb more easily on the QCS surface compared to the monovalent  $\text{Na}^+$  and  $\text{K}^+$  cations because of the higher electrostatic attraction. Finally, a  $\text{Na}^+$  or  $\text{K}^+$  ion biosorbed on the QCS surface occupies only one biosorption active site, whereas a  $\text{Mg}^{2+}$  or  $\text{Ca}^{2+}$  ion could occupy two sites, thus resulting in a greater decrease in the

biosorption of  $\text{Ni}^{2+}$  in the presence of  $\text{MgCl}_2$ ,  $\text{MgSO}_4$ , and  $\text{CaCl}_2$  [24,35,46]. These reasons indicate that the biosorption active sites present on the surface of the QCS are not specific to  $\text{Ni}^{2+}$  ions.

Additionally, the decrease in  $\text{Ni}^{2+}$  biosorption can be owing to the presence of the  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  ions, which could increase the formation of nickel chloride and nickel sulfate complexes, respectively, thus decreasing the free  $\text{Ni}^{2+}$  ions in solution [46]. It was observed that the  $\text{SO}_4^{2-}$  ion had a more negative effect on the biosorption of  $\text{Ni}^{2+}$  by QCS compared to the  $\text{Cl}^-$  and  $\text{NO}_3^-$  ions.

Okoronkwo et al. [46] reported that the  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  cations and the  $\text{SO}_4^{2-}$  anion cause a decrease in the ability of the Mexican sunflower (*Tithonia diversifolia*) stems to bind the  $\text{Ni}^{2+}$  ions in aqueous solutions.  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  cations also decreased the biosorption capacity of *Phellinus igniarius* and *Fomes fomentarius* for Methylene Blue and Rhodamine B dyes [41]. Moreover,  $\text{Mn}^{2+}$  and  $\text{Ag}^+$  ions decreased the  $\text{Cu}^{2+}$  removal efficiency of granular activated carbon [47]. Likewise, an important decrease in biosorption capacity of *Macrocystis pyrifera* for benzene and toluene was observed, when an artificial seawater solution (Instant Ocean®, Blacksburg, VA, USA) was used, probably because this solution contains different ions [42].

Table 4 presents the equilibrium  $\text{Ni}^{2+}$  biosorption capacity ( $q_{e \text{ exp}}$ ) for the control experiment (without background electrolytes) and at the different concentrations of background electrolytes. From Table 4, it is evident that the highest biosorption capacity was obtained in the absence of background electrolytes. Moreover, the heavy metal biosorption capacity decreased as the concentration of background electrolytes increased, and the lowest equilibrium  $\text{Ni}^{2+}$  biosorption capacities were obtained in the presence of  $\text{MgSO}_4$ ,  $\text{CaCl}_2$ , and  $\text{MgCl}_2$  at a concentration of 20 mM. The Elovich, fractional power, and pseudo-second order kinetic models described the kinetic profiles of the biosorption of  $\text{Ni}^{2+}$  at the three different concentrations of background electrolytes more adequately, as evidenced by the lower SSE, RMSE, and AIC values and the higher  $r^2$  values (Tables 4–6).

The equilibrium  $\text{Ni}^{2+}$  biosorption capacities predicted by the pseudo-second order model were close to the values obtained experimentally, and they decreased as the concentration of the background electrolytes increased. It is also evident that background electrolytes affected the parameters of all the tested kinetic models, and this occurred to a greater extent at a concentration of 20 mM. Among the observed changes, the decrease in the specific biosorption rate of the fractional power model with increasing concentrations of background electrolytes should be noted.

The goodness of fit between  $\text{Ni}^{2+}$  biosorption kinetics and pseudo-second order, Elovich, and fractional power models suggests that the biosorption process of  $\text{Ni}^{2+}$  by QCS from aqueous solutions, containing different types and concentrations of background electrolytes, has a chemical process (chemisorption) as the rate-limiting step of the overall rate of heavy metal biosorption [18,29].

To our knowledge, the effect of ionic strength and background electrolytes on the parameters of kinetic models of the biosorption of  $\text{Ni}^{2+}$  has not been previously reported.

The results obtained in the present study clearly demonstrate the remarkable capacity of QCS to biosorb  $\text{Ni}^{2+}$  from aqueous solutions containing various types of impurities with different concentrations.

Additionally, the results of this study show that the impurities in the water greatly affect the performance of the biosorption of heavy metals. Furthermore, this study provides valuable information on the effect of ionic strength, the type, and concentration of background electrolytes on the biosorption of  $\text{Ni}^{2+}$ , which is of great importance for the application of biosorption technology in the treatment of industrial wastewater contaminated with  $\text{Ni}^{2+}$ .

**Table 4.** Parameters of the pseudo-first order and pseudo-second order models for the biosorption of  $\text{Ni}^{2+}$  by QCS at 0.2, 2, and 20 mM of background electrolytes.

| Concentration (mM) | Background Electrolyte   | Pseudo-First Order Model    |                   |                   |        |        |        |        | Pseudo-Second Order Model |                   |        |        |        |        |
|--------------------|--------------------------|-----------------------------|-------------------|-------------------|--------|--------|--------|--------|---------------------------|-------------------|--------|--------|--------|--------|
|                    |                          | $q_e^{\text{exp}}$ (mmol/g) | $k_1$ (1/h)       | $q_{e1}$ (mmol/g) | $r^2$  | SSE    | RMSE   | AIC    | $k_2$ (g/mmol·h)          | $q_{e2}$ (mmol/g) | $r^2$  | SSE    | RMSE   | AIC    |
| 0.2                | Control                  | 1.000                       | $0.142 \pm 0.026$ | $0.957 \pm 0.047$ | 0.9724 | 0.0633 | 0.0629 | −94.0  | $0.170 \pm 0.035$         | $1.034 \pm 0.041$ | 0.9870 | 0.0299 | 0.0433 | −107.5 |
|                    | KCl                      | 0.929                       | $0.180 \pm 0.044$ | $0.887 \pm 0.058$ | 0.9431 | 0.0983 | 0.0784 | −86.1  | $0.280 \pm 0.077$         | $0.936 \pm 0.047$ | 0.9734 | 0.0459 | 0.0536 | −99.8  |
|                    | $\text{NaNO}_3$          | 0.943                       | $0.135 \pm 0.042$ | $0.901 \pm 0.050$ | 0.9274 | 0.1452 | 0.0953 | −91.6  | $0.176 \pm 0.079$         | $0.975 \pm 0.041$ | 0.9574 | 0.0852 | 0.0730 | −104.6 |
|                    | $\text{Na}_2\text{SO}_4$ | 0.937                       | $0.163 \pm 0.038$ | $0.902 \pm 0.057$ | 0.9493 | 0.0929 | 0.0762 | −87.1  | $0.239 \pm 0.068$         | $0.957 \pm 0.051$ | 0.9724 | 0.0506 | 0.0562 | −98.0  |
|                    | $\text{CaCl}_2$          | 0.909                       | $0.193 \pm 0.038$ | $0.876 \pm 0.046$ | 0.9628 | 0.0624 | 0.0624 | −94.3  | $0.308 \pm 0.068$         | $0.922 \pm 0.036$ | 0.9836 | 0.0276 | 0.0415 | −108.9 |
|                    | $\text{MgSO}_4$          | 0.912                       | $0.190 \pm 0.049$ | $0.869 \pm 0.060$ | 0.9355 | 0.1054 | 0.0812 | −84.8  | $0.309 \pm 0.094$         | $0.914 \pm 0.051$ | 0.9672 | 0.0537 | 0.0579 | −97.0  |
|                    | $\text{MgCl}_2$          | 0.909                       | $0.192 \pm 0.049$ | $0.870 \pm 0.059$ | 0.9371 | 0.1025 | 0.0800 | −85.3  | $0.315 \pm 0.095$         | $0.914 \pm 0.050$ | 0.9679 | 0.0523 | 0.0572 | −97.4  |
| 2                  | KCl                      | 0.931                       | $0.133 \pm 0.040$ | $0.882 \pm 0.070$ | 0.9355 | 0.1397 | 0.0935 | −79.7  | $0.159 \pm 0.065$         | $0.967 \pm 0.077$ | 0.9553 | 0.0968 | 0.0778 | −86.4  |
|                    | $\text{NaNO}_3$          | 0.865                       | $0.187 \pm 0.040$ | $0.829 \pm 0.072$ | 0.9529 | 0.0723 | 0.0672 | −79.1  | $0.308 \pm 0.066$         | $0.875 \pm 0.070$ | 0.9771 | 0.0352 | 0.0469 | −88.6  |
|                    | $\text{Na}_2\text{SO}_4$ | 0.897                       | $0.181 \pm 0.060$ | $0.844 \pm 0.075$ | 0.9093 | 0.1625 | 0.1008 | −77.0  | $0.272 \pm 0.121$         | $0.899 \pm 0.074$ | 0.9383 | 0.1105 | 0.0831 | −84.0  |
|                    | $\text{CaCl}_2$          | 0.785                       | $0.121 \pm 0.035$ | $0.782 \pm 0.066$ | 0.9454 | 0.0732 | 0.0750 | −71.7  | $0.174 \pm 0.071$         | $0.845 \pm 0.071$ | 0.9602 | 0.0533 | 0.0641 | −76.4  |
|                    | $\text{MgSO}_4$          | 0.709                       | $0.119 \pm 0.029$ | $0.765 \pm 0.062$ | 0.9644 | 0.0316 | 0.0562 | −62.2  | $0.192 \pm 0.059$         | $0.812 \pm 0.057$ | 0.9795 | 0.0181 | 0.0426 | −68.9  |
|                    | $\text{MgCl}_2$          | 0.757                       | $0.108 \pm 0.033$ | $0.755 \pm 0.065$ | 0.9465 | 0.0696 | 0.0732 | −72.4  | $0.153 \pm 0.065$         | $0.824 \pm 0.073$ | 0.9602 | 0.0517 | 0.0631 | −76.9  |
| 20                 | KCl                      | 0.810                       | $0.152 \pm 0.052$ | $0.737 \pm 0.063$ | 0.8968 | 0.1490 | 0.0910 | −90.5  | $0.230 \pm 0.088$         | $0.811 \pm 0.058$ | 0.9499 | 0.0724 | 0.0634 | −104.9 |
|                    | $\text{NaNO}_3$          | 0.795                       | $0.204 \pm 0.054$ | $0.737 \pm 0.048$ | 0.9309 | 0.0921 | 0.0715 | −100.1 | $0.350 \pm 0.104$         | $0.793 \pm 0.041$ | 0.9666 | 0.0445 | 0.0497 | −114.6 |
|                    | $\text{Na}_2\text{SO}_4$ | 0.667                       | $0.524 \pm 0.090$ | $0.642 \pm 0.020$ | 0.9779 | 0.0190 | 0.0325 | −131.7 | $1.325 \pm 0.355$         | $0.671 \pm 0.023$ | 0.9766 | 0.0201 | 0.0334 | −130.5 |
|                    | $\text{CaCl}_2$          | 0.470                       | $0.212 \pm 0.072$ | $0.427 \pm 0.040$ | 0.9031 | 0.0383 | 0.0505 | −95.8  | $0.698 \pm 0.335$         | $0.451 \pm 0.040$ | 0.9287 | 0.0282 | 0.0433 | −101.0 |
|                    | $\text{MgSO}_4$          | 0.307                       | $0.211 \pm 0.054$ | $0.211 \pm 0.016$ | 0.9576 | 0.0040 | 0.0175 | −115.4 | $1.158 \pm 0.572$         | $0.233 \pm 0.023$ | 0.9486 | 0.0048 | 0.0192 | −112.5 |
|                    | $\text{MgCl}_2$          | 0.584                       | $0.047 \pm 0.024$ | $0.627 \pm 0.098$ | 0.8653 | 0.1335 | 0.0913 | −80.6  | $0.089 \pm 0.063$         | $0.715 \pm 0.117$ | 0.9117 | 0.0875 | 0.0739 | −88.1  |

**Table 5.** Parameters of the Elovich and intraparticle diffusion models for the biosorption of  $\text{Ni}^{2+}$  by QCS at 0.2, 2, and 20 mM of background electrolytes.

| Concentration (mM) | Background Electrolyte          | Elovich          |                |        |        |        |        | Intraparticle Diffusion             |               |        |        |        |        |
|--------------------|---------------------------------|------------------|----------------|--------|--------|--------|--------|-------------------------------------|---------------|--------|--------|--------|--------|
|                    |                                 | $A_e$ (mmol/g·h) | $B_e$ (g/mmol) | $r^2$  | SSE    | RMSE   | AIC    | $k_{id}$ (mmol/g·h <sup>0.5</sup> ) | $c$ (mmol/g)  | $r^2$  | SSE    | RMSE   | AIC    |
| 0.2                | Control                         | 0.418 ± 0.110    | 5.369 ± 0.418  | 0.9916 | 0.0193 | 0.0348 | −115.3 | 0.087 ± 0.014                       | 0.183 ± 0.089 | 0.9126 | 0.2006 | 0.1120 | −73.22 |
|                    | KCl                             | 0.918 ± 0.196    | 6.937 ± 0.357  | 0.9960 | 0.0069 | 0.0208 | −133.9 | 0.074 ± 0.014                       | 0.236 ± 0.091 | 0.8803 | 0.2067 | 0.1137 | −72.69 |
|                    | NaNO <sub>3</sub>               | 0.856 ± 0.304    | 7.391 ± 0.632  | 0.9891 | 0.0168 | 0.0324 | −117.9 | 0.069 ± 0.015                       | 0.222 ± 0.091 | 0.8641 | 0.2088 | 0.1142 | −72.51 |
|                    | Na <sub>2</sub> SO <sub>4</sub> | 0.738 ± 0.248    | 6.552 ± 0.560  | 0.9890 | 0.0201 | 0.0354 | −114.7 | 0.077 ± 0.015                       | 0.224 ± 0.092 | 0.8840 | 0.2124 | 0.1152 | −72.20 |
|                    | CaCl <sub>2</sub>               | 1.051 ± 0.392    | 7.210 ± 0.625  | 0.9888 | 0.0188 | 0.0343 | −115.8 | 0.072 ± 0.016                       | 0.245 ± 0.102 | 0.8437 | 0.2623 | 0.1280 | −68.40 |
|                    | MgSO <sub>4</sub>               | 1.078 ± 0.331    | 7.306 ± 0.518  | 0.9925 | 0.0123 | 0.0278 | −123.4 | 0.072 ± 0.015                       | 0.243 ± 0.092 | 0.8690 | 0.2141 | 0.1157 | −72.06 |
|                    | MgCl <sub>2</sub>               | 1.128 ± 0.355    | 7.362 ± 0.529  | 0.9922 | 0.0126 | 0.0281 | −123.0 | 0.071 ± 0.015                       | 0.247 ± 0.094 | 0.8643 | 0.2211 | 0.1175 | −71.48 |
| 2                  | KCl                             | 0.284 ± 0.139    | 5.350 ± 0.845  | 0.9689 | 0.0674 | 0.0649 | −92.85 | 0.085 ± 0.013                       | 0.135 ± 0.084 | 0.9187 | 0.1760 | 0.1049 | −75.58 |
|                    | NaNO <sub>3</sub>               | 0.409 ± 0.137    | 5.765 ± 0.568  | 0.9863 | 0.0275 | 0.0414 | −109.0 | 0.082 ± 0.011                       | 0.172 ± 0.071 | 0.9372 | 0.1257 | 0.0886 | −81.65 |
|                    | Na <sub>2</sub> SO <sub>4</sub> | 0.600 ± 0.407    | 6.655 ± 1.190  | 0.9548 | 0.0810 | 0.0712 | −89.55 | 0.075 ± 0.016                       | 0.197 ± 0.098 | 0.8653 | 0.2412 | 0.1228 | −69.21 |
|                    | CaCl <sub>2</sub>               | 0.257 ± 0.135    | 6.352 ± 1.061  | 0.9715 | 0.0382 | 0.0542 | −81.42 | 0.070 ± 0.012                       | 0.125 ± 0.077 | 0.9223 | 0.1041 | 0.0895 | −66.38 |
|                    | MgSO <sub>4</sub>               | 0.342 ± 0.117    | 7.261 ± 0.740  | 0.9920 | 0.0071 | 0.0266 | −80.22 | 0.062 ± 0.014                       | 0.151 ± 0.084 | 0.9126 | 0.0774 | 0.0880 | −51.52 |
|                    | MgCl <sub>2</sub>               | 0.191 ± 0.098    | 6.178 ± 1.074  | 0.9709 | 0.0379 | 0.0540 | −81.55 | 0.069 ± 0.011                       | 0.100 ± 0.071 | 0.9329 | 0.0872 | 0.0819 | −69.02 |
| 20                 | KCl                             | 0.399 ± 0.128    | 6.997 ± 0.418  | 0.9865 | 0.0196 | 0.0330 | −131.1 | 0.069 ± 0.010                       | 0.167 ± 0.064 | 0.9142 | 0.1238 | 0.0830 | −94.19 |
|                    | NaNO <sub>3</sub>               | 0.815 ± 0.334    | 8.064 ± 0.639  | 0.9839 | 0.0214 | 0.0345 | −129.3 | 0.064 ± 0.014                       | 0.217 ± 0.083 | 0.8440 | 0.2077 | 0.1074 | −83.84 |
|                    | Na <sub>2</sub> SO <sub>4</sub> | 0.425 ± 0.587    | 15.69 ± 0.790  | 0.9219 | 0.0675 | 0.0612 | −106.3 | 0.041 ± 0.019                       | 0.323 ± 0.116 | 0.5324 | 0.4025 | 0.1495 | −70.61 |
|                    | CaCl <sub>2</sub>               | 0.576 ± 0.479    | 14.73 ± 1.665  | 0.9505 | 0.0195 | 0.0361 | −107.2 | 0.037 ± 0.009                       | 0.120 ± 0.054 | 0.8315 | 0.0665 | 0.0666 | −86.41 |
|                    | MgSO <sub>4</sub>               | 0.163 ± 0.170    | 24.01 ± 7.875  | 0.9113 | 0.0082 | 0.0253 | −104.3 | 0.024 ± 0.009                       | 0.050 ± 0.039 | 0.7443 | 0.0239 | 0.0429 | −88.45 |
|                    | MgCl <sub>2</sub>               | 0.074 ± 0.041    | 6.100 ± 1.519  | 0.9453 | 0.0542 | 0.0582 | −96.78 | 0.062 ± 0.009                       | 0.040 ± 0.042 | 0.9486 | 0.0509 | 0.0564 | −97.91 |

**Table 6.** Parameters of the fractional power model for the biosorption of  $\text{Ni}^{2+}$  by QCS at 0.2, 2, and 20 mM of background electrolytes.

| Fractional Power   |                          |                          |                   |                             |        |        |        |        |
|--------------------|--------------------------|--------------------------|-------------------|-----------------------------|--------|--------|--------|--------|
| Concentration (mM) | Background Electrolyte   | $k_{\text{FP}}$ (mmol/g) | $v$ (1/h)         | $k_{\text{FP}}v$ (mmol/g·h) | $r^2$  | SSE    | RMSE   | AIC    |
| 0.2                | Control                  | $0.289 \pm 0.043$        | $0.275 \pm 0.036$ | 0.0793                      | 0.9768 | 0.0534 | 0.0578 | −97.02 |
|                    | KCl                      | $0.340 \pm 0.025$        | $0.221 \pm 0.018$ | 0.0753                      | 0.9906 | 0.0162 | 0.0318 | −118.6 |
|                    | $\text{NaNO}_3$          | $0.320 \pm 0.036$        | $0.220 \pm 0.028$ | 0.0705                      | 0.9786 | 0.0329 | 0.0453 | −105.8 |
|                    | $\text{Na}_2\text{SO}_4$ | $0.327 \pm 0.035$        | $0.233 \pm 0.027$ | 0.0761                      | 0.9825 | 0.0321 | 0.0448 | −106.2 |
|                    | $\text{CaCl}_2$          | $0.352 \pm 0.041$        | $0.210 \pm 0.029$ | 0.0741                      | 0.9755 | 0.0411 | 0.0507 | −101.8 |
|                    | $\text{MgSO}_4$          | $0.347 \pm 0.027$        | $0.213 \pm 0.020$ | 0.0738                      | 0.9887 | 0.0185 | 0.0340 | −116.1 |
|                    | $\text{MgCl}_2$          | $0.351 \pm 0.028$        | $0.210 \pm 0.020$ | 0.0738                      | 0.9879 | 0.0197 | 0.0351 | −115.0 |
| 2                  | KCl                      | $0.232 \pm 0.053$        | $0.308 \pm 0.055$ | 0.0714                      | 0.9579 | 0.0911 | 0.0755 | −87.43 |
|                    | $\text{NaNO}_3$          | $0.269 \pm 0.028$        | $0.278 \pm 0.026$ | 0.0747                      | 0.9881 | 0.0237 | 0.0385 | −111.6 |
|                    | $\text{Na}_2\text{SO}_4$ | $0.295 \pm 0.061$        | $0.246 \pm 0.051$ | 0.0724                      | 0.9442 | 0.0999 | 0.0790 | −85.77 |
|                    | $\text{CaCl}_2$          | $0.209 \pm 0.047$        | $0.294 \pm 0.053$ | 0.0614                      | 0.9659 | 0.0457 | 0.0593 | −78.71 |
|                    | $\text{MgSO}_4$          | $0.236 \pm 0.021$        | $0.254 \pm 0.022$ | 0.0598                      | 0.9942 | 0.0051 | 0.0226 | −84.11 |
|                    | $\text{MgCl}_2$          | $0.179 \pm 0.045$        | $0.319 \pm 0.059$ | 0.0572                      | 0.9646 | 0.0461 | 0.0595 | −78.6  |
|                    |                          |                          |                   |                             |        |        |        |        |
| 20                 | KCl                      | $0.245 \pm 0.025$        | $0.264 \pm 0.026$ | 0.0645                      | 0.9845 | 0.0224 | 0.0351 | −128.4 |
|                    | $\text{NaNO}_3$          | $0.301 \pm 0.035$        | $0.216 \pm 0.030$ | 0.0652                      | 0.9719 | 0.0374 | 0.0456 | −118.1 |
|                    | $\text{Na}_2\text{SO}_4$ | $0.423 \pm 0.059$        | $0.107 \pm 0.038$ | 0.0451                      | 0.9078 | 0.0794 | 0.0664 | −103.1 |
|                    | $\text{CaCl}_2$          | $0.175 \pm 0.031$        | $0.213 \pm 0.045$ | 0.0372                      | 0.9472 | 0.0208 | 0.0373 | −106.1 |
|                    | $\text{MgSO}_4$          | $0.085 \pm 0.027$        | $0.239 \pm 0.093$ | 0.0204                      | 0.8746 | 0.0117 | 0.0300 | −99.14 |
|                    | $\text{MgCl}_2$          | $0.101 \pm 0.029$        | $0.404 \pm 0.069$ | 0.0406                      | 0.9564 | 0.0432 | 0.0520 | −100.9 |
|                    |                          |                          |                   |                             |        |        |        |        |



#### 4. Conclusions

The results obtained in this study demonstrated that QCS is a useful biosorbent for the bioremediation of aqueous solutions contaminated with  $\text{Ni}^{2+}$  and containing inorganic impurities. The effect of background electrolytes varied depending on the type and concentration of the electrolyte. The biosorption of  $\text{Ni}^{2+}$  was not affected by a NaCl ionic strength of 0.2 mM but was affected by higher ionic strengths. In addition, the background electrolytes (KCl,  $\text{NaNO}_3$ ,  $\text{Na}_2\text{SO}_4$ ,  $\text{CaCl}_2$ ,  $\text{MgSO}_4$ , and  $\text{MgCl}_2$ ) had no effect on the biosorption of the heavy metal at a concentration of 0.2 mM. However, at concentrations of 2 and 20 mM, the divalent  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  cations affected the biosorption of  $\text{Ni}^{2+}$  significantly, whereas the monovalent  $\text{Na}^+$  and  $\text{K}^+$  cations affected the biosorption of  $\text{Ni}^{2+}$  only slightly. The kinetic experimental data were well represented using the Elovich, fractional power, and pseudo-second order models. In order to determine the full potential of QCS as a commercial biosorbent,  $\text{Ni}^{2+}$  biosorption from industrial wastewater will be evaluated in a future research study.

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