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Removal of reactive dyes from their aqueous solutions using *Moringa oleifera* seeds and *Grewia venusta* peel

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ABSTRACT

Moringa oleifera seeds (MOS) and *Grewia venusta* peel (GVP) were used in this study as biosorbents. The adsorption behaviours on two dyes were investigated through batch experiments. The best conditions for the maximum indigo carmine removal (99%) were found to be at pH 2, with 0.5 g of GVP and 150 mg/L as initial concentration of dye solution. In the same conditions at pH 5, the maximum removal was 85% when using MOS. The equilibrium adsorption data were fitted with the Freundlich model when using MOS ($R^2 = 0.9968$) and Langmuir model when using GVP ($R^2 = 0.9957$). In a methyl orange case, the adsorption capacity of MOS is 31.25 mg g^{-1} and the one of GVP is 5.78 mg g^{-1}. In an indigo carmine case, the adsorption capacity of MOS is 91.74 mg g^{-1} and the one of GVP is 188.68 mg g^{-1}. This study contributed to a better understanding of methyl orange and indigo carmine dyes removal process from aqueous media by *M. oleifera* seeds and highlighted the sorption properties of *G. venusta* peel which is rarely used as absorbents in adsorption process. According to the obtained results, MOS and GVP may be potentially used in industry as natural biosorbents for textile dying wastewater treatment.

Keywords: *Moringa oleifera*; *Grewia venusta*; Methyl orange; Indigo carmine; Adsorption isotherms

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1. Introduction

Decontamination of environment has become a real challenge in recent years. Azo dyes such as methyl orange and indigo dyes are widely used in industries such as textiles, foodstuffs, leather and about 15% of them end up in wastewaters [1]. Dyes are hazardous pollutants which can modify the physical and chemical properties of any ecosystem. They are affecting water transparency, reducing light penetration and gas solubility in water [2], also being mutagenic to human. Indigo carmine is one of the most important dyes mainly used in the dyeing of clothes (blue jeans), foodstuffs and other blue denim. However, it is considered as highly toxic indigoid class of dye and its touch can cause skin and eye irritations to human beings [3]. It can also cause permanent injury to cornea and conjunctiva [4]. Textile industries generate large volumes of wastewater. Wastewaters resulted in the dye finishing industry have high BOD to COD values (>2,000 mg O_2 L^{-1}), while the discharge limits [5] are much lower (BOD < 40 mg O_2 L^{-1}; COD < 120 mg O_2 L^{-1}), indicating the need for wastewater treatment. The removal of dyes from textile and leather effluents is one of the most significant environmental problems because most of the dyes are slowly or no biodegradable.

There are various methods available for the removal of dyes, including membrane-separation [6], electrochemical and biological process [7], advanced oxidation process [8], etc. Conventional techniques such as precipitation, coagulation and flocculation have been also used in wastewater treatment, although these techniques are not very efficient for removing several common dyes, especially from dilute solutions [9]. Among all these techniques, adsorption processes remain the most common useful techniques for the decontamination of the effluents of textile and dyeing industries. Several workers, during the past few years, have tried different adsorbents such as mineral sorbents [10], activated carbons [11], peat [12] and chitin [13] for the removal of reactive dyes from aqueous solutions. A great deal of work has also been done in the use of hen feathers as adsorbent for the removal of hazardous dyes as an innovative initiative [14–16]. In recent past, many studies showed the effectiveness of agricultural materials in removal of dyes [17–19]. Especially, the exploitation of agricultural materials as biosorbents in wastewater treatment has a great advantage with respect to environmental management. These materials are low cost and environmental friendly due respectively to their availability and their biodegradability. In this regard, some agro waste such as sugarcane bagasse, corncobs and cotton sticks were successfully used very recently for dyes removal [20,21]. However, to improve adsorption performance, new adsorbents are still under development.

It is well known that *Moringa oleifera* can be used in wastewater treatment without consequences, due to the fact that it is reported as non-toxic and biodegradable material. *M. oleifera* seeds possess antibacterial properties, coagulant properties [22] and acts as biosorbent for heavy metal removal [23]. Similarly, *Grewia venusta* is known for its abundance in Benin, but very little scientific work is done for its development.

The present study aims to investigate the performance of *M. oleifera* seeds and *G. venusta* peel on methyl orange and indigo carmine dyes sorption from aqueous solutions. The investigation for dyes removal was carried out through a batch experiments. The attention has been drawn to the understanding of mechanisms, sorption kinetic and equilibrium processes involved in sorption. The effects of pH, initial concentration of dyes and sorbents doses on the sorption phenomena have been studied.

2. Materials and methods

2.1. Preliminary treatment of sorbents

M. oleifera seeds (MOS) were collected from a tree in Cotonou city (south of Benin). The sample was firstly dried at room temperature (29 ± 1˚C) for one week. Then, husks were removed and the dry seeds were ground and sift through 100 μm sieve size. The resulting powder was used for all experiments as raw materials. The infrared spectroscopy analysis of *M. oleifera* seed powder is widely done in literature. Interesting results were obtained by Kwaambwa and Maikokera [24] from their study in which crude extract of *M. oleifera* protein was characterised by FTIR.

G. venusta peel (GVP) were collected in Natitingou city (north of Benin) and dried under sunlight for one week, followed by oven drying at 45˚C for 24 h, in order to get constant weight. Then, the material was ground and powdered using domestic mixer and the resulting powder was sifted to 100 μm. The infrared spectrum of GVP peel powder was recorded with a Fourier transform infrared spectrometer (Bruker IFS55) equipped with a DTGS detector in the transmission mode. One (1) milligram of powder was mixed with 150 mg KBr (FTIR grade), and a pellet was prepared using a 10 T press. The spectra were recorded in the 4,000–400 cm^{-1} range with 64 scans collected at 2 cm^{-1} resolution.

2.2. Preparation of aqueous dye solution

All chemicals used in the present study are of analytical grade and used as such without further purification. Stock solutions (1,000 mg/L) were prepared by dissolving 1 g of dye in 1,000 mL of distilled water. The experimental solutions of different concentrations were made by further dilutions. The chemical structures of the two dyes are presented in Fig. 1.

2.3. Batch experiments

The batch tests were performed with aqueous solution containing methyl orange (MO) or indigo carmine (IND) dye. The effect of pH, initial concentration of dye solution and amount of MOS or GVP were studied. All pH measurements were carried out with a pH-metre VWR Symphony model. In each sorption experiment, 500 mL of dye solution with known concentration and known pH was put in 1,000 mL beaker at room temperature (28 ± 1˚C). Then, a known amount of biosorbent was added and the mixture was stirred at 90 rpm using flocculation tester equipment (VELP Scientifica FC6S). The dye sample solution was filtered using filter paper (Whatman N˚1) and the absorbance of filtrate was measured by spectrophotometer at 481 nm for methyl orange and 604 nm for indigo carmine. The initial pHs of suspension were adjusted from 2 to 10 using 0.1 M HCl or 0.1 M NaOH solutions. Effect of biosorbent dosage was studied with different doses ranging from 0.5 to 5 g of MOS or GVP, with time stirring ranging from 5 to 30 min. For isotherm experiments, 0.5 g of each biosorbent was mixed with 500 mL of dye solution with varied initial concentrations (5–40 mg L^{-1}) in the case of methyl orange and (20–200 mg L^{-1}) as far as indigo carmine is concerned. Each batch experience was done tree times and the average of data was considered. The removal per cent of dye (%*R*) and the capacity of dye adsorbed, q_e (mg g^{-1}) was determined.

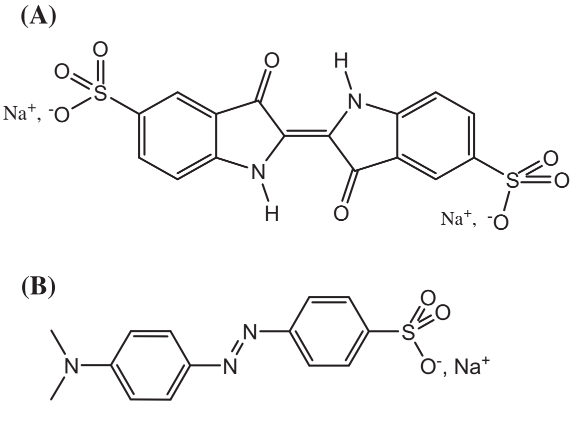

Fig. 1. Chemical structures of: (A) indigo carmine and (B) methyl orange.

2.4. Isotherm studies

The isotherms of dyes sorption describe how solute in solution interact with sorbents. It is well described in literature that isotherm can be well linearised by Freundlich or Langmuir model [13,16].

2.4.1. Freundlich isotherm

The well-known Freundlich isotherm [13,16] used for isothermal sorption is a special model for heterogeneous surface energy in which the energy term in the equation varies as a function of surface coverage strictly due to variation of the sorption. The Freundlich equation is given as:

$$\log q_e = \log K_F + n \times \log C_e \tag{1}$$

where K_F is an indicator of the adsorption capacity and n is an indicator of the adsorption intensity. K_F and n can be determined from the linear plot of $\log(q_e)$ vs. $\log(C_e)$.

2.4.2. Langmuir isotherm

The widely used Langmuir isotherm [13,16] has found successful application in many real sorption processes and is expressed as:

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{bC_eq_m} \tag{2}$$

where q_m (mg g^{-1}) is the maximum adsorbate amount that forms a complete monolayer on the surface, and b (L mg^{-1}) is the Langmuir constant related to adsorption heat. When $1/q_e$ is plotted vs. $1/C_e$ and the data are regressed linearly, q_m and b constants can be calculated from the slope and the intercept.

2.5. Adsorption kinetics

In order to understand the mechanism of adsorption and potential rate controlling step, a pseudo-first-order equation and pseudo-second-order equation models have been employed to test experimental data of dyes sorption. The conformity between experimental data and the model-predicted values was expressed by the correlation coefficients R^2 [14,18,21].

2.5.1. Pseudo-first-order kinetics

The pseudo-first-order model has been widely used to predict the sorption kinetics. It is expressed as:

$$\frac{\mathrm{d}q_{\mathrm{e}}}{\mathrm{d}t} = k_1(q_{\mathrm{e}} - q_t) \tag{3}$$

After integration, and at the initial condition $q_t = 0$ at $t = 0$, it becomes:

$$\log(q_{\mathrm{e}} - q_t) = \log\, q_{\mathrm{e}} - \frac{k_1 t}{2.303} \tag{4}$$

where k_1 (min^{-1}) is the rate constant of the pseudo-first-order adsorption.

The plot of $\log(q_{\mathrm{e}} - q_t)$ vs. t gives a straight line for the first-order sorption kinetics, which allows the computation of the sorption rate constant k_1.

2.5.2. Pseudo-second-order kinetics

The pseudo-second-order model was expressed as:

$$\frac{\mathrm{d}q_t}{\mathrm{d}t} = k_2(q_{\mathrm{e}} - q_t)^2 \tag{5}$$

Integrating equation for the bounding conditions gives:

$$\frac{t}{q_t} = \frac{1}{k_2 q_{\mathrm{e}}^2} + \frac{t}{q_{\mathrm{e}}} \tag{6}$$

where k_2 (g mg^{-1} min^{-1}) is the second-order rate constant of the pseudo-second-order adsorption.

The plot of t/q_t vs. t gives a straight line for the second-order sorption kinetics, which allows the computation of the sorption rate constant k_2.

3. Results and discussion

3.1. FTIR spectrum

From the results obtained by Kwaambwa and Maikokera [24], the IR spectrum of *M. oleifera* seeds showed two strong absorption bands at 1,656 cm^{-1} and at 1,540 cm^{-1} attributed of amide I and amide II vibration, respectively. It showed also a pronounced absorption band at 1,107 cm^{-1}, which is a characteristic of polysaccharides [25]. Ndabigengesere et al. [26] had earlier reported that carbohydrates represent 5% of the shelled *M. oleifera* seeds.

The photochemical screening experiments of *G. venusta* peel showed the presence of alkaloids, leucoanthocyans, triterpenoids, anthracene, mucilages and reductant compounds. The presence of mucilage and reductant compounds in GVP was confirmed by Nenonene et al. [27] and Nep and Conway [28]. In their studies, these authors found that, GVP contains 20–32% of mucilage and some polysaccharides with 5,925 kDa as molecular weight in which there are monosaccharide such as glucose, rhamnose, xylose, galactose, arabinose and galacturonic acid.

The IR spectrum of *G. venusta* peel (Fig. 2) exhibits the typical bands and peak characteristic of polysaccharides. The broad band around 3,400 cm^{-1}, showed the presence of hydroxyl (–OH) groups due to humidity, but could also arise from the hydroxyl of sugar rings. The peak obtained at 2,927 cm^{-1} results from stretching modes of the C–H bonds of methyl groups (–CH$_3$). Absorption bands around 1,616 and 1,432 cm^{-1} are typical of carboxylate groups of the galacturonic acid residues [29]. The wave numbers between 800 and 1,200 cm^{-1} represent the finger print region for carbohydrates [30].

3.2. Effect of contact time

The effect of contact time on removal of both dyes was studied at different pHs. It showed in Fig. 3 the results of methyl orange and indigo carmine removal

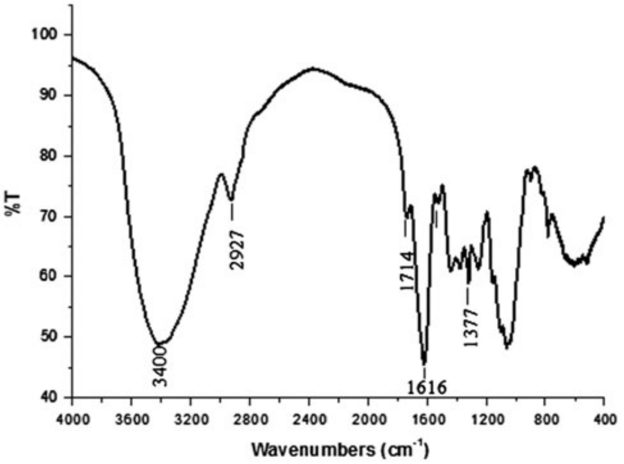

Fig. 2. FTIR spectrum for *G. venusta* peel. %*T* is the percentage transmittance plotted against wave number.

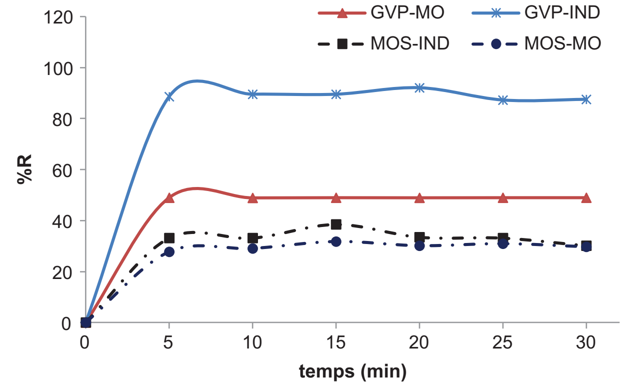

Fig. 3. Effect of contact time on removal dyes by MOS and GVP at pH 2. (Concentration of methyl orange solution: 30 mg/L; concentration of indigo carmine solution: 150 mg/L; adsorbent mass 1 g; T = 28°C).

using MOS and GVP at pH 2. It appears that, the removal of both dyes was rapid at the process beginning (05 min), but becomes constant till 30 min. The rapid removal at initial contact time may be related to the availability of high sorption sites on both biosorbents at the beginning, and the slow sorption rate in the next minutes was due to slower diffusion of solute inside the biosorbent. The same result was obtained by Umpuch and Sakaew [31] in their study on removal of methyl orange from aqueous solutions by adsorption using chitosan intercalated montmorillonite.

3.3. Effect of pH

The initial pH of dye solution has a great influence on sorption of dye onto sorbent due to the effect of the surface properties or the dissociation of dye molecules. Fig. 4(a) shows the effect of pH on the removal of both dyes. The experiments with methyl orange showed that as the pH increased from 2.0 to 4.0, the removal efficient rapidly decreased from 31.71 to 18.65% when using MOS and from 48.9 to 21.15% when using GVP. It then slowly decreased from 18.65 to 8.45% for MOS and 21.15 to 12.30% for GVP with a further increase in the pH from 4.0 to 10.0 as far as the indigo carmine is concerned, as the pH increased from 2.0 to 4.0, the removal efficient rapidly decreased from 69.35 to 43.80% when using GVP and slowly decreased from 43.80 to 10.81% with a further increase in the pH from 4.0 to 10.0. But when using MOS, the removal efficiency increased from 38.46 to 86.54% with an increasing of pH from 2.0 to 5.0. It then slowly decreased from 86.54 to 25.32% when the pH increased from 5.0 to 10.0.

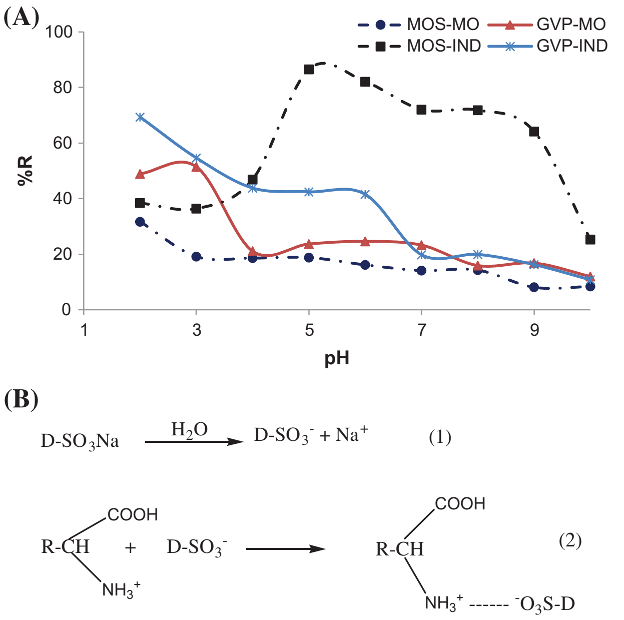

Fig. 4. (A) Effect of pH on dyes removal (Concentration of methyl orange solution: 30 mg/L; concentration of indigo carmine solution: 150 mg/L; adsorbent mass 1 g; T = 28°C) and (B) Mechanism of dyes removal.

The removal efficiency can be explained through two mechanisms. Firstly, an electrostatic attraction between the protonated groups of biosorbents and dyes molecules, and secondly, the chemical interaction between dyes molecules and biosorbents surface [32]. At lower pH, more protons were available causing an increase in electrostatic attraction between negative charge of dye surface and protonated groups of biosorbents. Methyl orange and indigo carmine are anionic dyes. In aqueous solution, both of them are dissolving as shown in first equation (Fig. 4(b)). The second equation explains the reaction between dyes molecules and protoneted biosorbent molecules.

As the initial pH of the dye solution increased, the positive charge on the sorbent surface decreased and the number of negative charge sites increased in suspension. Then, occurred a competition between the hydroxide ions and the dye anions. The electrostatic repulsion between anionic dye molecules and the excessive hydroxide ions resulted in a sharp decrease in sorption. These results agree well with those reported by Sumalatha et al. [11]. They found highest removal of indigo carmine from aqueous solution at pH 3 using activated carbon while Ramesh et al. [33] found maximum adsorption of indigo carmine at pH 6 when using magnesium oxide to remove the dye. The same results were obtained by Umpuch and Sakaew [31] and Saha et al. [34] during their study on methyl orange onto chitosan from aqueous solutions.

3.4. Effect of biosorbent dosage

The effect of MOS and GVP amount on removal of both dyes was investigated at initial dye concentration of 30 mg L^{-1} for methyl orange solution and 150 mg L^{-1} for indigo carmine solution. Fig. 5 shows the removal efficiency obtained with the variation of biosorbent mass at different times. It was noticed that the variation of MOS and GVP mass had not significant effect on equilibrium time. In Fig. 5, it was noticed that the removal efficiency of each dye increases significantly as the GVP and MOS amount increases due to increasing of available sorption sites. The removal per cent of methyl orange and indigo carmine increases, respectively, from 40.46 to 74.60% and 88 to 99% when increasing GVP amount from 0.5 to 5 g. Removal per cent of methyl orange and indigo carmine increases from 28.13 to 31.71% and from 82.58 to 84.37%, respectively, when increasing MOS dosage from 0.5 to 5 g.

3.5. Effect of initial dye concentration

The removal of both dyes by MOS and GVP was clearly dependent on the initial dye concentration of the solution (Fig. 6). An increase in the initial dye concentration leads to an increase in the biosorbent sorption capacity. This is due to the increase in the driving force of the concentration gradient, as the initial dye concentration has increased [35]. When using GVP, dye removal capacity was increased from 23 to 40.46% for methyl orange and from 69 to 99% for indigo carmine as the initial concentration of dye solution increases, respectively, from 5 to 40 mg L^{-1} and from 20 to 200 mg L^{-1}. Despite the fact that this increase is low in case of MOS, the same phenomenon was observed.

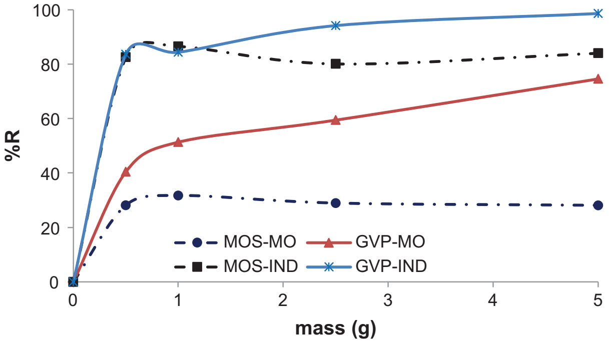

Fig. 5. Effect of biosorbent dosage on dyes removal (MOS-MO, pH 2; GVP-MO, pH 3; MOS-IND, pH 5; GVP-IND, pH 2; concentration of methyl orange solution: 30 mg/L; concentration of indigo carmine solution: 150 mg/L; $T = 28$°C).

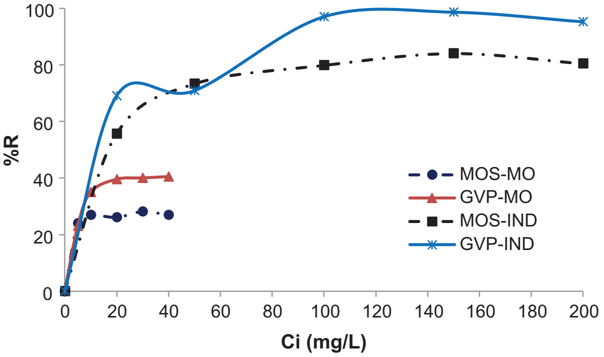

Fig. 6. Effect of initial dye concentration. (MOS-MO, pH 2; GVP-MO, pH 3; MOS-IND, pH 5; GVP-IND, pH 2; adsorbent mass 0.5 g; $T = 28$°C).

3.6. Isotherm sorption

The adsorption isotherm of dyes was analysed by models given by Langmuir and Freundlich (Fig. 7). The parameters of adsorption isotherm are show in Table 1. From Table 1, the values of correlation coefficients (R^2) show that the experimental data are more correct with

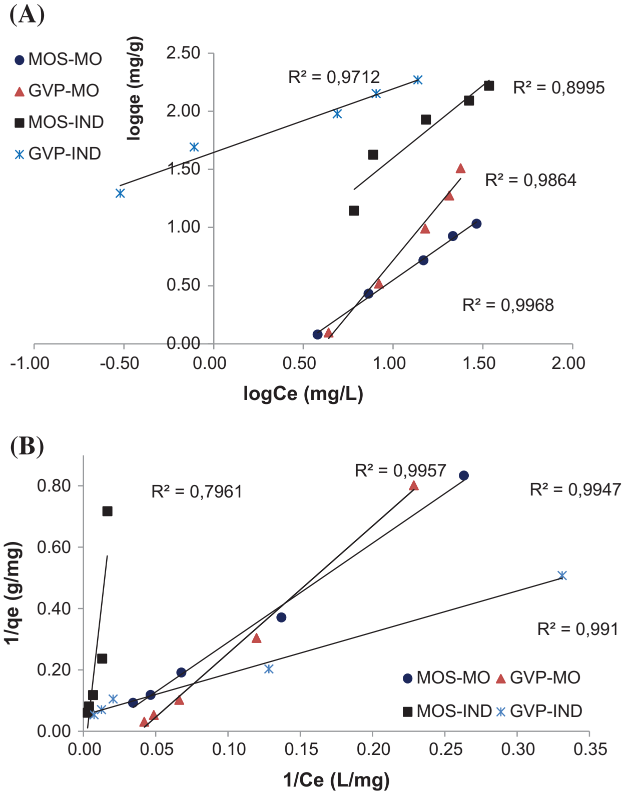

Fig. 7. Adsorption isotherm: (A) Freundlich model isotherm and (B) Langmuir model isotherm. (MOS-MO, pH 2; GVP-MO, pH 3; MOS-IND, pH 5; GVP-IND, pH 2; adsorbent mass 0.5 g; $T = 28$°C).

Table 1
Values of parameters of each isotherm model

		Langmuir model			Freundlich model		
		q_m (mg g^{-1})	b (l mg^{-1})	R^2	K_F	n	R^2
MOS	Methyl orange	31.25	0.01	0.9947	0.298	1.07	0.9968
	Indigo carmine	91.74	0.03	0.7961	2.244	1.25	0.8995
GVP	Methyl orange	5.78	1.37	0.9957	0.065	1.90	0.9864
	Indigo carmine	188.68	0.39	0.9910	44.33	0.55	0.9712

the Freundlich model than the Langmuir model for both dyes when using MOS. So, there is no saturation phenomenon on the surface of sorbent. This may be the availability of multilayer on the surface. In case of GVP, Langmuir model is more closed to experimental data, especially in indigo carmine removal, where the maximum sorption capacity was 188.68 mg g^{-1}. Here, the sorption was done on specific sites according to Langmuir's hypothesis.

The dyes sorption capacity of MOS and GVP was compared with other sorbents. In the case of methyl orange, the sorption capacity of MOS (31.25 mg g^{-1}) and GVP (5.78 mg g^{-1}) was lower than the ones of cross-linked chitosan (89.29 mg g^{-1}) [13] and chitosan intercalated montmorillonite (70.42 mg g^{-1}) [31]. As far as carmine indigo is concerned, the sorption capacity of MOS (91.74 mg g^{-1}) and GVP (188.68 mg g^{-1}) was better than ones of zeolite-iron oxide magnetic composite (0.58 mg g^{-1}) [10] and magnesium oxide (0.44 mg g^{-1}) [33], except pol(acrylic acid)/SiO$_2$ composite (523.11 mg g^{-1}) [34]. The comparison with an agro waste such as native sugarcane bagasse [20,21] showed that this material has a comparative sorption capacity of reactive dyes (43.17 mg g^{-1}) with MOS and GVP. From these results, MOS can be used for removal of both dyes while GVP is better to remove indigo carmine than methyl orange.

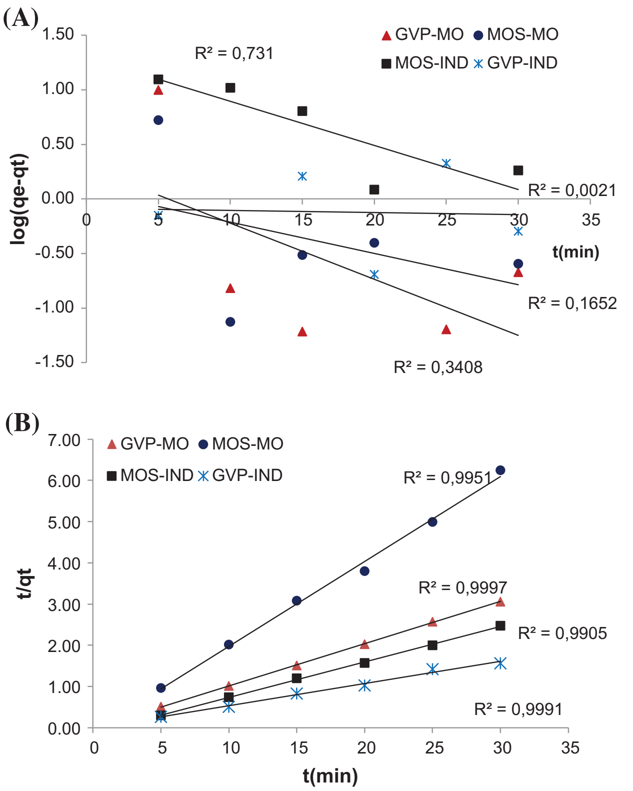

Fig. 8. Kinetic model: (A) pseudo-first-order kinetic and (B) pseudo-second-order kinetic. (MOS-MO, pH 2; GVP-MO, pH 3; MOS-IND, pH 5; GVP-IND, pH 2; concentration of methyl orange or indigo carmine solution: 20 mg/L; adsorbent mass 0.5 g; T = 28°C).

3.7. Adsorption kinetics

Fig. 8 reports the results of the dyes sorption kinetic tests. The pseudo-first-order kinetic did not

Table 2
Kinetics parameters

	Pseudo-first-order kinetic				Pseudo-second-order kinetic			
	MOS		GVP		MOS		GVP	
	k_1 (min^{-1})	R^2	k_1 (min^{-1})	R^2	k_2 (g mg^{-1} min^{-1})	R^2	k_2 (g mg^{-1} min^{-1})	R^2
Methyl orange	0.07	0.1652	0.12	0.3408	0.48	0.9951	0.97	0.9997
Indigo carmine	0.09	0.7310	0.01	0.0021	0.06	0.9991	0.34	0.9905

apply of the investigated processes because of relatively low R^2 value. The pseudo-second-order kinetic fits very well to all the processes data. These results show parallel kinetics, running on small and large pores. In Table 2, the kinetic constants of the adsorption mechanisms on the investigated substrates are presented. A relatively high R^2 value indicates that the model successfully describes the kinetics of adsorption of methyl orange and indigo carmine.

4. Conclusion

The adsorption of methyl orange and indigo carmine dyes by *M. oleifera* seeds and *G. venusta* peel was a pH-dependent process with the maximum removal efficiency at acid pH. *M. oleifera* seeds are efficient in removal of both dyes while *G. venusta* peel is efficient in indigo carmine removal. The use of *G. venusta* peel as a biosorbent is a new method for the treatment of effluents, especially when related to methyl orange and indigo carmine compounds. The use of *G. venusta* peel and *M. oleifera* seeds as biosorbent presents operational, simplicity and adsorption capacity for the removal of organic compounds from aqueous solutions. But further studies should be done for simultaneous removal of two dyes and for complex wastewater treatment (containing dyes and heavy metals) using both biosorbents before their use in industrial applications. More characterisation techniques like scanning electron microscopy (SEM) and atomic force microscopy (AFM) should be done on biosorbents.

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