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Effect Of Solution Medium And Temperature On The Sorption Of Silver Ions By Impregnated Sorbents

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Abstract: The influence of the medium’s acidity and temperature on the sorption of silver ions by impregnated polymer sorbents was investigated. The sorption process was studied in the pH range of 1–12 using PAD400, PAD600, and FK-based sorbents modified with chelating ligands such as MAB, Dtz, and DEDTK. The results demonstrated that the sorption efficiency strongly depends on the acidity of the solution, with maximum sorption observed at pH = 3. The kinetics of silver ion sorption were analyzed using pseudo-first-order, pseudo-second-order, and intra-particle diffusion models. Experimental data showed that the sorption process reaches equilibrium within 60 minutes. Comparison of the kinetic parameters revealed that the pseudo-second-order model best describes the sorption process for all sorbents, with correlation coefficients ($R^2 > 0.99$) and calculated equilibrium capacities (Q_e,calc) consistent with experimental data (Q_e,obs). These findings indicate that the rate-limiting step of the sorption process is the surface chemical reaction between the functional groups of the sorbent and silver ions.

Keywords: Silver ion sorption; impregnated polymer sorbents; pH effect; pseudo-second-order kinetics; intra-particle diffusion; chelating ligands; adsorption equilibrium.

Introduction

The acidity of the medium plays a crucial role in the separation of metal ions by sorbents, as it determines the state and reactivity of the functional groups present on the sorbent surface. Furthermore, the speciation of metal ions in the solution also depends on the acidity of the medium, which allows the sorption process to be controlled by adjusting the pH conditions.

Results

The effect of pH on the sorption of silver ions by solid extractants was investigated within the pH range of 1–12 (Figure 1.). To control the pH of the solution, 0.1 M HNO_3 and NaOH solutions were used.

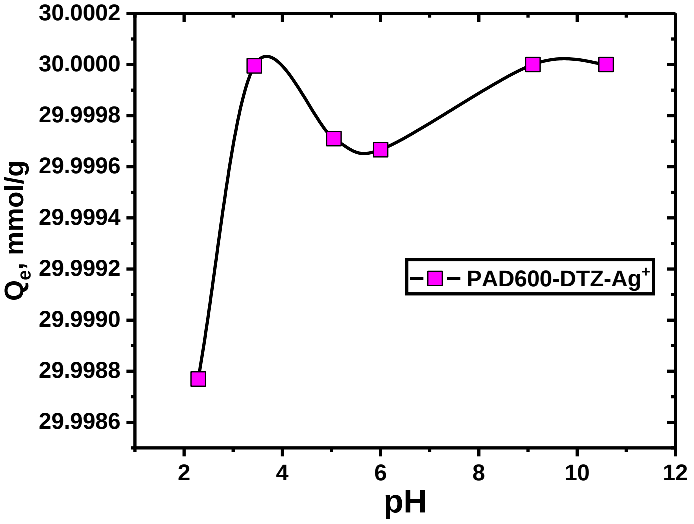

Figure 1. Dependence of silver ion sorption on the acidity of the medium (C⍰$_e$ = 50 mmol/L, m⍰$_{or}$b = 10 mg, t = 2 h, V = 30 mL).

The study of the effect of the medium on the sorption capacity of the PAD600-Dtz sorbent revealed that an increase in the pH value of the solution beyond a certain optimal point leads to a slight decrease in the sorption efficiency of silver ions. This phenomenon can be explained by the formation of hydrolyzed Ag(OH) species in the solution. The saturation of the free coordination capacity of the metal ion occurs due to interactions with water molecules or anions present in the medium. According to the obtained data, the pH corresponding to maximum silver ion sorption on the PAD600-DTZ solid extractant shifted toward a more acidic region. The highest sorption efficiency was observed at pH = 3.

Kinetics of Silver Ion Sorption.

The rate at which equilibrium is achieved during the sorption process is one of the most important characteristics of any sorbent, having both practical and theoretical significance. Based on kinetic data, it is possible to determine the time required to establish equilibrium in the sorbent–salt solution system. By applying diffusion and chemical kinetics principles to the theoretical interpretation of experimental data, it becomes possible to draw conclusions about the progression of the sorption process and to identify the rate-limiting steps that can be utilized to address certain practical issues in sorption applications.

The pseudo-first-order kinetic model is expressed by the following equation:

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2{,}303}t$$

The pseudo-second-order kinetic model is expressed by the following equation:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^{\,2}} + (\frac{1}{q_e})t$$

The kinetic curves describing the uptake of silver ions by the obtained solid extractants are presented in Figures 2–4. Kinetic studies showed that the sorption process proceeds relatively rapidly, and equilibrium between the silver ions in solution and the sorbent is established after approximately 60 minutes.

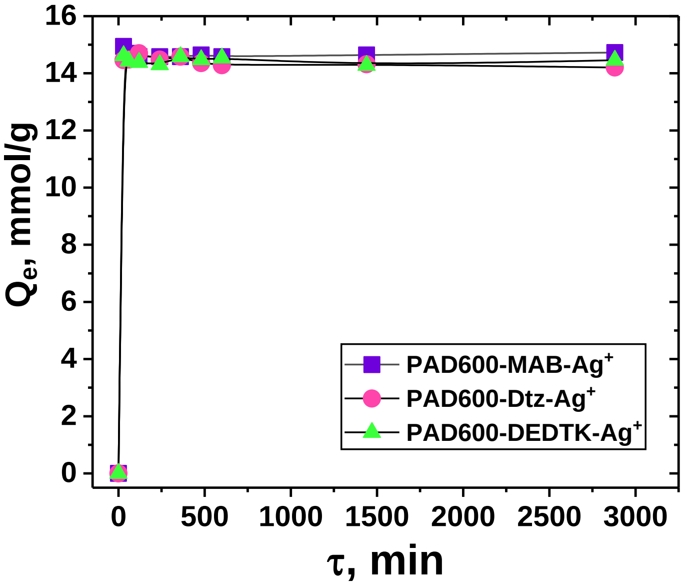

Figure 2. Time dependence of silver ion sorption on PAD600-based sorbents.

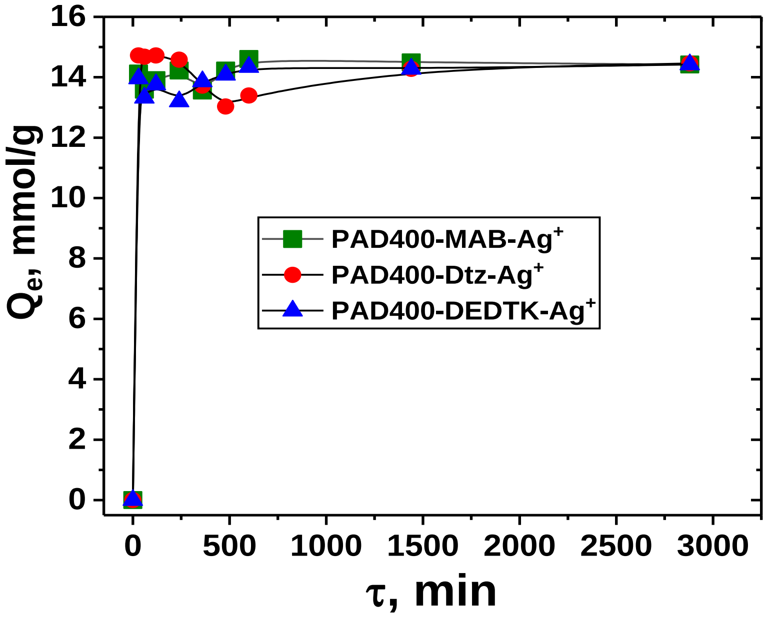

Figure 3. Time dependence of silver ion sorption on PAD400-based sorbents.

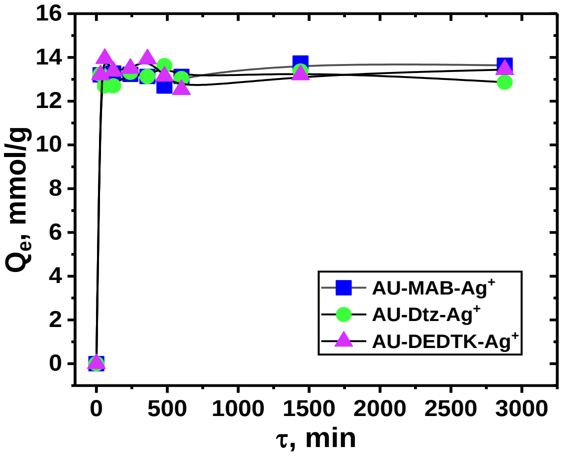

Figure 4. Time dependence of silver ion sorption on FK-based sorbents.

The study of sorption kinetics makes it possible to determine the contribution of the chemical stage of sorption for a given solution composition and temperature, as well as to establish the interaction mechanism between the modifier and the sorbent. For sorbents containing chelating groups, the chemical reaction between the sorbed ions and the functional groups of the sorbent may significantly contribute to the overall sorption kinetics [1–2]. The adsorption kinetic curves were analyzed using the pseudo-first-order and pseudo-second-order kinetic models, as well as the intra-particle diffusion equation.

In the case of intra-particle diffusion, the rate of the process depends on the particle size of the sorbent and the thickness of the formed adsorption layer. If the rate-limiting step is a chemical reaction, the degree of sorption is independent of these factors and depends solely on the ion concentration and temperature. When the kinetics of the process follow the pseudo-first-order model, sorption initially begins with diffusion. In the pseudo-second-order model, it is assumed that a chemical exchange reaction limits the sorption process. In this case, the reaction between the sorbate and the functional groups of the sorbent is considered to be a second-order reaction, where both interact in a 1:1 ratio [3]. Thus, the chemical interaction stage of heavy metal ions also contributes significantly to the overall rate of the sorption process.

The parameters of the pseudo-first-order kinetic model for the impregnated sorbents were determined from the linear relationship plotted in the coordinates $\ln(Q_e – Q\tau)$ versus τ (Figures 5–7).

To obtain the coefficients of the pseudo-second-order and intra-particle diffusion kinetic models, linear plots were constructed in the coordinates $\tau/Q\tau$ versus τ and Q_e versus $\tau^{1/2}$ (Figures 5–6).

The results of processing the integral kinetic curves for the sorption of Ag^+ ions on impregnated sorbents, based on various chemical kinetic models, are presented in Tables 1–2.

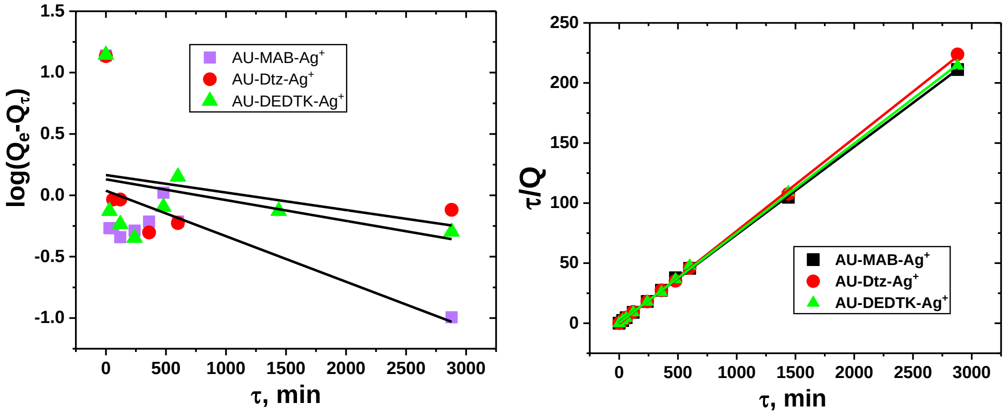

Figure 5. Dependence of silver ion sorption on FK-based solid extractants according to the pseudo-first-order model "$\ln(Q_e – Q\tau)$" (a) and the pseudo-second-order model "$Q/(1–Q)$" (b).

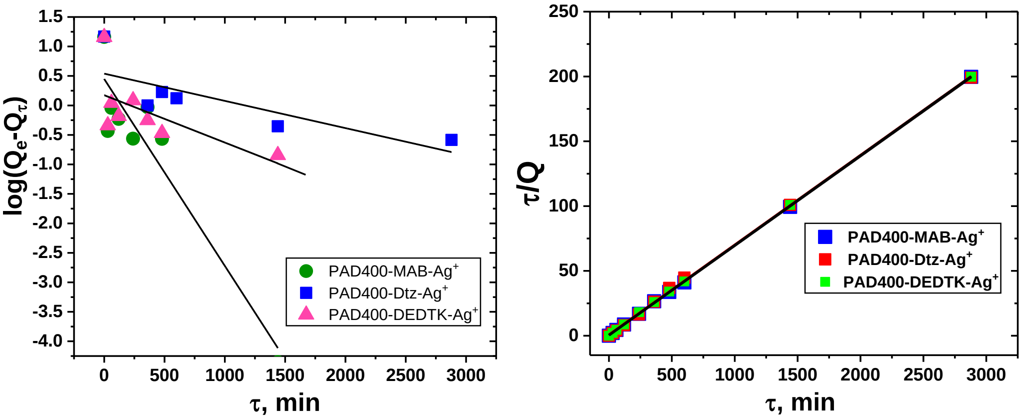

Figure 6. Dependence of silver ion sorption on PAD400-based solid extractants according to the pseudo-first-order model "$\ln(Q_e – Q\tau)$" (a) and the pseudo-second-order model "$Q/(1–Q)$" (b).

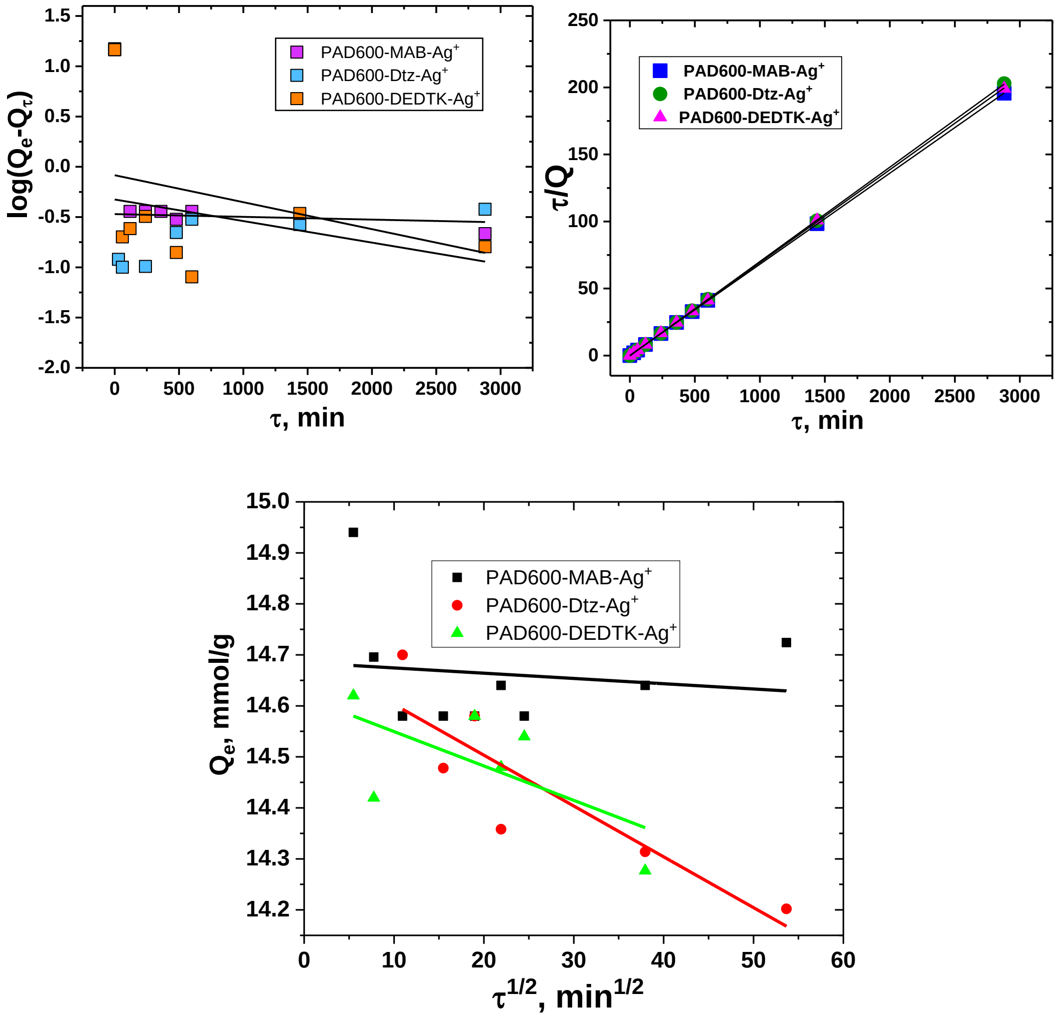

Figure 7. Dependence of silver ion sorption on PAD600-based solid extractants according to the pseudo-first-order model “ln(Q_e − Qτ)” (a), the pseudo-second-order model “Q/(1−Q)” (b), and the diffusion model “Q_e − $τ^{1/2}$” (c).

To establish the most appropriate model describing the sorption of Ag^+ ions on impregnated sorbents, the correlation coefficients (R^2) for each sorbent were compared. Analysis of the determination coefficient values presented in the tables showed that for the pseudo-first-order model, R^2 values ranged between 0.72 and 0.77, whereas for the pseudo-second-order model, R^2 values exceeded 0.99. In addition, for the first-order model, the experimentally obtained Q_e^{obs} values were slightly higher than the calculated Q_e^{calc} values, indicating the need to apply an alternative kinetic model. It should also be noted that, although the diffusion model provides an equilibrium adsorption value consistent with the experimental data, it exhibits a very low correlation coefficient (R^2).

It was observed that the data plotted in the coordinates of the pseudo-second-order equation provided the most accurate description for all the studied sorbents. A comparison of the results obtained using the pseudo-first-order, pseudo-second-order, and intra-particle diffusion models to characterize the kinetics of silver ion sorption showed that, in all cases, the pseudo-second-order model offered the best fit to the experimental data, exhibiting the highest correlation coefficients (R^2). Moreover, the calculated Q_e^{calc} values showed good agreement with the experimentally obtained Q_e^{obs} values.

Table 1. Kinetic parameters of silver ion sorption on PAD400-based sorbents.

Kinetic model	Parametr	PAD400-MAB	PAD400-Dtz	PAD400-DEDTK
Experimental results.	Q_e^{obs} (mmol/g)	14.49306	13.626	13.944
1 Order	Q_e^{calc} (mmol/g)	2.423818	1.2414	1.493826
	k_1 (min^{-1})	0.000606457	0.00061	0.000606457
	R^2	0.7707	0, 6435	0,4213
2 Order	Q_e^{calc} (mmol/g)	14.57726	14.47178	14.45087
	k_2 (g/ mmol./min)	0.008	0.005	0.007
	R^2	0.9996	0.9994	0.9999

Table 2. Kinetic parameters of silver ion sorption on PAD600-based sorbents.

Kinetic model	Parametr	PAD600-MAB	PAD600-Dtz	PAD600-DEDTK
Experimental results	Q_e^{obs} (mmol/g)	14.49306	14.58	14.62
1 Order	Q_e^{calc} (mmol/g)	2.929544	1.627047	1.672245
	k_1 (min^{-1})	0.000606457	0.000606457	0.000606457
	R^2	0.4252	0,0809	0,1195
2 Order	Q_e^{calc} (mmol/g)	14.72754	14.20455	14.41961
	k_2 (g/ mmol./min)	0.028	0.016	0.267
	R^2	1.00	1.00	1.00
Intra-particle diffusion	Q_e^{calc} (mmol/g)	14.618	14.703	14.58
	k_3 (mmol/g min$^{-0.5}$)	0.0002	0.001	0.0053
	R^2	0.0026	0.7741	0.2375

Table 3. Kinetic parameters of silver ion sorption on FK-based sorbents.

Kinetic model	Parametr	FK-MAB	FK-Dtz	FK-DEDTK
Experimental results	Q_e^{obs} (mmol/g)	13.7395	13.626	13.944
1 Order	Q_e^{calc} (mmol/g)	1.1301	1.2414	1.3344
	k_1 (min^{-1})	0.00061	0.00061	0.00061
	R^2	0.2893	0, 2301	0,0473
2 Order	Q_e^{calc} (mmol/g)	13.6986	13.4048	13.1062
	k_2 (g/ mmol./min)	0.006	0.017	0.031
	R^2	0.9998	0.9997	0.999

The pseudo-second-order model adequately describes the sorption kinetics of silver ions on the impregnated sorbents, indicating that the chemical reaction occurring on the sorbent surface is the rate-limiting step of the sorption process [4–6]. This equation accounts not only for the sorbate–sorbent interactions but also for the intermolecular interactions among the adsorbed species, thereby confirming the high adequacy and reliability of applying the second-order kinetic model.

Conclusion

The study demonstrated that the pH of the medium significantly affects the sorption efficiency of silver ions on impregnated polymer sorbents modified with chelating ligands. The maximum sorption capacity was achieved in the acidic region at pH = 3, indicating the key role of protonation–deprotonation equilibria of the sorbent's functional groups. Sorption equilibrium was established within 60 minutes, confirming the rapid interaction between Ag^+ ions and active sorption centers. Kinetic analysis revealed that the pseudo-second-order model best describes the sorption process for all investigated sorbents, with correlation coefficients ($R^2 > 0.99$) supporting high model adequacy. The calculated equilibrium capacities (Q_e,calc) were in strong agreement with experimental data (Q_e,obs), confirming the reliability of the proposed kinetic model. The results indicate that the rate-determining step is the surface chemical reaction rather than intra-particle diffusion. The effective performance of PAD400, PAD600, and FK-based sorbents demonstrates their potential use in selective silver ion removal.

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