



## Removal of paraquat from drinking water by activated carbon prepared from waste wood

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### ABSTRACT

In this study, activated carbon was produced from the chips of two wood species, namely Ayous (*Triplochiton schleroxylon*) and Sapelli (*Entandrophragma cylindricum*) and characterized for its texture by scanning electron microscopy, granulometry using a master sizer, and the structure by determining the iodine and methylene blue indices. The obtained carbon was used to study the adsorption of paraquat in aqueous solution. Results from the characterization of carbon revealed that it consists essentially of micropores and mesopores, a tubular structure, and a fine coating of very small granules on the surface (Ayous) and elongated fibers (Sapelli). The adsorption kinetics of paraquat in solution followed a maximum exponential growth model with three stages: the first, which corresponds to an increase in adsorption of the molecule on sites located on the outer surface of the carbon, the second phase, which corresponds to the diffusion of the molecules on the internal pores, and the third constant phase corresponding to the adsorption equilibrium. The equilibrium time obtained was 10 min for Ayous with 87% of paraquat adsorbed and 20 min for Sapelli with 90% of paraquat adsorbed. The biosorption kinetics of paraquat in the solution followed a pseudo-first order and intra-particle diffusion model. Results from the influence of paraquat concentration and mass of carbon showed that, an increase in concentration and mass of the adsorbent impel respectively in the amount of the paraquat molecule adsorbed. Change of solution pH does not affect the amount of paraquat adsorbed whatever the type of carbon. The adsorbed amounts of paraquat decreased (from 88 to 60 mg g<sup>-1</sup>) as the ionic strength increased from 5 × 10<sup>-3</sup> to 5 × 10<sup>-2</sup> M. This study shows that activated carbon products obtained from Sapelli and Ayous wood chips, which are wastes released into the environment, can be used as low-cost biosorbents for the removal of organic micropollutants in drinking water.

*Keywords:* Chips of wood; Activated carbon; Paraquat; Adsorption kinetics; Drinking water

### 1. Introduction

Methods used to obtain drinkable water sometimes include filtration with activated carbon to remove

organic micropollutants such as pesticides [1]. The physicochemical characteristics of carbon can influence the adsorption of these organic pollutants. The adsorption capacity of organic compounds is proportional to the micro-porous surface of the adsorbent [2]. It is known that adsorption using low-cost adsorbents is

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an effective and economic method for water treatment. Many non-conventional adsorbent materials have been proposed and studied for their ability to remove toxic azo dye in water by using hen feather [3–11], for the removal and recovery of toxic halogen, hazardous coloring agent by using bottom ash [12,13], hazardous dye by using waste material of de-oiled soya [14] and for the removal of phenol and pesticides in water [15,16]. A research on waste materials can yield useful adsorbents, which have numerous uses and applications in water treatment. Chips and sawdust are the wastes from wood processing. A study in 28 sawmills in the city of Ngaoundere, Cameroon revealed a very high use of lignocellulosic wood species (260t/month) such as Ayous, Sapelli and Iroko. The transformation of these species generates about 266.6 kg of waste per month [17]. Therefore, any attempt to reuse this waste will be useful for the country.

The expansion of agricultural activities in recent years has created an impact on ecosystems equilibrium due to the intense use of pesticides. Due to their high solubility ( $620\text{ g L}^{-1}$ ) [18] and ease to seep into soil, these molecules are found in ground and surface waters including those waters which are intended for human consumption.

Ranked number two worldwide, paraquat or 1, 1-dimethyl-4,4- bipyridiniumdichloride according to IUPAC and commonly called Gramoxone on the local market is the most controversial herbicide, because it's the most toxic to humans when ingested deliberately or accidentally [19]. This is however one of the best-selling herbicide in the world because it is present in more than 120 countries and in 19 African countries, including Nigeria, Cameroon and Burkina Faso.

In Cameroon, agriculture is practiced by about 80% of the active population. However, this activity is accompanied by an increase and sometimes uncontrolled use of chemical products. In nearly ten years, the sale of pesticides in Cameroon has increased more than ten folds. Indeed, the destruction of the microflora and fauna of a cultivated soil contaminated by pesticides is estimated at 90%. About 60% of the dose applied to the plant directly touches the ground and after infiltration and run-off, are found in surface and ground waters intended for human consumption [20]. The European Union has set standards with regard to the acceptable concentration for each pesticide (paraquat included) in water for consumption at  $0.1\text{ }\mu\text{g L}^{-1}$  and  $1\text{--}3\text{ }\mu\text{g L}^{-1}$  for surface waters [21]. Reducing the amount of paraquat in these waters is a major concern.

Conventional methods used for potable water treatment have proven ineffective for the removal of these compounds. Thus several methods for their

elimination are being developed over the years. In particular, chemical (advanced oxidation, photolysis) and physical (nanofiltration, reverse osmosis) techniques have shown very good results, but their application is still very expensive for under-developed and developing countries [22]. Adsorption using activated carbon, clay, zeolite, and alumina has proven to be very effective for the removal of these compounds in water [23]. Activated carbon is one of the best adsorbents used in the adsorption of organic micropollutants due to its high adsorption capacity [24]. About 50% of industrially available activated carbons are derived from precursors of lignocellulosic origin [2,16] which is very expensive. It is for this reason that we are moving towards low-cost materials which are readily available such as chips and sawdust, which can be exploited in the manufacture of activated carbon.

In order to valorize these wastes which are often underutilized in Cameroon, activated carbons were prepared from the wood chips. The former was used to test their capacity to eliminate paraquat in solution. The aim of the present work was thus: To produce and characterize activated carbons from chips of two different wood species notably Ayous (*Triplochiton-schleroxylon*) and Sapelli (*Entandrophragmacylindricum*), and to study the influence of physicochemical parameters (concentration, weight, ionic strength, and pH) on the adsorption kinetics of paraquat on the activated carbon in aqueous solution.

## 2. Materials and methods

### 2.1. Sampling and preparation of Ayous (*T. schleroxylon*) and sapelli (*E. cylindricum*)

Ayous tree is present in several African countries. In Cameroon, it is mostly found in the Eastern forests. This wood is white, soft and porous. It has low durability and so is widely used for light carpentry. It consists of approximately 98% of cellulose, hemicellulose and lignin. The remaining 2% is made up of extractable (tannins, pectins, polymers with low degree of polymerization) and minerals ( $\text{Ca}^{2+}$ ,  $\text{Na}^+$ ,  $\text{Cl}^-$ , and  $\text{SO}_4^{2-}$ ) [25,26].

Sapelli is a wood with a red color and it is highly demanded for their durability. It is very strong and therefore widely used for various purposes and in particular in the manufacture of small bridges. This wood contains plant constituents such as cellulose, hemicellulose, lignin, and pectin at a very high rate of about 98%.

Sampling of these two types of chips was carried out in sawmills in Ngaoundere city and 30 kg of each

type was collected. The sampled wood chips were directly taken out of the wood plainer in order to avoid mixing them with other sample types. After wood chips sampling, parameters such as water and ash content were determined by AFNOR method (1981).

The Wood chips (*Ayous* and *Sapelli*) utilized in this work was dried at 105°C in the oven for 24 h. It was then placed in the furnace at 350°C for 1 h 30 min. After this step, the carbon produced was cooled, crushed and sieved to obtain a powder with ( $\varnothing < 50 \mu\text{m}$ ).

The preparation of activated carbon was adopted from the method of [27]. To prepare the activated carbon adsorbent, a known amount of previously carbonized material (ANA and SNA) was mixed with  $\text{H}_3\text{PO}_4$  (85%) to obtain a pulp, and then placed in a furnace at 450°C for 1 h 30 min. After Cooling, the material was soaked with nitric acid (10 mL of nitric acid/g of carbon). Subsequently, the slurry was filtered in the Wattman paper No. 1, and its solid phase was thoroughly washed with water, until the filtrate reached the pH of distilled water. Subsequently, the material was dried at 105°C in an oven for 8 h. The activated carbon produced was assigned to AA and SA.

## 2.2. Characterization of activated carbons

The textural and surface chemical properties of the activated carbons were obtained by the following techniques:

The morphological characterization was done by scanning electron microscopy (SEM). The microscopic analysis of carbon was performed using a scanning electron microscope Hitachi S-4500. The particle size distributions were measured using a Malvern Master Sizer (Master sizer 2000).

Infrared analysis (1 mg carbon mixed with 250 mg KBr) was carried out with a Bruker alpha spectrometer in transmission mode (200 scans collected at  $2 \text{ cm}^{-1}$  resolution in the  $4,000\text{--}400 \text{ cm}^{-1}$  range).

The values of the iodine index and the methylene blue index were obtained according to the method of [16] and [28], respectively.

## 2.3. Adsorption studies

### 2.3.1. Preparation of paraquat synthetic solution

Pure paraquat molecule (99%) was obtained from Riedel de Haen (Germany). The structure of this molecule is as presented below (Fig. 1):

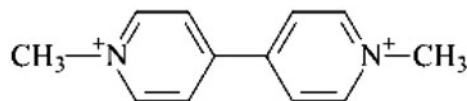


Fig. 1. Paraquat molecular structure.

The stock solution of paraquat of concentration 1 g/L was prepared and used for the adsorption study. Successive dilutions of 100, 200, 300, and 400  $\text{mg L}^{-1}$  were prepared from this stock solution.

### 2.3.2. Batch adsorption kinetics

The adsorption of this molecule (paraquat) was carried out in well-closed flasks of 120 mL into which a volume of 30 mL of solution was introduced. The whole was shaken in a water bath at a temperature of  $25 \pm 1^\circ\text{C}$  and a speed of 35 strokes  $\text{min}^{-1}$ . Studies were conducted at time intervals of 10 min over a period of 60 min. Sodium hydroxide (Merck) or hydrochloric acid (Merck) and potassium chloride (Merck) were used to vary the pH and the ionic strength, respectively. The mixture was filtered using a filter paper with a porosity of  $0.45 \mu\text{m}$ . The concentration of paraquat in the solution was analyzed using a UV/Vis-6405 spectrophotometer (JENWAY, Shimadzu UV-6405) by monitoring the absorbance changes at a wavelength of maximum absorbance (634 nm). Blank tests were also run to estimate the amount of paraquat adsorbed on walls of flasks tubes. The amount of adsorbed paraquat was calculated from the difference between the total added paraquat concentration and the supernatant concentration.

The influence of parameters such as the concentration of paraquat, the mass of activated carbon introduced, the pH, and ionic strength were also studied.

In order to investigate the adsorption kinetics of paraquat, the intra-particle diffusion model, pseudo-first order model and pseudo-second order model were carried out in this study.

## 3. Results and discussion

### 3.1. Raw materials analysis

The results of the proximate analysis of the sampled biomass are presented in the Table 1 below.

It appears from immediate analysis that the moisture contents of both wood chips are three-fold higher than the standards, which provides a value of 5%. These high values may be explained by the exposure of wood to rains, since sampling was done during the

Table 1  
Sampled biomass analyses

Biomass	Humidity (%)	Ash (%)
Ayous wood chips	14.53 ± 0.6	10.09 ± 0.01
Sapelli wood chips	16.48 ± 0.2	10.42 ± 0.05

rainy season. However, other studies by [29] also revealed water content greater than 5% for coconut shells and peanut shells.

The analyses also presented an average ash content. We can therefore conclude that the biomasses chosen are suitable to be used for obtaining good activated carbon because they contain low ash.

### 3.2. Carbon characterization

Fig. 2 presented the MEB of activated carbon from Ayous wood chips (a)–(d) and Sapelli wood chips (e)–(h).

Fig. 2(a)–(h) above present SEM images of AA and SA carbon obtained at different magnification. We can notice that both carbons have a porous and tubular structure (Fig. 2(a), (c)–(f)). This structure is very compact for both types of carbon, but it is slightly more pronounced in AA. This observation of the compact structure of carbon has already been reported by some authors for different types of carbon including the fruit hulks of Brazilian pine [30], Hulks of *Jatropha curcas* [31], and Aqai stem [32]. The surface of the material is heterogenous and presents pores of varying sizes (macropores, mesopores and micropores) and polygons of irregular shapes (Fig. 2(a)–(c), (e) and (h)). An enlargement of Fig. 2(a) of AA carbon shows the presence of deposits of masses of granules of very small and medium size at its surface (Fig. 2(b)). Similarly an enlargement of Fig. 2(f) of SA carbon reveals the presence of deposits of interspersed fibers on its surface (Fig. 2(g)). Therefore, treatment of materials with phosphoric acid ( $H_3PO_4$ ) allowed the generation

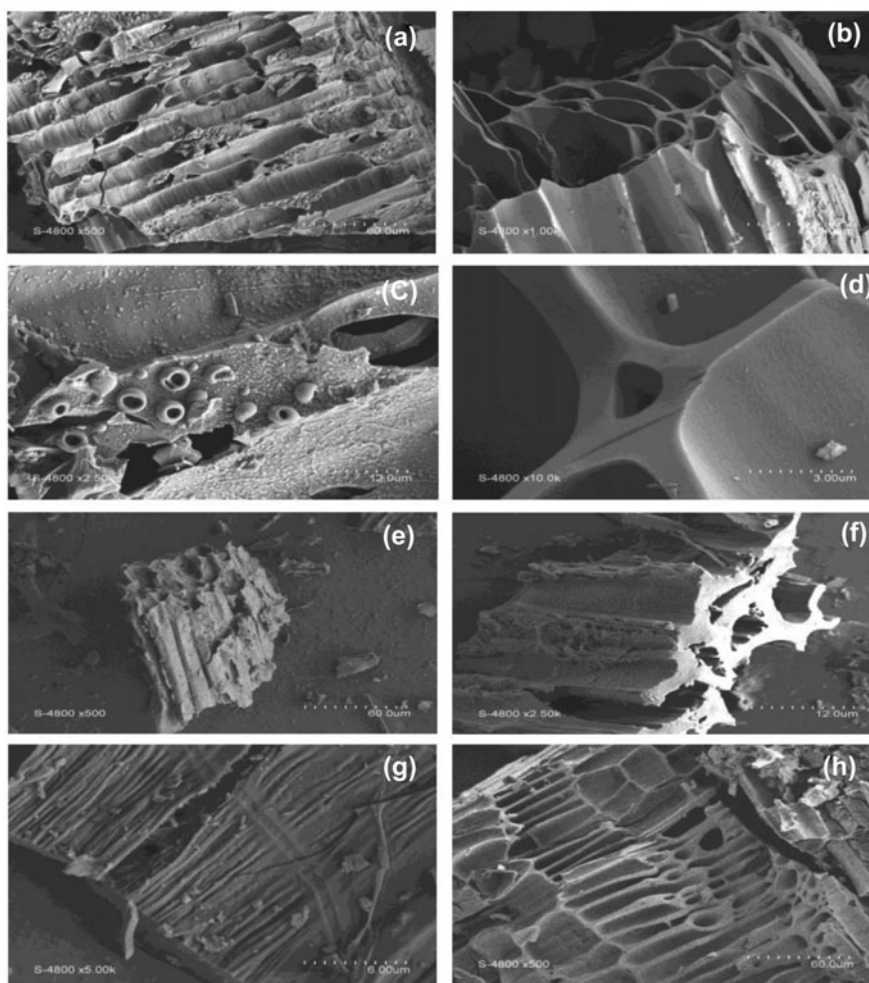


Fig. 2. MEB of activated carbon from Ayous wood chips (a)–(d) and Sapelli wood chips (e)–(h).

of macropores ( $\varnothing > 50$  nm) on the surface in small numbers, and mesopores ( $\varnothing$  between 2 and 50 nm) and micropores ( $\varnothing < 2$  nm) in large numbers. This suggests a very high adsorption capacity of these materials.

The size distribution of the charcoal giving by laser diffraction particle sizer analyzer is reported in Fig. 3.

Fig. 3(a) shows that 77% of the particles have sizes between 10 and 100  $\mu\text{m}$  with a mean size of 40  $\mu\text{m}$ . Fig. 3(b) shows that 84% have sizes between 10 and 100  $\mu\text{m}$  with a mean size of 50  $\mu\text{m}$ . Fig. 3(c) show that 3.6% of the particles have sizes lower than or equal to 5  $\mu\text{m}$ ; 79% of the particles have sizes between 5 and 100  $\mu\text{m}$ , and 16.6% have size greater than 100  $\mu\text{m}$ . Fig. 3(d) shows that 29% of the particles have sizes lower than or equal to 10  $\mu\text{m}$ ; 56% of the particles have sizes between 10 and 50  $\mu\text{m}$ , 12.75% of the particles have sizes between 50 and 100  $\mu\text{m}$ , and 2.25% have sizes greater than 100  $\mu\text{m}$ .

The spectrum (Fig. 4) exhibits the main infrared bands from non activated and activated carbons of Ayous (a), (b) and Sapelli (c), (d).

The surface groups of coals studied, were determined by infrared spectroscopy and Fourier transmission shown in Fig. 4. It is clear from this figure that the low carbon wood chips Ayous and Sapelli activated and non-activated absorb throughout the infrared range. It is observed in this figure a broad absorption band at 3,500–3,000  $\text{cm}^{-1}$  common to all coals, with a

minimum around 3,200  $\text{cm}^{-1}$  characteristic of the stretching vibration of the hydrogen of the hydroxyl groups (carboxyls, phenols or alcohols) and water in the activated carbon. The presence of alcohols and/or phenols is confirmed by their characteristic bands of C–O and OH bonds observed at 1375.59, 1313.68, and 1254.24  $\text{cm}^{-1}$  for ANA (Fig. 4(a)); at 1241.75  $\text{cm}^{-1}$  for AA (Fig. 4(b)); at 1208.84 and 1113.50  $\text{cm}^{-1}$  for SNA (Fig. 4(c)) and finally at 1240.84  $\text{cm}^{-1}$  for SA (Fig. 4(d)). However, we note that there are some peaks disappear after activation of coals, indicating that these groups participate in the chemical modification.

The  $\text{CH}_2$  stretching band at 3,000 and 2,840  $\text{cm}^{-1}$  are assigned to asymmetric and symmetric stretching of  $\text{CH}_2$  groups [33,34], or aromatic groups méthoxyls [35], respectively, which presents practically the same wavenumbers before and after the chemical modifications, indicating that these groups did not participate in the chemical modification [36].

The aromatic nuclei also contribute to the broad absorption band around 3,300  $\text{cm}^{-1}$ . The bands between 913.66 and 657.45  $\text{cm}^{-1}$  correspond to the deformation vibration of the C–H outside the plane of aliphatic structures.

The small band around 1,700  $\text{cm}^{-1}$  is usually attributed to the vibrations stretching of C=O of ketones, aldehydes, lactones or carboxylic groups. It clearly appears in all spectra at 1698.16  $\text{cm}^{-1}$  (ANA), 1705.97

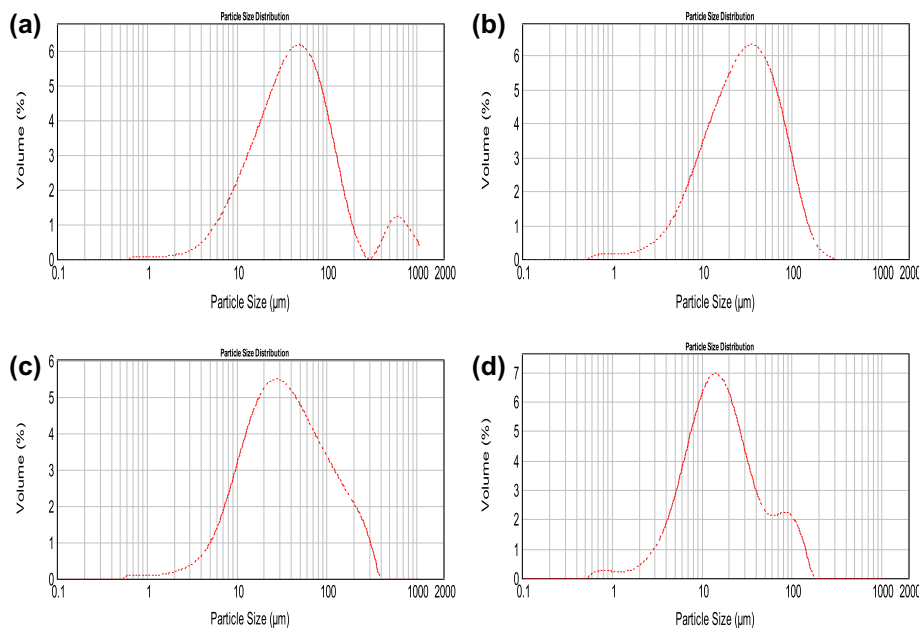


Fig. 3. Particle size distribution of (a) non-activated carbon from Ayous wood, (b) activated carbon from Ayous wood, (c) non-activated carbon from Sapelli wood, and (d) activated carbon from Sapelli wood.

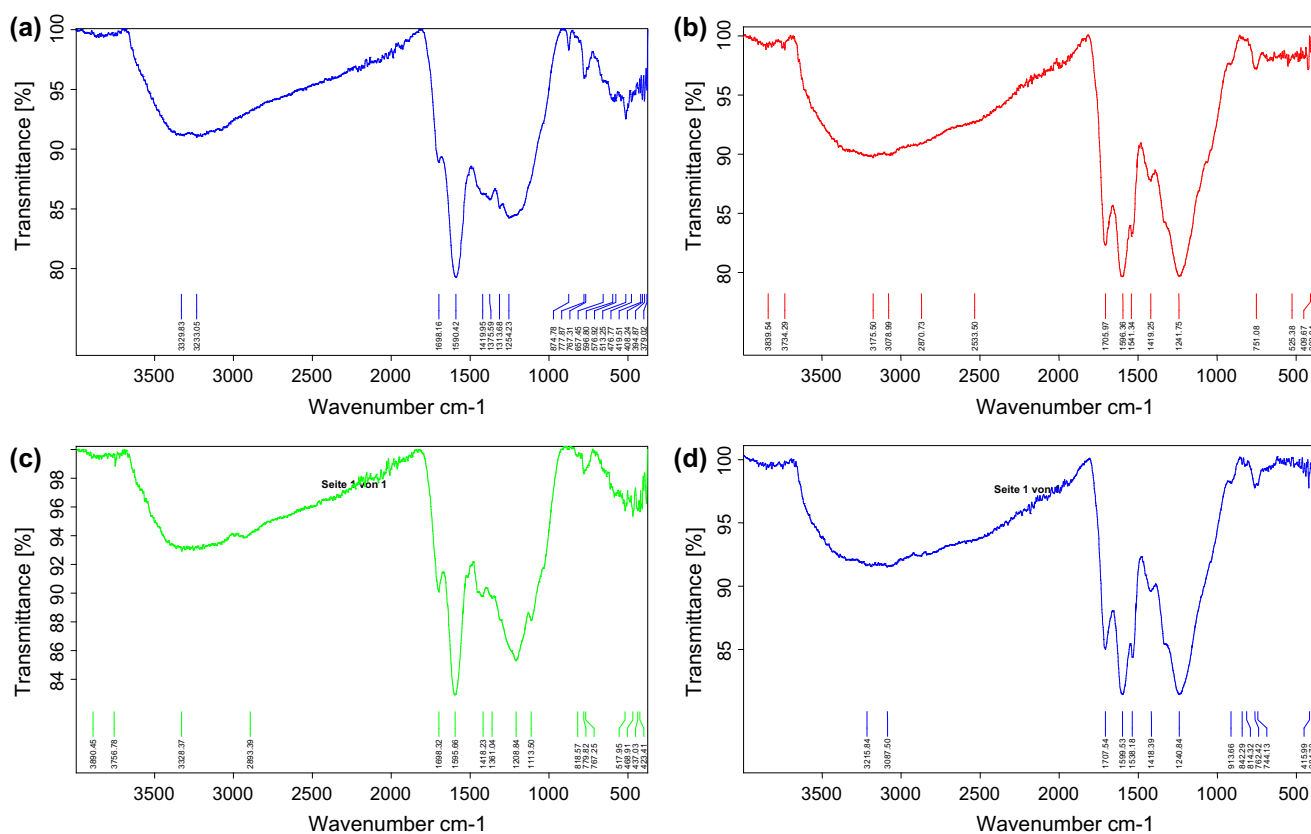


Fig. 4. Infrared spectrum of non-activated carbon and activated carbon of Ayous and Sapelli (a) non-activated carbon of Ayous (ANA), (b) activated carbon of Ayous (AA), (c) non-activated carbon of Sapelli (SNA), (d) activated carbon of Sapelli (AS).

cm<sup>-1</sup> (AA), 1698.32 cm<sup>-1</sup> (SNA), and 1707.54 cm<sup>-1</sup> (SA). The spectra also showed a strong band before and after the chemical modification at 1599.53–1590.42 cm<sup>-1</sup> due to vibration of C=C bonds in the aromatic rings. The region around 1,400 cm<sup>-1</sup> on the other hand gives some idea of the relative abundance of CH<sub>2</sub> and CH<sub>3</sub> groups.

All spectra (Fig. 4(a)–(d)) shows a broad band in the spectral region between 1,300 and 900 cm<sup>-1</sup>. This band is commonly described in the oxidized carbon, and is attributed to the stretching of C–O groups in the acids, alcohols, phenols, ethers, and esters, but it also a characteristic of phospho carbon and phosphorus compounds in activated carbon by phosphoric acid [37].

The results of the characterization of the carbon products are listed in the Table 2 below.

It is observed from this table that the yields are lower than 50% which means that there is a production of micropores and mesopores at the expense of macropores [38]. High values of the iodine index and low methylene blue index values confirmed that the carbon product consists mainly of mesopores and micropores and therefore present a greater ability to bind molecules. These results are consistent with those observed in SEM described above.

Table 2  
Iodine and methylene blue index of activated carbon

Biomasses	Yield (%)	Iodine Index (mg g <sup>-1</sup> )	Methylene blue index (mg g <sup>-1</sup> )
Ayous	27	1172.70 ± 1.07	49.6 ± 0.5
Sapelli	29	1133.50 ± 2.03	51.1 ± 0.3

### 3.3. Adsorption of paraquat on activated carbon

#### 3.3.1. Influence of activation

Fig. 5 shows changes in the quantities of paraquat adsorbed on activated and non-activated carbons as a function of the contact time at  $200 \text{ mg L}^{-1}$ , pH 6.5, and at a temperature of  $25^\circ\text{C}$  and 50 mg of carbon.

It can be noted from Fig. 5 that the amount of adsorbed paraquat varied significantly as a function of the nature and type of carbon, irrespective of the concentration of the paraquat molecule in solution. This result could be explained by the increase in the specific surface area after acid activation and in turn increase in the adsorption sites.

#### 3.3.2. Kinetics and modeling of paraquat adsorption

Changes in the quantities of paraquat adsorbed as a function of the contact time for varying concentrations (100–400  $\text{mg L}^{-1}$ ), for the various types of carbon

(Ayous and Sapelli) is presented in Fig. 6. Generally, these curves present three phases: (1) rapid growth phase, which indicates that paraquat is adsorbed very rapidly regardless of the concentration, type and nature of the carbon in solution; (2) a second slow growth phase, and (3) the third phase which is in the form of a plateau where the adsorption of the molecule is maximal.

The first step corresponds to the adsorption of paraquat on the most accessible sites located on the external surfaces of the carbon. The second phase in turn corresponds to the start of saturation of the available sites on the surface of the carbon, as well as the fixation of the molecule inside the pores due to their tubular structure. After this phase, the quantities paraquat retained stopped evolving and the presence of a plateau on the kinetics in the third stage indicates that the adsorption equilibrium has been attained. The equilibrium time observed from the kinetics is identical and varies between 10 and 20 min. These

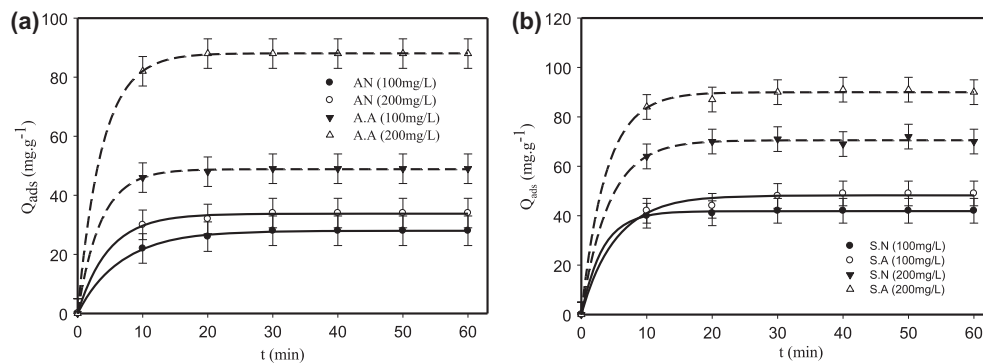


Fig. 5. Influence of carbon activation on paraquat adsorption at  $25^\circ\text{C}$  ( $200 \text{ mg L}^{-1}$ ;  $m = 50 \text{ mg}$  and pH 6.5, (a) Ayous wood chips, (b) Sapelli wood chip). (pH = 6.5; FI =  $5 \times 10^{-3} \text{ M}$ ;  $m = 50 \text{ mg}$ ; temperature of  $25^\circ\text{C}$ ) (a) activated carbon from Ayous wood chip, (b) unactivated carbon from Sapelli wood chip.)

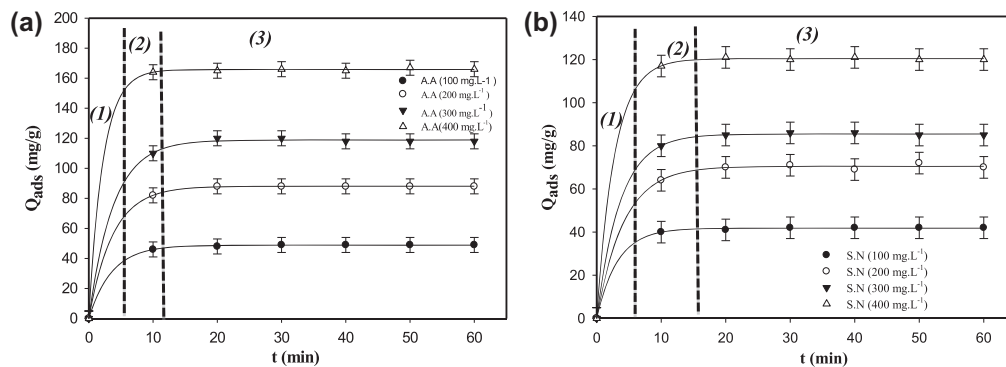


Fig. 6. Kinetic curves for the adsorption of paraquat on wood carbon at  $25^\circ\text{C}$ . (pH = 6.5; FI =  $5 \times 10^{-3} \text{ M}$ ;  $m = 50 \text{ mg}$ ; temperature of  $25^\circ\text{C}$ ) (a) activated carbon from Ayous wood chip, (b) unactivated carbon from Sapelli wood chip.)

results are consistent with that reported by [39], who observed a very high removal of paraquat by 10 min on CAP from Ayous wood.

In order to investigate the adsorption kinetics of paraquat, three kinetic models, namely: pseudo-first-order, pseudo-second-order and intra-particle diffusion models, were used in this study. Table 3 shows the different parameters of the kinetic models applicable to the adsorption of paraquat in solution. It can be noticed from the pseudo-first order and pseudo second order model that the calculated amounts adsorbed ( $q_{cal}$ ) increases with increasing concentration regardless of the type or nature of the carbon. The values of the coefficient of determination ( $R^2$ ) are all greater than 0.99 showing that this model is appropriate to explain the kinetics of paraquat adsorption on this carbon. The linearity of this kinetic model can be better seen in Fig. 7.

Finally, it is apparent from Table 3 above that the intra-particle diffusion constant obtained for the adsorption of paraquat ( $K_{int1}$ ) and also the slope of the first part of the diffusion curve (Fig. 7(c) and (d)) increases with the concentration irrespective of the nature or type of carbon. This first phase is almost linear as the coefficients of determinations ( $R^2$ ) are all greater than 0.95. On the contrary, at the last stage, the intra-particle diffusion constant ( $K_{int2}$ ) varies slightly and non-linearly as the values of  $R^2$  are all less than 0.95. This observation had already been made by [40] during the adsorption of the atrazine on montmorillonite.

### 3.4. Influence of paraquat concentration, activated carbon mass, pH, and ionic force on the adsorption kinetics of paraquat

#### 3.4.1. Influence of paraquat concentration and activated carbon mass

Fig. 8 shows the variation in the quantities of paraquat adsorbed on carbon (Fig. 8(a) and (b)) and the changes in the amounts of paraquat adsorbed (Fig. 8(c) and (d)) as a function of the contact time.

In Fig. 8(a) and (b) the shape of the kinetic curves is the same as described above regardless of the mass of carbon introduced. It was however noted that the amount of paraquat adsorbed decreases as the mass of carbon introduced into the medium increases. This amount drastically reduces as we move from 25 mg to 1 g of carbon for the two types of wood. This result could be explained by the fact that increase in mass can reduce mobility of molecules in solution. This result is concordance with the work of [41] who evaluated the adsorption of methoxychlor, methyl

Table 3  
Kinetic parameters of the pseudo-first-order, pseudo-second-order and intra-particle diffusion models for the adsorption of paraquat on activated carbon from Ayous and unactivated carbon from Sapelli wood chips

Carbon type	Concentration ( $\mu\text{g L}^{-1}$ )	Pseudo-first order model		Pseudo-Second order model		Intra-particle diffusion model							
		$K_1$ ( $\text{min}^{-1}$ )	$q_{e\text{ cal}}$	$R^2$	$q_{e\text{ cal}}$	$K_2$ ( $\text{g mg}^{-1} \text{min}$ )	$R^2$	$K_{int1}$ ( $\text{g mg}^{-1} \text{min}$ )	$C_1$	$R_1^2$	$K_{int2}$ ( $\text{g mg}^{-1} \text{min}$ )	$C_2$	$R^2$
AA	100	0.273	46.586	0.999	7.235	0.447	0.996	17.480	-3.531	0.986	0.960	42.733	0.755
	200	0.270	89.184	0.999	9.713	0.414	0.996	30.981	-6.697	0.987	2.353	74.383	0.807
	300	0.263	119.701	0.999	11.322	0.398	0.996	42.244	-10.251	0.988	3.638	97.763	0.814
	400	0.453	166.817	1	13.363	0.659	0.997	78.282	-14.328	0.984	9.548	126.06	0.716
SAN	100	0.308	41.625	0.999	6.742	0.479	0.994	14.88	-1.892	0.980	0.817	37.295	0.804
	200	0.241	71.715	0.999	8.704	0.353	0.996	23.703	-5.871	0.988	1.888	58.914	0.749
	300	0.273	84.953	1	9.578	0.429	0.996	29.949	-5.815	0.985	2.047	73.546	0.797
	400	0.353	117.778	0.999	11.396	0.597	0.994	43.694	-2.185	0.972	2.161	109.72	0.835



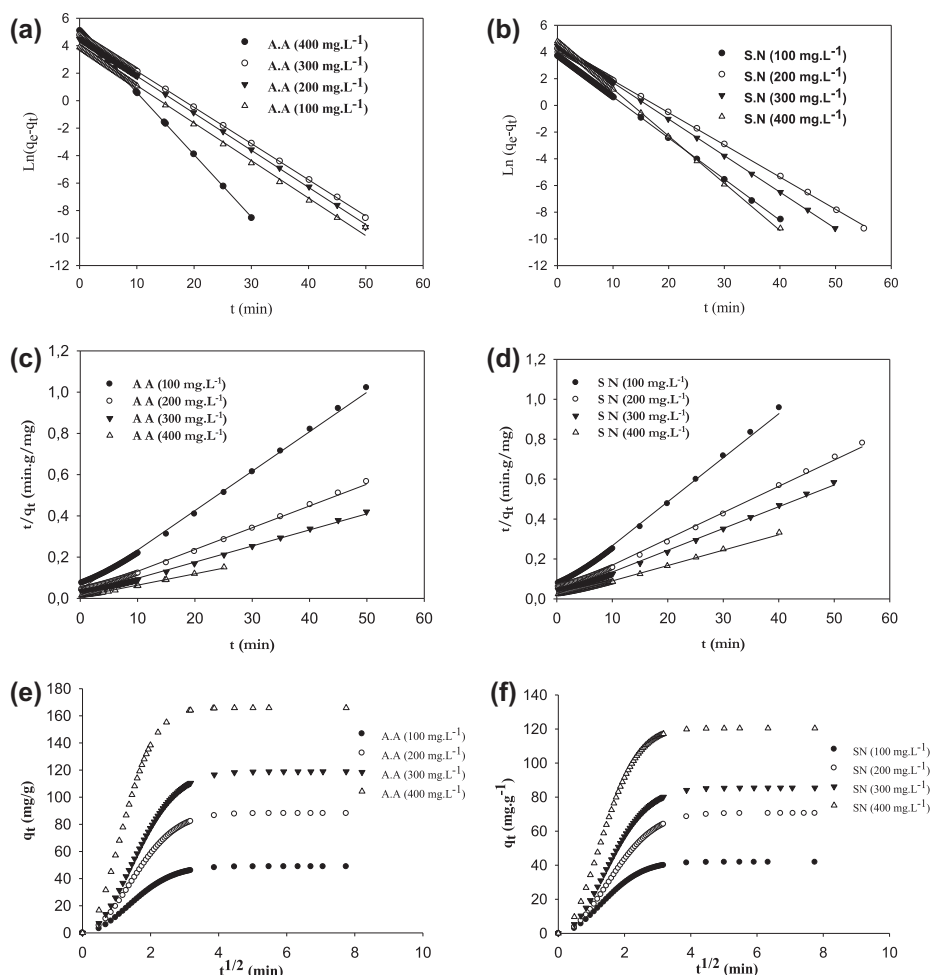


Fig. 7. Kinetic model of pseudo-first order (a), (b), pseudo second order (c), (d) and intra-particle diffusion (e), (f) for the adsorption of paraquat at pH 6.5;  $FI = 5 \times 10^{-3} M$ ;  $m = 50 \text{ mg}$ ; temperature of  $25^\circ\text{C}$ , (AA = on activated carbon from Ayous wood chips, and SN = unactivated carbon from Sapelli wood chips).

parathion, and atrazine on activated carbon, [42] also observed similar results. On the contrary, [43] had opposite results observed during the removal of paraquat in solution on activated bleaching earths.

In Fig. 8(c) and (d), it can be noted that the variation of the concentration of paraquat in solution does not influence the time at which adsorption is at equilibrium. However, the amounts of adsorbed molecule increases with the concentration of paraquat introduced. The amount adsorbed is rapid in the first phase and nearly constant in the second phase, whatever the type and nature of the carbon. This result could be explained by the fact that at lower concentrations of paraquat, the diffusion of the molecule to the adsorption sites on the surface of carbon is much lower than at higher concentrations. Paraquat is a molecule which is very soluble in water and gets fixed on the surface of CAP via hydrogen bonds or by van der Waals bonds. This result agrees with that of [42], who studied

the adsorption of paraquat in solution on activated carbon. They also noticed that the adsorption of paraquat increases with its concentration in the medium.

### 3.4.2. Influence of pH and ionic force of the mixture

Fig. 9 shows the variations in the quantities of paraquat adsorbed on CAP with contact time for different values of pH and ionic strength. It was noted that, the amount of paraquat adsorbed did not vary significantly with an increase in pH of the mixture whatever the type of carbon. This can be observed from the values of the adsorbed quantities which are substantially identical. This result is consistent with the work of [43] that showed that the quantity of adsorbed molecules in solution was almost constant at varying pH (5, 7, and 9). However, contrary results

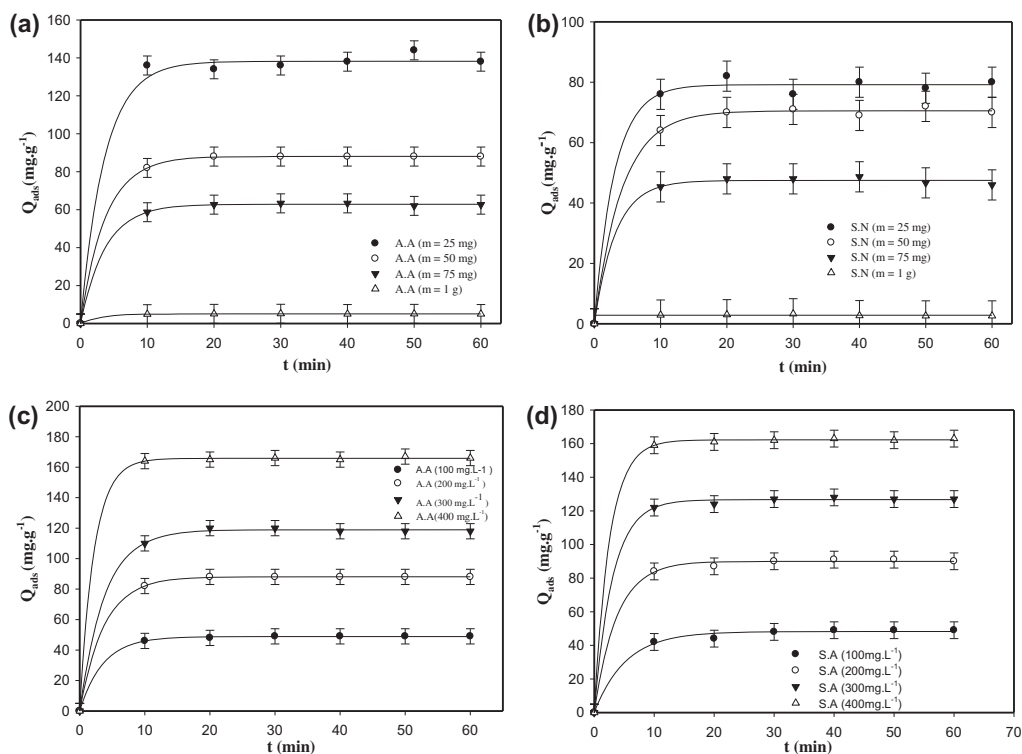


Fig. 8. Influence of the mass of carbon (a), (b) at  $200 \text{ mg L}^{-1}$  of paraquat and paraquat concentration (c), (d) at  $25 \text{ mg}$  of carbon) on the adsorption kinetics at  $\text{pH } 6.5$  and  $\text{FI} = 5 \times 10^{-3} \text{ M}$ , temperature of  $25^\circ\text{C}$ , (AA) activated carbon from Ayous wood chips, (SA) activated carbon from Sapelli wood chip, (SN) non-activated carbon from Sapelli wood chips.

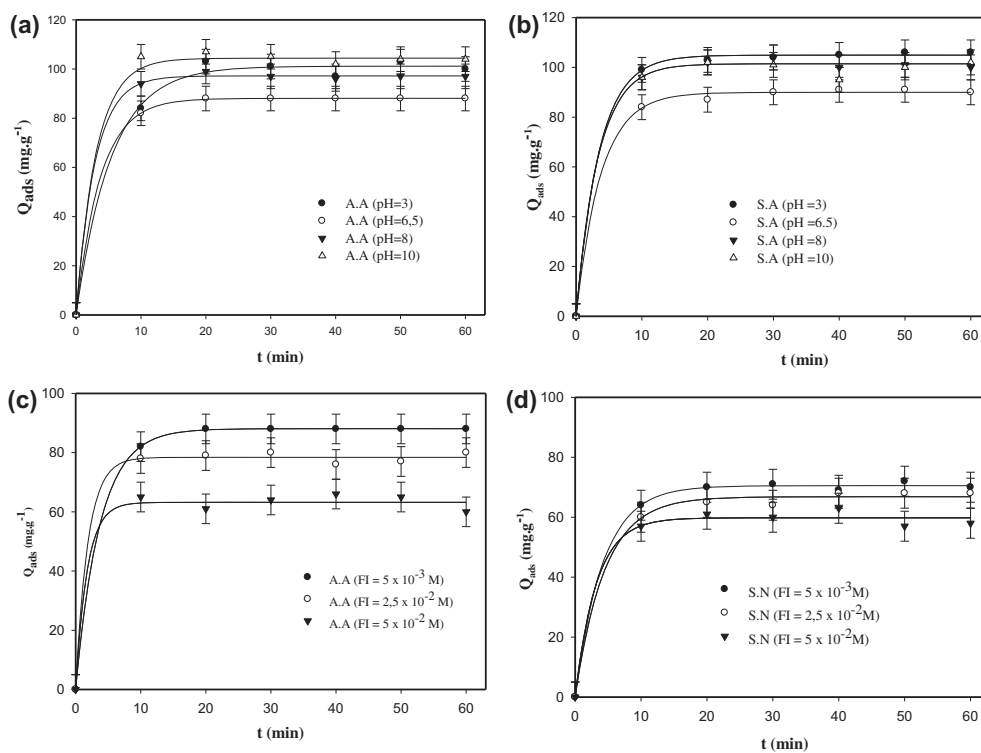


Fig. 9. Influence of  $\text{pH}$  (a), (b) and ionic strength ((c), (d) at  $\text{pH } 6.5$ ) on the adsorption kinetics of paraquat at  $200 \text{ mg L}^{-1}$ ,  $m = 25 \text{ mg}$ , temperature of  $25^\circ\text{C}$ , (AA) activated carbon from Ayous and (SN) non activated carbon from Sapelli.

were obtained by [44] who rather observed an increase in the amount adsorbed when the pH of the medium dropped (11, 7, and 3).

It can be noted on Fig. 9 that increase in ionic strength of the mixture causes a slight decrease in the amount of adsorbed paraquat, regardless of the nature and type of carbon. Thus, it can be observed for the activated carbon from Ayous (AA) that the adsorbed amount decreased from 88 to 60 mg g<sup>-1</sup> when the ionic strength increased from 0.005 to 0.05 M. This could be explained by the effect of competition between the potassium ion (K<sup>+</sup>) and paraquat molecule. In fact, increasing the salt concentration (KCl) in the medium resulted to an increase in the amount of monovalent K<sup>+</sup> ion which also attached on the adsorption sites which would have been occupied by paraquat molecules (R–N<sup>+</sup>). This observation was also made by some authors including [44–46].

#### 4. Conclusion

The results reported in this study show that activated carbon produced from wood chips of Ayous (AA) and Sapelli (SA) have good capabilities to adsorb paraquat molecule from aqueous solution.

The adsorption kinetics of paraquat in solution by activated carbon was best described by pseudo-first order model and intra-particle diffusion model. The adsorption process is initially mass transfer controlled and later intra-particle diffusion controlled.

From the study of the influence of the physico-chemical parameters on the adsorption kinetics, it appeared that, an increase in concentration and mass of the adsorber impel respectively an increase and a decrease in the adsorbed quantity of the paraquat molecule. Variation in the pH did not affect the amount of paraquat adsorbed whatever the type of carbon. The amounts of paraquat adsorbed decreased as the ionic strength increased.

Ultimately, this study shows that activated carbon products obtained from Ayous and Sapelli wood chips which are wastes released into the environment, can be used as low-cost biosorbents for the removal of organic micropollutants (paraquat for example) in aqueous solution.

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