

Preliminary Studies of Arsenic Adsorption Using Activated Carbons Synthesized from *Kagneckia Lanceolata* and *Passiflora Ligularis*

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Abstract—Arsenic is a chemical element that is found in freshwater in volcanic areas or because of activities like mining. It is a toxic element that can cause diseases like cancer, if taken in high amounts. The South of Peru is a volcanic region with intensive mining activity, and several governmental studies have reported arsenic in various rivers that supply water for the local population. Conventional arsenic removal techniques, such as reverse osmosis, can be very costly, making it necessary to study new, efficient and low-cost arsenic removal methods. The present study seeks to analyze the efficiency of activated carbons synthesized from *Passiflora Ligularis* (PL) shells and from a plant native to the Peruvian Andes, *Kagneckia lanceolata* (KL), at removing As from water by adsorption. The iodine number of these carbons were 964.39 and 962.13 mg/g, respectively. Images taken using scanning electron microscopy (SEM) corroborate how porous these new adsorbents are. In addition, it appears the adsorption equilibrium best fits the Langmuir model, with an adsorption capacity (Q max) of 5.07 mg/g for *Passiflora ligularis* (PL) and 4.51 mg/g for *Kagneckia lanceolata* (KL). The results show these new eco-friendly and low-cost adsorbents have great potential as As removing agents, and their performance can be improved with a treatment to give them As adsorption specificity.

Keywords—arsenic removal, adsorption, activated carbon, green adsorbents

I. INTRODUCTION

Arsenic is a harmful element to human health when ingested in large quantities through drinking water. Prolonged exposure to such elevated levels of arsenic in water can cause a wide range of health issues, leading to serious consequences such as miscarriage, or diseases like bronchitis, hypertension, skin cancer, kidney cancer and lung cancer [1].

Arsenic can enter the water supply through natural sources or as the result of human activities. Some of the common ways arsenic enter water include contamination via filtration through arsenic-rich soils, arsenic release from volcanic activity, and arsenic contamination from mining and industrial activities [2–4]. Due to the increase in industrial activities and the increasing use of groundwater as a source for drinking water, this problem has escalated to global importance. The World Health Organization (WHO) established a maximum limit of 0.01 mg/L of this element in drinking water, a value that is exceeded in many regions around the world [5].

There are several methods for the effective treatment of arsenic-contaminated water, the most common processes

being adsorption, coagulation-flocculation, membrane technologies, ion exchange, microfiltration, microbiological methods and electrochemical methods [6, 7]. While the methods mentioned are efficient, they have disadvantages associated with energy costs, chemical inputs and formation of toxic byproducts. This makes adsorption technology emerge as the most promising technique due to its cost-effectiveness and operational simplicity to remove potentially toxic elements [8].

The adsorption method was recently pointed out as the best option, where it is the adsorbent that plays a very important role in this process, the best adsorbent materials studied with interesting efficiency results are based on carbon, polymers, zeolites and organometallic structures [9].

Activated carbon can be used to adsorb this element, as it has a high affinity for certain contaminants such as heavy metals due to its large surface area and porous structure which allows it to trap and retain impurities [10]. However, its low arsenic adsorption capacity and the high cost of commercial activated carbon produced from conventional raw materials such as coconut shell and wood [11], have led various researchers to study new materials for the synthesis of activated carbon. Examples of successful materials include agricultural waste, such as rice polish, rice husk, activated carbon, industrial waste materials, beet pulp and corn straw [12–14].

Therefore, this article studies the potential of activated carbon produced from residues of a fruit native to the Andes, such as the granadilla peel *Passiflora Ligularis* (PL) and a plant native to the southern region of Peru such as the “Lloque” *Kagneckia Lanceolata* (KL), as arsenic adsorbents, since these materials are renewable, cost-effective and widely available, making them an environmental and economically attractive option.

There are few investigations that synthesize activated carbon from agro-industrial residues or native materials for the adsorption of As. In addition, this investigation is the first to report the adsorption capacity of these new raw materials native to and from the Andes, having as the main advantage a high adsorption of As with an unmodified activated carbon, as well as being a more financially viable and sustainable alternative to the use of conventional raw materials – having the potential to become, on the one hand, an excellent solution for waste management in the agri-food industry, and a way to achieve a circular economy with the use of PL waste;

as well as an option to improve the biodiversity of our country and a positive effect on the local economy due to the cultivation of KL as a raw material for activated carbon, becoming a source of income and job creation for native peoples.

The new adsorbents will also be characterized and the performance of As adsorption carried out by batches where the isotherms and adsorption kinetics were determined will be analyzed. Additionally, the article discusses the current state of research on this topic and provides insights into the future direction of this field. The specific objective of the research is to highlight the potential of activated carbon based on agro-industrial residues and native plants as a sustainable alternative to remove arsenic from water.

II. MATERIALS AND METHODS

A. Chemical Reagents

The chemicals used for the synthesis and characterization of activated carbon were 85% phosphoric acid (H_3PO_4), 0.5 M sodium hydroxide (NaOH), hydrochloric acid (HCl), 0.1 N sodium thiosulfate solution ($Na_2S_2O_3$), 0.1 N iodine solution, a starch solution and nitrogen gas (99.5%). The chemical reagent used for the adsorption tests was a stock solution (100 mg/L) prepared dissolving sodium arsenite ($NaAsO_2$) using deionized (DI) water. The Initial As concentration tests were prepared by gradually diluting the stock solution in DI for each test. 0.5 M (NaOH) and 0.1 M (HCl) were used to adjust the pH of the water samples. All the reagents used in this investigation were purchased from commercial sources (Merck and Sigma Aldrich) and are of pure analytical grade.

B. Preparation of Activated Carbons

The authors used their own procedure [15] to synthesize adsorbents from *Passiflora ligularis* (PL) and *Kageneckia lanceolata* (KL), with some modifications. The precursors were washed with distilled water to remove impurities and dried at room temperature for 48 hours. When obtaining the dry precursors, their size was reduced with a mill; then, the resulting particles went through a sieve, ASTM No. 50 mesh (0.297 mm), in order to obtain a uniform particle size before activation. The coals were prepared for activation following the optimal parameters for the (PL) found in a previous work by the authors [15]. Accordingly, they were activated in a ratio of 1 to 4 (precursor: H_3PO_4) at 60% for PL, in a shaker at 80 °C for 1 h, on the other hand the effect of the concentration of the acid at 60% and 40% for the precursor of (KL) for its activation. The impregnated precursors were carbonized in a tubular pyrolytic furnace (Model: OTF-1200X MTI Corporation) at a temperature of 500 °C for PL and 700 °C for KL with a heating rate of 10 °C·min⁻¹ for 120 min under a constant nitrogen flow rate (0.2 L min⁻¹) and allowed to cool under the same flow rate. Finally, the activated carbons were labeled as follows: PL, KL-40% and KL-60%, were washed with distilled water and a NaOH solution to eliminate residual acid and reach the pH of water. The washed samples were dried at 105 °C for 4 h in the oven and then weighed to determine their yield using the following equation:

$$R\% = \frac{(1-C_e/C_0)}{M} \quad (1)$$

where in the equation C_0 and C_e mg L⁻¹ represent the initial and equilibrium concentration of As in solution respectively.

C. Characterization of the Adsorbents

1) Adsorption capacity index

The authors used their own procedure [15] to synthesize adsorbents from *Passiflora ligularis* (PL) and *Kageneckia lanceolata* (KL), with some modifications. The precursors were washed with distilled water to remove impurities and dried at room temperature for 48 hours. When obtaining the dry precursors, their size was reduced with a mill; then, the resulting particles went through a sieve, ASTM No. 50 mesh (0.297 mm), in order to obtain a uniform particle size before activation. The coals were prepared for activation following the optimal parameters found in a previous work by the authors [15]. Accordingly, they were activated in a ratio of 1 to 4 (precursor: H_3PO_4) at 60% and 40% for PL and KL respectively, in a shaker at 80 °C for 1 h. The impregnated precursors were carbonized in a tubular pyrolytic furnace (Model: OTF-1200X MTI Corporation) at a temperature of 500 °C for PL and 700 °C for KL with a heating rate of 10 °C·min⁻¹ for 120 min under a constant nitrogen flow rate (0.2 L·min⁻¹) and allowed to cool under the same flow rate. Finally, the activated carbons were washed with distilled water and a NaOH solution to eliminate residual acid and reach the pH of water. The washed samples were dried at 105 °C for 4 h in the oven and then weighed to determine their yield using the following equation.

$$\frac{X}{M} = \frac{(N_1 \times 126.93 \times V_1) - \frac{V_1 + V_{HCl}}{V_F} \times (N_{Na_2S_2O_3} \times 126.93) \times V_{Na_2S_2O_3}}{M_C} \quad (2)$$

2) Physicochemical characterization of the adsorbent

A SEM (Thermo Scientific Scios Dual Beam FIB) scanning electron microscope was used to determine the morphological characteristics of the coals surface and its pore size distribution, coupled with an EDX Ray detector to determine the adsorbents elemental composition.

D. Adsorption Experiments

To determine the adsorption isotherms that better describe the arsenic adsorption capacity of the newly obtained activated carbons, a series of 7 flasks containing 50ml arsenic solutions at different concentrations of the metal from (0.1 mg/L to 80 mg/L) were prepared then shaken at 150 rpm. After 24 h of stirring, the adsorbent solutions were filtered to measure their concentrations. The total arsenic was determined using the guidelines described in APHA-SMWW 2006 – Method 3114-C using the atomic absorption spectrophotometry equipment brand Thermo Scientific ICAPQ model by hydride generation [16].

Afterwards, the determination of the adsorption capacity was carried out using the following equation:

$$q_e = V \cdot \frac{(C_0 - C_e)}{M} \quad (3)$$

where, is the equilibrium adsorption capacity (mg g⁻¹); (C_0 y C_e) mg L⁻¹ represent the initial and equilibrium concentration of As in the solution, respectively; M is the

mass of the adsorbent (g) and V is the volume of the solution used (L)

E. Adsorption Kinetics

The influence of time on the adsorption capacity was studied, the adsorption tests were performed in 250 mL flasks containing 50 mL of arsenic solution at a concentration of 10 mg/L adjusted to pH6, shaken at 150 rpm for 0, 30, 60, 90, 120 and 150 min. The mixtures were filtered and the filter solution was analyzed as described above. The adsorption capacity at each time interval was calculated and the kinetics determined.

Both the nonlinear adsorption kinetics and isotherm models were fitted to the experimental adsorption data using Origin 9.0 software as a nonlinear curve fitting tool.

III. RESULTS AND DISCUSSION

A. Characterization of Activated Carbons

1) Iodine number

One of the widely used parameters to measure the performance in adsorption capacity of carbon is the iodine number. This is an indirect determination of porosity and surface area. The resulting values for *Passiflora ligularis* and *Kageneckia lanceolata* are shown in Table 1. All iodine numbers were within the average range for activated carbon, which is from 500 to 1450 mg/g [17]. Table 1 shows the iodine number of these samples are higher than that of other agro-industrial residues. The higher the iodine number, the more significant the microcytic structures and the better porosity structure [18]. The values obtained indicate that the carbon samples have a microporous nature according to ASTM D 2866–94.

Table 1. Values of the iodine number for activated carbons

Material	% H ₃ PO ₄	T (°C)	Iodine number (mg/g)	Ref.
Commercial activated carbon	-	-	738.62	[18]
Tea waste <i>Camellia sinensis</i>	-	300	593.41	[18]
Cardoon waste <i>Cynara Cardunculus</i>	-	-	418	[19]
Cardoon waste <i>Cynara Cardunculus</i>	5	-	426	[19]
Coal Bottom Ash	-	-	234.95	[20]
Shell of granadilla <i>Passiflora ligularis</i>	60	500	978.55	[15]
<i>Kageneckia lanceolata</i>	40	700	905.82	[15]
Shell of granadilla <i>Passiflora ligularis</i>	60	500	964.39	Present work
<i>Kageneckia lanceolata</i>	60	700	948.17	Present work
<i>Kageneckia lanceolata</i>	40	700	962.13	Present work

2) Scanning Electron Microscopy (SEM)

The surface morphology of the activated carbons was studied by scanning electron microscopy (SEM) with an EDS X-ray detector to identify elements present in the sample. In Fig. 1, we can see the microphotographs that were taken at two different magnifications. Fig. 1 (a), (c) and (e) at 3000X and Fig. 1 (b), (d) and (f) at 80000X magnification. An irregular morphology is observed for the PL, KL-60% coals, and KL-40%.

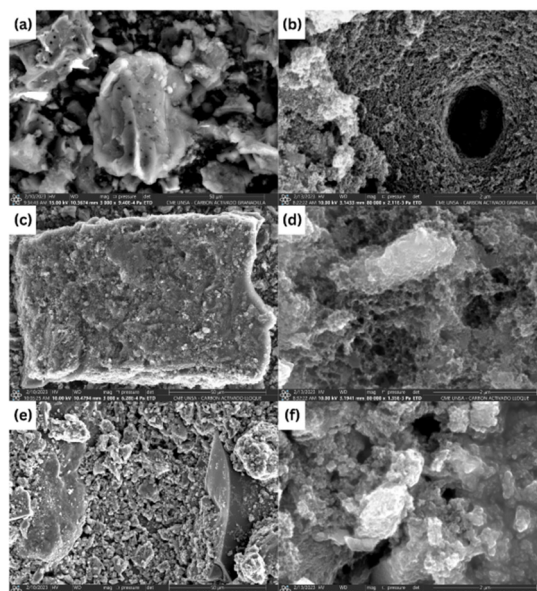


Fig. 1. SEM image (a) and (b) PL, (c) and (d) KL -60% H₃PO₄, (e) and (f) KL -40% H₃PO₄.

In the three coals, diatoms of different shapes, round, ovoid and elongated, are observed. The carbonization of PL resulted in a porous carbon with pore diameters ranging between 0.83–2.55 μm , while for KL-60% between 0.63–1.79 μm and for KL-40% between 0.99–3.6 μm . There is a difference in structure between PL and both KL samples, the latter presenting a more homogeneous porosity.

SEM images in general indicate granular surfaces with different pore sizes due to the liberation of volatile organic compounds [15], The pore structure is one of the factors that determines the performance and quality of the adsorbent [21] and this in turn depends on the raw material used, since the chemical composition varies according to the precursor, where the precursors rich in carbon are of main interest for the synthesis of high-yield adsorbents [22–24].

As seen in Fig. 1(c), (e) the pore diameters become almost undetectable and it is only at a higher magnification, Fig. 1(d), (f), where they are best visualized, noticing a random mixture of coarse and fine grains without a particular pattern. Fig. 1(f) shows pores up to 3.6 μm , with a mean of 2.7 μm . These large pores widened due to activation conditions.

The chemical composition of the surface of the activated carbons obtained is presented in Table 2 and indicates the majority presence of (Carbon, Oxygen, Sodium, Silicon and Phosphorus). It is observed that there was a small loss of C (carbon), this due to a greater use of phosphoric acid that promotes the gasification of coal and will increase the loss of total carbon mass [25].

Table 2. Elemental composition of the activated carbon samples obtained

Sample	Code	C (%w)	O (%w)	Na (%w)	Si (%w)	P (%w)
Shell of granadilla (<i>Passiflora ligularis</i>)	PL	26.22	71.89	0.43	0.03	1.43
Lloque (<i>Kageneckia lanceolata</i>)	KL-60	26.57	72.18	0.25	0.02	0.98
Lloque (<i>Kageneckia lanceolata</i>)	KL-40	26.81	72.22	0.45	0.13	0.38

B. Adsorption Equilibrium

The adsorption isotherms describe the balance between the concentrations of a material in the aqueous phase and on the surfaces of the adsorbent particles. The mathematical models of Langmuir and Freundlich were used to describe the adsorption equilibrium in this investigation.

1) Langmuir isotherm

The Langmuir Adsorption model assumes that (a) the surface is homogeneous, (b) it has a specific number of sites where a molecule can be adsorbed and so, when all sites are occupied, it is not possible for adsorption to continue as the system has become saturated, (c) the heat of adsorption is independent of the degree of coverage and (d) all sites are homogeneous and the energy of the adsorbed molecules is independent of the presence of other molecules [26].

The Langmuir isotherm can be described by the following equation:

$$q_e = \frac{K_L q_{max} C_e}{1 + K_L C_e} \quad (4)$$

where C_e is the equilibrium concentration of arsenic in the solution ($\text{mg} \cdot \text{L}^{-1}$), q_e is the amount of arsenic adsorbed by the activated carbon ($\text{mg} \cdot \text{g}^{-1}$), q_{max} is the maximum adsorption capacity of the activated carbon monolayer ($\text{mg} \cdot \text{g}^{-1}$), and K_L is the Langmuir adsorption constant related to energy adsorption. ($\text{L} \cdot \text{mg}^{-1}$).

2) Freundlich isotherm

The Freundlich model was used to analyze the adsorption equilibrium of arsenic on the surface of activated carbon.

This model is applied to sorption processes on heterogeneous surfaces and reversible adsorption and admits multilayer adsorption.

The Freundlich isotherm is expressed according to Eq. (5):

$$q_e = K_F C_e^{1/n} \quad (5)$$

where q_e is the amount of arsenic adsorbed per unit weight of adsorbent at equilibrium ($\text{mg} \cdot \text{g}^{-1}$), C_e is the equilibrium concentration of arsenic in solutions ($\text{mg} \cdot \text{L}^{-1}$), $y(\text{L} \cdot \text{g}^{-1})$, and K_F and n are the Freundlich sorption isotherm constants, related to the adsorption capacity of the adsorbent and the adsorption intensity, respectively.

Adsorption isotherms are important because, through them, the interactions between the adsorbate and the active sites present on the adsorbent surface are understood. Both help predict the adsorption capacities of the adsorbent.

The Langmuir theory was better adjusted according to the highest correlation coefficients obtained (Table 3) ($R^2 = 0.96874$ and $R^2 = 0.98951$) with a maximum adsorption capacity of 5.07 and 4.51 mg/g for PL and KL respectively, which describes that the adsorption is monolayer and a relatively homogeneous adsorbent surface of As [27]. The isotherm data confirm the higher adsorption of As for PL, reaffirming the results obtained in the Iodine Index.

Table 3 presents the parameter values of the Langmuir and Freundlich adsorption isotherms, where the corresponding coefficients of determination (R^2) of the Langmuir model for samples made from granadilla shells (PL) and lloque (KL) are greater than those of the Freundlich model, showing the

Langmuir model better represents the adsorption mechanism for both adsorbents. These results can also be seen in Fig. 2(a) Granadilla shell adsorption isotherm (PL), (b) Lloque adsorption isotherm (KL).

Table 3. Parameters of Langmuir and Freundlich adsorption

Adsorption Isotherms	Type of adsorbent	
	PL (a)	KL (b)
Langmuir model		
q_{max} (mg/g)	5.076 ± 0.881	4.509 ± 0.564
K_L (L/mg)	0.026 ± 0.009	0.018 ± 0.004
R^2	0.968	0.989
Freundlich model		
K_F	0.263 ± 0.112	0.157 ± 0.048
N	1.666 ± 0.309	1.519 ± 0.183
R^2	0.932	0.970

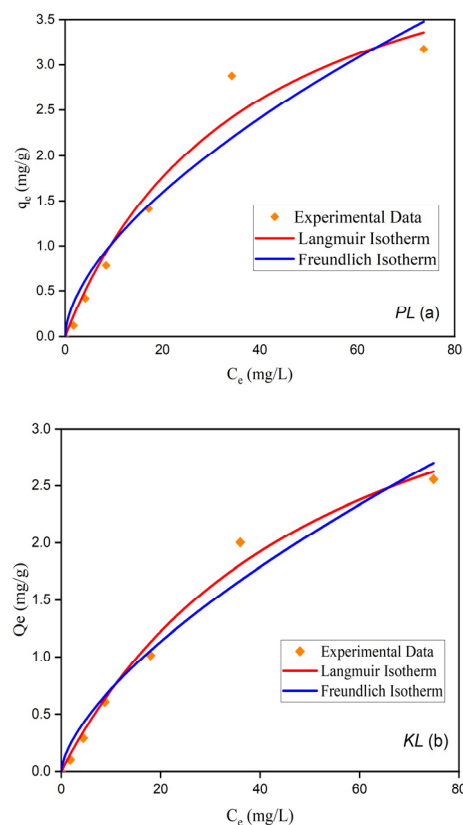


Fig. 2. Langmuir and Freundlich isotherms fitted to the experimental data of activated carbons PL (a) and KL (b).

Langmuir's equation considers the sorbent has a limited number of adsorption sites, which the adsorption energies are the same and so, there are no interactions between molecules [15].

The maximum adsorption capacities of the PL and KL precursors exceed those reported in the literature (Table 4). Sattar *et al.* (2019) [28] reported a maximum adsorption capacity of 5.01 (mg g^{-1}) as an adsorbent for peanut shells, which is close to the results obtained in this work, this can be attributed to the particular chemical composition of the precursors where it is used. It maximizes the availability of active sites on the surface due to the greater presence of fixed carbon. The authors Pączkowski *et al.* (2021) [29] carried out the compositional analysis of the peanut shell, the results indicated that it is made up of 46.05% carbon, of equal similarity to the Shell of passion fruit *Passiflora ligularis* with a carbon content of 47.35% [30]. The presence of carbon constitutes the presence of cellulose, lignocellulose,

hemicellulose, lignin and proteins that assign certain characteristics and properties to the precursor [31, 32].

Table 4. Comparison of arsenic adsorption capacity (q_{max}) with other lignocellulosic adsorbents

Adsorbent	Maximum Adsorption capacity ($mg\ g^{-1}$)	pH	Ref.
Acid treated eucalyptus bark	0.944	6	[33]
Peanut shells	4.75	7.2	[28]
	5.01	6.2	
Guava leaves	1.05	6	[34]
Mango peel	1.25	6.5	
Sugar cane bagasse	1.35	7.5	
Guava seeds	4	6	[35]
Soya	0.005	2	[36]
Rice peel	0.225	8	[37]
Pine bark	3.5	4	[38]
Shell of granadilla <i>Passiflora ligularis</i>	5.07	6	Present work
<i>Kageneckia lanceolata</i>	4.51	6	Present work

C. Adsorption Kinetics

The adsorption kinetics of *Passiflora ligularis* (PL) was carried out in this research because it presents the best results of maximum adsorption capacity (q_m). The kinetic model will allow determining the type of adsorption mechanism and the effectiveness of the adsorbent, evaluating the pseudo first and second order kinetics, since they are kinetic models that are commonly used for adsorption kinetics simulations.

1) Pseudo First Order (PFO)

According to this model, it is assumed that the adsorption rate in relation to time is proportional to the difference between the equilibrium adsorption capacity (q_e) and the amount adsorbed at each time (q_t) [39], described by the following equation:

$$q_t = q_e(1 - e^{-k_1 t}) \quad (6)$$

where: (q_t) is the adsorption capacity at any time ($mg \cdot g^{-1}$), (q_e) is the equilibrium adsorption capacity ($mg \cdot g^{-1}$), (k_1) is the first order rate constant (min^{-1}) and t is the time spent on the adsorption test (min).

2) Pseudo Second Order (PSO)

The pseudo second order model describes the adsorption process described predominantly as chemisorption [40], represented by the following non-linear equation:

$$q_t = \frac{q_e^2 k_2 t}{1 + q_e k_2 t} \quad (7)$$

where k_2 is the pseudo second order rate constant ($g \cdot mg^{-1} \cdot min^{-1}$), and t is adsorption time (min).

The experimental data were correlated with the PFO and PSO as kinetic models to determine the kinetic parameters of arsenic adsorption (Fig. 3), which are represented in Table 5.

The determination coefficient (R^2) (0.99647) obtained in the kinetic model suggest that the adsorption with PL could be described by the Pseudo First Order (PFO) or Lagergren kinetic model, this model describes a physical adsorption, consequently the control mechanism of the adsorption process is based on the mass transfer of the adsorbate to the surface of the adsorbent [41]

Table 5. Parameters of the kinetic models

Kinetic Models	Type of adsorbent
	PL
Pseudo First Order (PFO)	
q_e (mg/g)	0.46028 ± 0.00503
k_1 (min^{-1})	0.08629 ± 0.00539
R^2	0.99647
χ^2	1.02×10^{-4}
Pseudo Second Order (PSO)	
q_e (mg/g)	0.50019 ± 9.4266
k_2 (min^{-1})	0.2746 ± 0.0042
R^2	0.96426
χ^2	2.51×10^{-4}

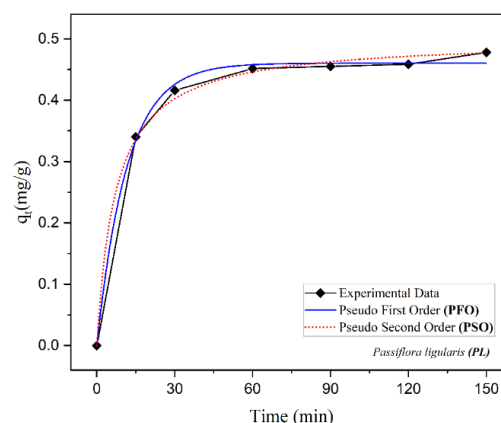


Fig. 3. Total arsenic adsorption kinetics by *Passiflora ligularis* (PL).

IV. CONCLUSION

The present study considers removing As using new adsorbents made from granadilla shells *Passiflora ligularis* and *Kageneckia lanceolata*, a plant native to the Peruvian Andes. A SEM analysis of the synthesized activated carbons show they have a satisfactory surface area and are sufficiently porous. The iodine number was calculated accordingly and the results were 964.39 mg/g PL and 962.13 mg/g KL, showing a strong adsorption performance, and that it is mostly microporous.

The experimental results showed the Langmuir isotherm fits the adsorption capacities of both activated carbons better. The maximum adsorption capacity of PL was 5.07 mg/g and of KL, 4.51 mg/g. Likewise, the kinetics of PL adsorption is described by means of the kinetic model of Pseudo First Order (PFO). The aforementioned data confirm activated carbons made from these particular raw materials are a good inexpensive alternative to remove arsenic from water sources.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

J.S: Analyzed the conceptualization, methodology, investigation, writing - original draft. P.P: Performed the methodology, investigation, data curation, writing - original draft. R.A: Investigation, formal analysis, visualization, writing - original draft. W.V: Writing - original draft, investigation, J.M: Validation, supervision, writing - review & editing, resources. All authors had approved the final version.

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