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Study of the removal of residual aluminum through the biopolymers carboxymethylcellulose, chitin, and chitosan

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ABSTRACT

Aluminum is currently associated to health problems, especially dementia, and drinking water is one of the most potential sources to Al^{3+} ingestion. In this work, the Al^{3+} removal capacities of the biopolymers carboxymethylcellulose (CMC), chitin (QTN), and chitosan (QUIT) were evaluated for synthetic water with a drinking water Al^{3+} range level and well water with a high concentration of natural Al^{3+}. Isothermal and kinetics essays were carried out. The laboratorial tests have demonstrated that CMC is not an efficient Al^{3+} removal agent, whereas QTN and QUIT have shown very promising results. QTN and QUIT best fitted Sips isotherm model. However, the Freundlich model cannot be discarded for QUIT. Pseudo-second-order kinetic models fitted very well for the experimental conditions for both QTN and QUIT. It was notice that QUIT removes Al^{3+} faster than QTN, with complete decontamination of well water occurring within 120 min of contact time.

Keywords: Drinking water; Aluminum removal; Carboxymethylcellulose; Chitin; Chitosan

1. Introduction

It is wellknown that aluminum is present at various concentrations and in different forms in the environment. Over the years, the contact and ingestion of this element by population became more usual, especially due to the development of industrialized products and its utilization in several different ways in this regard. Deodorants, sun screens, antacids, treated drinking water, among others, are examples of common products containing aluminum [1]. Recent studies have shown that the absorption of aluminum by the human body is often higher than that reported by early studies, particularly in the case of the most hazardous species like Al^{3+}, $Al(OH)_2^+$, and $AlOH^{2+}$. These species are more likely to be ingested by humans through drinking water [2–4].

During the past years, several researchers studied the relation between aluminum and health problems. It is now recognized that aluminum can cause kidney and bone complications, lung disability, and is

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associated with dementia processes [2,5–7]. Due to the difficulties of measuring small aluminum amounts and identifying the mechanisms by which it reaches human's tissues, it is hard to define a safe concentration for human ingestion [2]. For drinking water, most countries establish a maximum value determined by the World Health Organization (WHO) guide of $0.20\,mg\,L^{-1}$ of total aluminum [8].

The dissolved aluminum in water from artesian wells usually comes from a region that contains aluminum in soil, acid surface or rainfall water, and areas contaminated with industrial wastewater that produces large amounts of aluminum [9]. The drinking water analyzed in this work comes from artesian wells from south of Brazil, and has a high content in natural monomeric Al^{3+}. Given the problems associated with the excess of aluminum and other metals in water supplies, many techniques and materials have been proposed to remove metallic ions from water [10–15]. The present study utilizes concurrently three different biopolymers, namely, carboxymethylcellulose (CMC), chitin (QTN), and chitosan (QUIT) to remove the Al^{3+} from drinking water under controlled conditions [16] .

These biopolymers have N- or O-donor functional groups that promptly form compounds with high polarized ions, like the Al^{3+}. The molecular structure of QUIT is chemically similar to cellulose, differing only by the functional hydroxyl group (–OH) of the latter and the amino group ($-NH_2$) of the former [16]. QUIT has been highlighted in studies to remove metal ions, due to its intrinsic adsorbent characteristics associated to the ability of forming complexes with transitional metallic ions due to the presence of amino groups in its structure [17–19]. By its turn, QTN, precursor of QUIT, has been less studied in regard to its adsorption capacity [20,21]. With respect to CMC, previous studies have demonstrated the effectiveness in removing aluminum in contaminated river water, despite the presence of humic material [22].

Considering the literature survey provided above, the purpose of this study is to evaluate and compare the potential of CMC, QTN, and QUIT biopolymers in removing Al^{3+} at low concentrations, typical for water supply sources. For this intend, batch isothermal experiments with synthetic water were conducted and results analyzed according the Langmuir, Freundlich, and Sips models. Batch kinetics experiments were also performed with artesian well water containing high Al^{3+} concentration ($0.80\,mg\,L^{-1}$).

2. Materials and methods

CMC (Sodium Carboxymethylcellulose 250.000 Daltons) and QUIT (75–85% deacetylated chitosan) were provided by Sigma-Aldrich®. QTN was extracted from shrimps shell, pulverized, and sieved before used. The partial structure of all the biopolymers is shown in Fig. 1.

Similar experimental procedures were carried for the three biopolymers (CMC, QTN, and QUIT), with all the experiments conducted in duplicate. Due to the presence of aluminum in the laboratorial environment, glassware were previously rinsed with hydrochloric acid solution (HCl 1:1) and subsequently washed with distilled water and dried out. In order to avoid contaminating glassware with aluminum, high density polypropylene erlenmeyers were used.

Isothermal experiments were carried out with synthetic water with predefined Al^{3+} concentration, whereas kinetics tests were conducted with well water with high Al^{3+} concentration ($0.80\,mg\,L^{-1}$). All experiments were performed with a thermostatic bath, keeping constant both the temperature and the agitation during the whole experiment period.

A Hach DR2010 spectrophotometer was used for Al^{3+} concentration measurements. The Aluminon Method with Hach® kits for colorimetric spectrophotometry was utilized in this regard. This method allows quantification for ion Al^{3+} at concentrations in the range from 0.00 up to 0.80 $mg\,L^{-1}$, with a $0.01\,mg\,L^{-1}$ of accuracy.

2.1. Batch isotherm experiments

Al^{3+} stock solutions were prepared by dissolving Alum ($Al_2(SO_4)_3\cdot(14–18)\cdot H_2O$). For isothermal experiments, each test was prepared diluting the stock solution in deionized water to produce nine different Al^{3+}

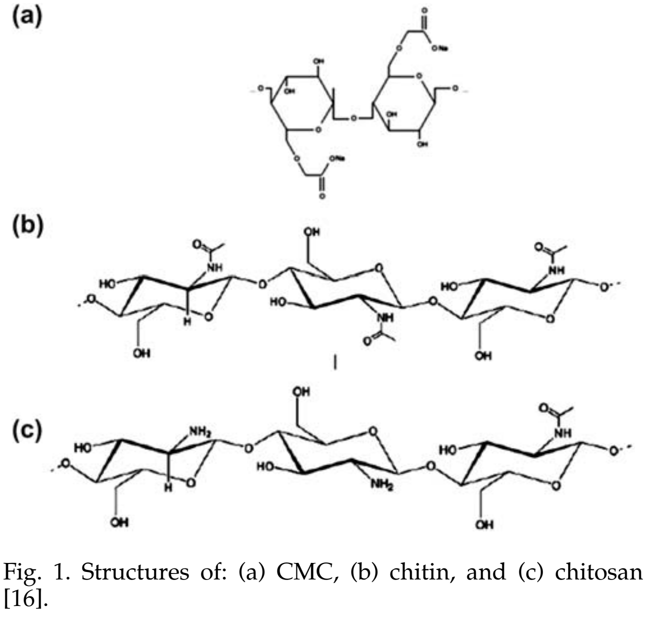

Fig. 1. Structures of: (a) CMC, (b) chitin, and (c) chitosan [16].

concentrations (ranging from 0.05 to 1.50 mg Al^{3+} L^{-1} or from 0.05 to 3.00 mg Al^{3+} L^{-1}) and a blank sample. Then, 200 mL of each solution were poured in 250 mL polypropylene erlenmeyers with the pre-establish amount of biopolymers (see Fig. 2), then agitated in a thermostatic bath during 24 h to ensure that the equilibrium was reached. When necessary, pH reduction was achieved by adding nitric acid (HNO_3 54%). Temperature was controlled at 298 K, except for one experiment that was carried out at 308 K. After the 24 h, the erlenmeyers were withdrawn, the contents filtered in a cellulose acetate filter (porosity 45 μm) and the corresponding Al^{3+} concentrations were accurately measured.

Al^{3+} adsorption capacity by the biopolymers (q_t, mg Al^{3+} g^{-1} biopolymer) was calculated according to the Eq. (1), where C_0 (mg L^{-1}) is the initial Al^{3+} concentration, C_t (mg L^{-1}) is the Al^{3+} concentration at time t (min.), V is the batch volume (L) and m (g) is the biopolymer mass utilized.

$$q_t = \frac{(C_0 - C_t)V}{m} \tag{1}$$

Three different isothermal models were tested with the experimental data collected from biopolymer/Al^{3+} adsorption experiments, namely, Langmuir, Freundlich, and Sips models. Langmuir adsorption isotherm assumes that adsorption occurs in a finite adsorption monolayer with a fixed number of active sites. This model considers that all sites exhibit equal affinity for the adsorbate. Langmuir's adsorption curve ($q_e \times C_e$) is characterized by a plateau when the equilibrium saturation is reached, since it assumes that when the molecules occupy an active site no further adsorption can take place on it [23,24]. The model is represented by the Eq. (2):

$$q_e = \frac{q_{max} K_L C_e}{1 + K_L C_e} \tag{2}$$

where q_e is the equilibrium adsorption capacity, q_{max} is the maximum adsorption capacity for the Langmuir model, C_e is the equilibrium concentration of Al^{3+}, and K_L is the Langmuir adsorption constant (L mg^{-1}). The linearization of Langmuir model in the form of $C_e/q_e \times C_e$ plot makes possible to calculate the K_L and q_{max} values.

Freundlich isotherm model establishes a nonideal and reversible adsorption process, which is not restricted to the formation of a monolayer in the adsorbent. This empirical model can be applied to multilayer adsorption, since a nonuniform distribution of adsorption sites and affinities over the heterogeneous surface is considered. This means that the adsorbed amount is the summation of adsorption in all sites, with the strongest binding sites occupied preferentially, until the adsorption energy is exponentially decreased upon the completion of the adsorption process. The model is represented by Eq. (3):

$$q_e = K_F C_e^{1/n} \tag{3}$$

where C_e represents the equilibrium concentration of Al^{3+}, K_F represents the Freundlich adsorption constant

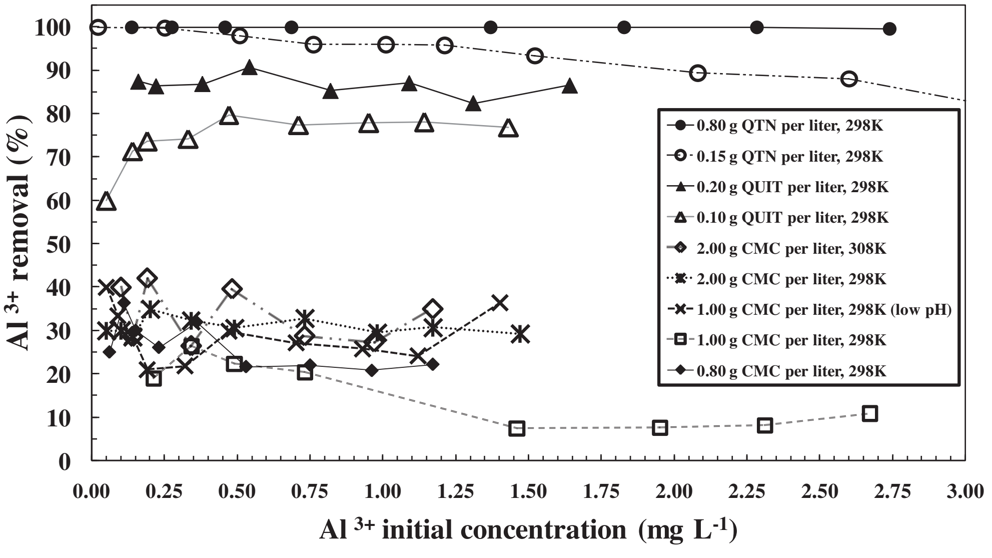

Fig. 2. Al^{3+} removal percentage plotted against initial Al^{3+} concentration in the aqueous solution, for three different biopolymeric removal agents in various different amounts (V = 200 mL, agitation = 200 rpm).

for the reaction ($L\,g^{-1}$), and n represents the measure of surface heterogeneity. The linearization of Freundlich model is in the form of log $q_e \times \log C_e$ plot [23,24].

Sips isotherm model is basically a combination of Langmuir and Freundlich models. It is deduced from predicting the heterogeneous adsorption systems and circumventing the limitation of the rising adsorbate concentration associated with Freundlich isotherm model. At low adsorbate concentrations, it reduces to Freundlich isotherm model, while at high concentrations it predicts a monolayer adsorption capacity typical of the Langmuir isotherm model. The equation parameters are governed mainly by conditions like pH, temperature, and concentration [23–25]. It is represented by Eq. (4):

$$q_e = \frac{q_{\max} K_S C_e^m}{1 + K_S C_e^m} \tag{4}$$

where q_e is the equilibrium adsorption capacity, C_e is the equilibrium concentration of Al^{3+}, K_S is the Sips adsorption constant ($L\,mg^{-1}$) of the reaction, and m is a measure of surface heterogeneity. In this study, Sips model was calculated by nonlinear regression, utilizing Solver add-in from Microsoft Excel®. The linearization of this model is given by ln $K_s\ q_e{}^{-1} \times \ln C_e$ plot.

The sum of the squares of the errors (ERRSQ) is one of the most commonly used error functions. The results fit better to a particular model when smaller differences are calculated from both the model and the corresponding experimental data. ERSSQ is give by Eq. (5).

$$\text{ERRSQ} = \sum_{i=1}^{n} (q_{e\,\exp.} - q_{e\,\text{cal}.})_i^2 \tag{5}$$

2.2. Batch kinetics studies

Kinetic experiments and Al^{3+} removal from well water were conducted utilizing natural water from artesian wells from a south Brazilian town (Araranguá-SC), with Al^{3+} concentrations around $0.80\,mg\,L^{-1}$. Erlenmeyers containing 200 mL of the natural water were added with 0.16 g of biopolymer during different periods of time (from 5 up to 1,440 min), at controlled agitation and temperature conditions (respectively, 200 rpm and 298 K). Three different kinetic models have been used extensively in the literature [11,13,15,17,26]. The pseudo-first-order equation, given by Eq. (6).

$$\log(q_e - q_t) = \log(q_e) - \frac{K_1 t}{2.303} \tag{6}$$

where K_1 is the rate constant of pseudo-first-order adsorption (min^{-1}) and q_e and q_t are the amount of Al^{3+} adsorbed per gram of biopolymer ($mg\,g^{-1}$) at equilibrium and time t (min), respectively. The linearization of this model is given by the plotting of log ($q_e - q_t$) vs. t. A straight line would suggest the applicability of this kinetic model to fit the experimental data. The first-order rate constant K_1 and equilibrium adsorption density (q_e) can be calculated from the slope of the straight line and its interception with y axis, respectively.

The pseudo-second kinetic model is based on experimental information of solid phase sorption. Generally, it has been applied to heterogeneous systems, where the sorption mechanism is attributed to chemical sorption. The pseudo-second-order kinetic model relies on the assumption that the rate-limiting step may be chemisorptions involving valence forces through sharing or exchange of electrons. The pseudo-second-order equation is generally expressed as Eq. (7):

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t \tag{7}$$

where K_2 is the constant rate of sorption ($g\,mg^{-1}\,min^{-1}$), q_e is the amount of solute sorbed at equilibrium, and q_t is the amount of solute sorbed on the sorbent surface at time t. The adsorption constant rate h ($mg\,g^{-1} min^{-1}$) is given by the term $K_2 q_e^2$. The constants q_e and K_2 can be determined by plotting t/q_t vs. t.

The intra-particle diffusion kinetic model consists of a simple model in which the rate of intra-particle diffusion can be obtained by the linearization of the curve represented by Eq. (8):

$$q_t = K_d t^{1/2} + I \tag{8}$$

where K_d is the constant rate of sorption ($g\,mg\,min^{-1/2}$), q_t is the amount of solute sorbed on the sorbent surface at time t and I is the interception with y axis.

3. Results and discussion

3.1. Al^{3+} removal from synthetic water

Several tests were performed with the three biopolymers (CMC, QTN, and QUIT) with the aim of verifying the lowest amount required to remove a satisfactory amount of Al^{3+} from the synthetic water. At first, an amount of 0.80 g of biopolymer per liter of solution was established for the initial tests [22]. Removal results are displayed in Fig. 2.

Experiments with a CMC contents of 0.80 g L^{-1} at 298 K and 200 rpm agitation showed unsatisfactory removal results, with no more than 37% of Al^{3+} being removed from any tested solution. Also, the higher initial Al^{3+} concentration the worse were the results for this biopolymer.

Modifications of the controlled conditions were then implemented in order to improve Al removal by CMC. The first attempt was to increase the CMC content while maintaining pH level, temperature, and bath agitation of the initial test. Unfortunately, the results with 1.00 and 2.00 g L^{-1} CMC were not satisfactory, with an average removal percentage of 30.31% only. Another attempt was to decrease the initial pH of the solution, from approximately 4.4–2.8 by adding nitric acid, using 1.00 g L^{-1} CMC maintaining fixed other testing parameters. Al^{3+} removal was just slightly increased, 2.2% in average. One last try was to keep the highest biopolymer concentration and increase temperature (from 298 to 308 K), which once more resulted in poor removal percentages (below 42%). Owing to the disappointing results, CMC biopolymer was not considered a potential agent for Al^{3+} removal in the drinking water typical range of concentrations; therefore it was discarded from the next set of experiments in this work.

By contrast, QTN showed the best results on Al^{+3} removals (Fig. 2). With 0.80 g L^{-1} of this biopolymer it was possible to completely remove Al^{3+} contained in all aqueous concentrations tested. An even lower QTN concentration of 0.15 g L^{-1} was tested, which still so showed very satisfactory removal percentage (94% on average). Thus, after treatment with only 0.15 g L^{-1} QTN and initial Al^{3+} concentrations equal or lower than 2.00 mg L^{-1}, the residual aluminum concentration was lower than 0.20 mg L^{-1}, maximum allowed value by the WHO [8].

Due to the high cost of QUIT, this biopolymer was essayed in similar and lower quantities than QTN in order to evaluate an economically affordable removal procedure. Thus, both the 0.10 and the 0.20 g L^{-1} testing conditions were analyzed. The results were considered satisfactory, with removal percentages of 76 and 86% on average, respectively, standing below the level attained by QTN for similar testing conditions.

It is worth noting that, as expected, the pH of the treated solutions varied within the experimental period of 24 h due to they were no buffered. The experiments showed initial and final pH values around 4.5 and 6.5, respectively. It can be inferred that the pH increased due to the Al^{3+} removal from the solution (which exhibits Lewis acid characteristics) by adsorption on the biopolymer. However, the possibility of Al^{3+} removal via precipitation as $Al(OH)_3$ at pH level of order of 6.0 cannot be discarded [27]. Nevertheless, the fact of the solution treated with CMC showed a similar behavior relative to the pH variation to those treated with the others biopolymers, despite the low Al^{3+} removal, indicates that the Al^{3+} removal by the biopolymers was the main removal via in the cases of QTN and QUIT.

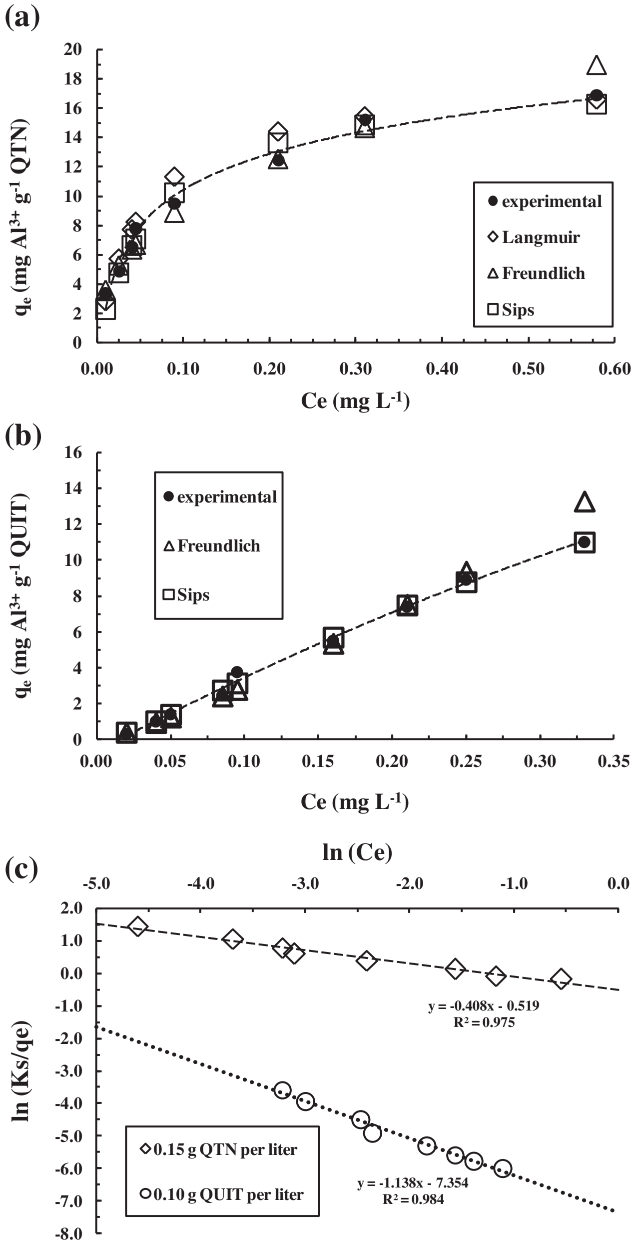

Fig. 3. Isotherm models for (a) 0.15 g L^{-1} QTN; (b) 0.10 g L^{-1} QUIT; and (c) Sips linearization for the biopolymers (conditions 298 K, 200 rpm, 24 h).

3.2. Isothermal experiments

In regard to the adsorption isothermal experiments, the tests with 0.15 g QTN L^{-1} and 0.10 g QUIT L^{-1} were analyzed on the basis to the removal experiments. Fig. 3 shows the experimental data and the Langmuir, Freundlich, and Sips concurrent isothermal models for QTN (Fig. 3(a)) and QUIT (Fig 3 (b)). Fig. 3(c) exhibits the Sips linearization for both biopolymers.

The isotherms that best fit the experimental data were chosen by comparing the linearization correlation coefficient R^2 (higher values) and the sum of the square of the errors ERSSQ (lower values, Eq. (5)), both values listed in Table 1. The Langmuir model was discarded for QUIT due to the very low R^2 value. The others R^2 values were very similar (~0.98) and then the isothermal model that best fit the experimental data was selected in function of the ERSSQ values. The results allow one to conclude that both QTN and QUIT fit the Sips isotherm model (Fig. 3(a) and (b)). Nevertheless, in the case of QUIT the Freundlich model cannot be discarded, and further experiments with higher initial Al^{3+} concentrations may be performed in order to find which model is more suitable for the biopolymer. The Al^{3+} removal results suggest that QTN behaves according to both the physisorption (Freundlich model) and the chemisorptions (Langmuir model) mechanisms, at very low and higher concentrations, respectively, as shown in Fig. 3(a). The predominance of one or other mechanism over the removal process depends upon the adsorbate concentration, characterizing a Sips behavior adsorbent.

Table 1
Isothermal parameters for QTN and QUIT essays

		0.15 g QTN^{-1}	0.10 g QUIT L^{-1}
Langmuir	$q_{\max}$ (mg g^{-1})	18.05	–
	K_L (L mg^{-1})	17.31	–
	R^2	0.980	0.331
	ERRSQ	12.296	–
Freundlich	K_F (L g^{-1})	23.66	53.46
	n	2.451	0.796
	R^2	0.975	0.980
	ERSSQ	11.716	6.447
Slips	$q_{\max}$ (mg g^{-1})	18.28	23.57
	K_L (L mg^{-1})	11.46	4.1
	m	0.922	1.394
	R^2	0.975	0.984
	ERSSQ	9.020	0.526

According to the Sips model, the maximum removal capacity found for the biopolymers was 18.28 and 23.57 mg g^{-1} for chitin and chitosan, respectively (Table 1). These values were compared to those found in the literature for aluminum removal capacity. The values found in this work for QTN and QUIT are close to that found for aluminum removal by refused cast beach seaweed, which was 22.50 mg g^{-1} [28]. In others studies with QUIT, the results showed a maximum adsorption capacity of 45.50 mg g^{-1} [29] and 26.35 mg g^{-1} [30] at others initial Al^{3+} levels and conditions. These data indicate that both biopolymers show high adsorption capacities for Al^{3+}.

3.3. Kinetic experiments and removal from well water

Kinetic experiments and Al^{3+} removal were carried out with 0.80 g L^{-1} of QTN or QUIT and a well water sample containing 0.80 mg Al^{3+} L^{-1}. Fig. 4 shows the Al^{3+} variation (C_t/C_0) plotted against time of contact with both biopolymers. It is observed from the graphic that QUIT reduces Al^{3+} concentration faster than QTN. According to the data, in the first 20 min QTN removes 76% of Al^{+3} ions, whereas QUIT does it by around 90%. The WHO maximum Al concentration value for drinking water (0.20 mg L^{-1}) is reached in 20 min by QTN and in 10 min by QUIT. The complete removal of Al^{3+} for the analyzed water is reached by QTN in approximately 160 min against approximately 120 min taken by QUIT.

The pseudo-first-order, pseudo-second-order and intra-particle diffusion models were tested in this study in order to evidence the controlling mechanism

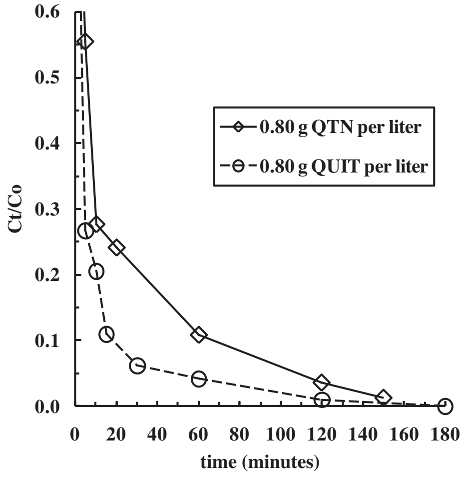

Fig. 4. Variation of concentration against for artesian well water (V = 200 mL, testing conditions 298 K and 200 rpm).

Table 2
Kinetic linearization's equations for well water and pseudo-second-order parameters

	$0.80\,g$ QTN L^{-1}	$0.80\,g$ QUIT L^{-1}
Pseudo-first-order		
Equation	$\mathrm{Log}(q_e - q_t) = -0.01t - 0.336$	$\mathrm{Log}(q_e - q_t) = -0.0009t - 0.0816$
R^2	0.972	0.921
Pseudo-second-order		
Equation	$t/q_1 = 0.960t + 0.082$	$t/q_1 = 1.109t + 1.538$
q_e ($mg\,g^{-1}$)	1.042	0.902
K_2 ($g\,mg^{-1}\,min^{-1}$)	0.181	0.800
h ($mg\,g^{-1}\,min^{-1}$)	0.197	0.650
ERSSQ	0.00939	0.00620
R^2	0.9999	0.9999
Intra-particle diffusion		
Equation	$q_t = 0.010t^{1/2} + 0.737$	$q_t = 0.002t^{1/2} + 0.818$
R^2	0.389	0.301

of Al^{+3} adsorption. The mathematical expressions containing the kinetics parameters, as well as the corresponding correlation coefficients resulting from the kinetic models linearization are provided in Table 2.

The results demonstrate that experimental data are best fitted by the pseudo-second-order model, confirming previous assumption that the controlling kinetic mechanism is chemical adsorption. This indicates that the adsorption of Al^{3+} by the biopolymers is dependent on the ions concentration in the adsorbent and also on the equilibrium concentration of adsorbate. The pseudo-second-order linearization for QTN and QUIT and the comparison between experimental data and pseudo-second-order model, as well of equilibrium adsorption capacity for QTN and QUIT biopolymers, are shown in Fig. 5.

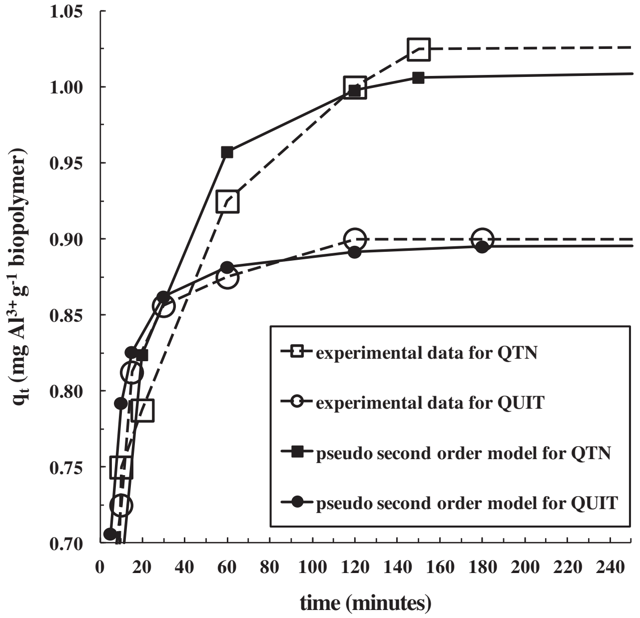

Fig. 5. Comparison between experimental data and pseudo-second-order models for $0.80\,g\,L^{-1}$ of QTN and QUIT in well water ($V = 200\,mL$, testing conditions 298 K and 200 rpm).

The results show that the pseudo-second-order models fit very well with the experimental data, showing chemisorptions properties that best fit for both biopolymers. The chemisorption processes suggested by the isothermal and kinetic results can be indicative of the formation of complexes between Al^{3+} and the coordinative sites of the biopolymers. Spectroscopic studies will be subsequently performed in order to confirm this hypothesis.

5. Conclusions

- CMC is not an efficient Al^{3+} removal agent from aqueous solutions at low Al^{3+} concentrations, as found in drinking water.
- The biopolymers showed a very good maximum adsorption capacity, with.18.28 $mg\,g^{-1}$ for QTN and 23.57 $mg\,g^{-1}$ for QUIT.
- The adsorption isotherms of Al^{3+} via QTN and QUIT biopolymers were best described by Sips model; however, the Freundlich model cannot be discarded for QUIT at the analyzed Al^{3+} concentrations.
- In kinetic experiments using well water ($[Al^{3+}] = 0.80\,mg\,L^{-1}$) QTN and QUIT removed completely Al^{3+} in 160 and 120 min, respectively. The WHO health safety level of Al^{3+} was reached in 20 min by QTN and 10 min by QUIT.

- The kinetic results suggest a pseudo-second-order model to fully describe the behavior of QTN and QUIT biopolymers to Al^{3+} removal in typical drinking water concentrations.
- Both QTN and QUIT biopolymers can be considered potential agents for effective Al^{3+} removal under the studied conditions and so, studies in continuous and in larger scale can be performed.

Acknowledgments

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Symbols

C_0	—	Al^{3+} initial concentration, $mg\,L^{-1}$
C_t	—	Al^{3+} concentration at time t, $mg\,L^{-1}$
C_e	—	Al^{3+} concentration at equilibrium, $mg\,L^{-1}$
q_e	—	equilibrium adsorption capacity, $mg\,g^{-1}$
q_t	—	adsorption capacity at time t, $mg\,g^{-1}$
q_{max}	—	maximum adsorption capacity, $mg\,g^{-1}$
K_L	—	Langmuir isotherm constant, $L\,mg^{-1}$
K_F	—	Freundlich isotherm constant, $L\,g^{-1}$
n	—	Freundlich exponent
K_s	—	Sips isotherm constant, $L\,mg^{-1}$
m	—	Sips exponent
K_1	—	Pseudo-first-order kinetic constant, min^{-1}
K_2	—	Pseudo-second-order kinetic constant, $g\,mg^{-1}\,min^{-1}$
h	—	initial adsorption rate, $mg\,g^{-1}\,min^{-1}$
K_d	—	intra-particle diffusion constant, $mg\,g^{-1}\,min^{-1/2}$
R^2	—	correlation coefficient
ERSSQ	—	sum of the squares of the errors

Abbreviations

WHO	—	World Health Organization

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