Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2017.20372

71 (2017) 327–333
April

Cadmium removal from aqueous solutions using *Moringa oleifera* seed pod as a biosorbent

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Received 24 May 2016; Accepted 23 November 2016

ABSTRACT

Biosorption is an effective, environmental friendly and cheap process for removal of contaminants from aqueous solutions. The aim of this study was to determine the *Moringa oleifera* seed pod (Mosp) efficiency in removal of cadmium from aqueous solutions. After determining optimum pH (pH = 6.8) and activation temperature (500°C) by pretests, the biosorption experiments were performed in batch system at room temperature. The effects of experimental parameters such as biosorbent dose (0.1–10 g/L), contact time (3–120 min) and initial Cd concentration (1–300 mg/L) were studied. The highest removal efficiency was 100% and was reached only after a few minutes contact time in the optimum operation condition. The biosorption was found better fitted to the Freundlich model, which was indicative of multilayer adsorption. It was concluded that the Mosp can be used as a successful and environmental friendly Cd biosorbent from aqueous solution especially industrial effluent containing high level of Cd.

Keywords: Biosorption; Cadmium; Equilibrium capacity; *Moringa oleifera*

1. Introduction

Organic and inorganic contaminants can be present in the atmosphere, groundwater, and food [1–12]. From these contaminants, heavy metals are widely distributed in the environment and is hence of particular concern. Toxic heavy metals may be discharged into the environment as industrial wastes and causing serious soil and water pollution [13]. Some of heavy metals including cadmium have no known important role in living organs and are toxic even in trace amounts. Cadmium has biological half-life in the range of 10–30 years and has the ability to accumulate in living organisms. The US Environmental Protection Agency [14] has classified cadmium as a priority pollutant and has set a value of 0.005 mg/L as maximum contaminant level goal and maximum contaminant level. Cadmium is non-biodegradable and must be removed from aqueous solution [15,16]. In the past decades, various traditional technologies for the removal of heavy metal ions such as ion exchange, precipitation, reverse osmosis, co-precipitation, electrochemical treatment, oxidation, and adsorption were widely used (Table 1). Among these strategies, adsorption is one of the most recommended physicochemical treatment processes for removal of inorganic contaminants and specially heavy metals [17–34].

Biosorbents application for the removal of different pollutants from aqueous solutions is one of the most recent developments in environmental and bioresource technology. The main advantages of this technology over conventional ones are its low cost, high efficiency, minimization of chemical sludges, and regeneration of biosorbent [35–41].

Many measures have been studied and developed for the effective removal of heavy metals using biosorbents, such

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Table 1
Advantages and disadvantages of different techniques in removal of heavy metals from aqueous solutions [46]

Method	Advantages	Disadvantages
Chemical precipitation	Inexpensive, simple, most of the metals can be removed	Disposal problems, large amounts of sludge produced [47]
Ion exchange	Metal selective, high regeneration of materials	Fewer numbers of metal ions removed, high cost [33]
Chemical coagulation	Dewatering, sludge settling	Large consumption of chemicals, high cost [47]
Membrane process and ultrafiltration	High efficiency (>95% for single metal), less solid waste produced, less chemical consumption	Removal % decreases with the presence of other metals, high initial and running cost, low flow rates [17]
Natural zeolite	Relatively less costly materials, most of the metals can be removed	Low efficiency [17]
Adsorption using activated carbon	High efficiency (>99%), most of the metals can be removed	No regeneration, cost of activated carbon, performance depends upon adsorbent [47]
Electrochemical methods	Pure metals can be achieved, metal selective, no consumption of chemicals	High running cost, high capital cost, initial solution pH and current density [33]

as sugar [42], gum arabic [43], peanut shell [44], and papaya wood [45]. One of these useful biosorbent is *Moringa oleifera* that grown in tropical areas and has been consumed as food in some African regions. *Moringa oleifera* is the most widely cultivated species of the genus *Moringa*, which is the only genus in the family *Moringaceae*. *Moringa oleifera* also is grown in semiarid, tropical, and subtropical areas of Iran including Bushehr Province in southwestern part of Iran. The coagulating demeanor of *Moringa oleifera* seed powder has been studied for different aspects of water treatment, but small studies have been directed toward its sorption demeanor for the removal of pollutants from water [28].

In the present study, the efficiency of the *Moringa oleifera* seed pod (Mosp) activated carbon in the removal of cadmium from aqueous solution was investigated. Furthermore, the effects of activation temperature, pH, biosorbent doses, cadmium concentration, and contact time were also studied.

2. Material and methods

2.1. Preparation of the biosorbent

After collection and transfer to laboratory, Mosp were washed by tap water and then two times by distilled water to remove sand, clay, and other impurities. The washed Mosp was placed in a muffle furnace and heated at 500°C for 1 h, then allowed to cool at room temperature and finally ground and sieved through a 0.71-mm screen. The prepared Mosp stored in a dark and dry place in laboratory for experiments.

2.2. Batch experiments

A stock solution of 1,000 mg/L cadmium was prepared by dissolving a corresponding amount of $Cd(NO_3)_2.4H_2O$. Specific concentration levels of cadmium solutions were prepared at 1, 2, 5, 10, 20, 50 and 300 mg/L concentration levels by diluting the stock solution. For experiments, fresh dilutions were prepared daily. At each stage of the experiment, 100 mL of Cd solution with a specific initial Cd concentration at neutral pH (pH = 7) was agitated at 120 rpm at room temperature. The effects of seven contact times (3, 5, 10, 15, 30, 60, and 120 min), seven initial Cd concentrations (1, 2, 5, 10, 20, 50, and 300 mg/L), and six biosorbent concentrations (0.1, 0.5, 1, 2, 5, and 10 g/L) were investigated in the batch experiments.

2.3. Analytical procedures

After each run, sample was filtered through a 0.45-μm membrane. Then, remaining cadmium in the solution after biosorption was determined by using an atomic absorption spectrometry (AAS) method (Varian, SpectrAA 240, Australia) with cadmium hallow cathode lamp and an air acetylene flame at 228.8 nm. The removal efficiency was calculated using Eq. (1) [48]:

$$\text{Biosorption yield} = \frac{(C_i - C)}{C_i} \times 100 \quad (1)$$

where C_i and C are the concentrations of cadmium before and after the experiment in any time (mg/L).

The equilibrium biosorption capacity of Mosp at different cadmium concentration levels was calculated by using Eq. (2):

$$q_e = \frac{(C_i - C_e)V}{m} \quad (2)$$

where q_e is the equilibrium biosorption capacity (mg/g); C_i is the cadmium concentration at initial time (mg/L); C_e is the concentration of cadmium in solution at equilibrium time (mg/L); V is the solution volume (L); and m is the biosorbent dosage (g).

2.4. Characterization of biosorbent

Fourier transform infrared spectroscopy (FTIR) spectra of Mosp samples were recorded by using an FTIR spectrometer (PerkinElmer FTIR, model Spectrum RXI) over the range of 400–4,000 cm^{-1} wave number.

3. Results and discussion

3.1. Characterization of the Mosp

FTIR analysis was used to identify the characteristic functional group on the surface of biosorbent. FTIR spectra of Mosp before and after Cd biosorption are shown in Fig. 1. The FTIR spectrum of Mosp exhibited a broad peak at 3,421.64 cm^{-1}, which is indicative of the existence of macromolecular association and may dente the pressure of both H-bonded amine and OH groups. The strong C–O band at 1,051.5 cm^{-1} due to $-OCH_3$ group also confirms the presence lignin structure in Mosp. The C–O stretching vibrations of aliphatic primary and secondary alcohols in cellulose, hemicellulose, and lignin. The broad biosorption between 1,350 and 1,550 cm^{-1} confirm the existence of N=O groups. A 1,053 cm^{-1} band could be vibration of O–C and C–N of polysaccharides. Spectra analysis of FTIR spectrum after divalent cadmium biosorption showed vibration of C–O and C–N at 1,053.44 cm^{-1} played the most important role in binding cadmium. The FTIR spectra of biosorbent indicated that the functional groups such as carboxyl and hydroxyl groups on the biosorbent surface could be the potential biosorption sites for Mosp biosorption. The shifts in the biosorption peaks generally observed indicate the existence of a metal-binding process, taking place on the surface of the Mosp.

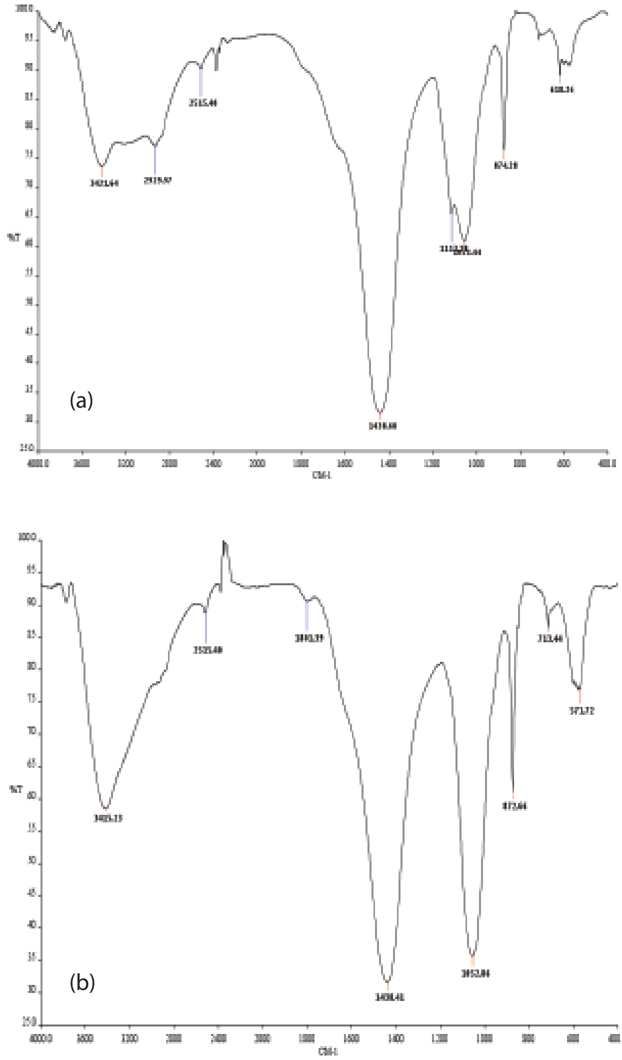

Fig. 1. FTIR spectra of Mosp before (a) and after (b) cadmium biosorption.

3.2. Effect of activation temperature

In order to determine the optimum activation temperature for biosorbent, a series of pretests was done at four different temperatures (100°C, 300°C, 400°C, and 500°C), biosorbent dose of 0.5 g/L, contact time of 30 min, initial Cd concentration of 1 mg/L, and pH value of 6.8.

The effect of Mosp activated temperature values on the removal efficiency of Cd is shown in Fig. 2. By increasing activation temperature, the Cd removing efficiency increased. So an activation temperature value of 500°C was selected as the best activation temperature to perform all experiments in this study.

3.3. Effect of pH

The effect of pH on Cd biosorption was investigated as pretests at three pH values from 3 to 10 (3, 6.8, and 10), biosorbent dose of 1 g/L, and initial Cd concentration of 1 mg/L. The effect of pH values of solution on the removal efficiency of Cd is shown in Fig. 3. The Cd removal efficiency

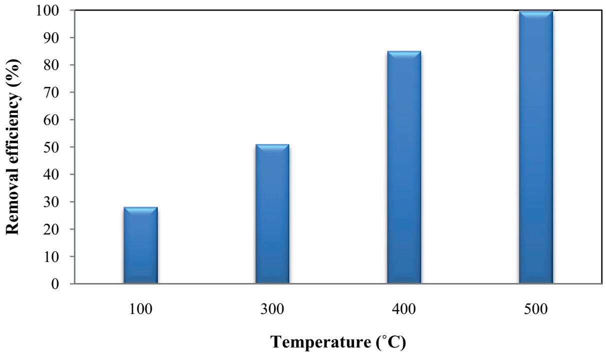

Fig. 2. Effect of temperature on Cd biosorption on to Mosp (biosorbent dose = 0.5 g/L, initial Cd concentration = 1 mg/L).

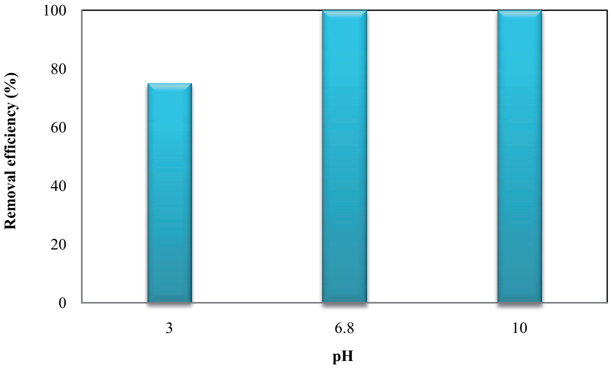

Fig. 3. Effect of pH on Cd biosorption on to Mosp (biosorbent dose = 1 g/L, initial Cd concentration = 1 mg/L).

was higher in pH value of 6.8 and alkaline pH value of 10 in comparison with acidic pH value of 3. It should be noted that beside sorption, precipitation could be another factor causing high percentage of Cd removal at pH value of 10. As there wasn't difference between pH values of 6.8 and 10 for Cd sorption onto Mosp, the pH value of 6.8 has been selected as the optimum pH to perform all experiments. pH controls biosorption by influencing the surface charge of the biosorbent, the degree of ionization, and the species of adsorbate. It is important to note here that in the case of some metals, the metal uptakes may decrease when pH is higher than the optimum pH. The same results have been reported for the metal removal by sea urchin test as biosorbents; the study results indicated that the metal sorption was negligible at pH 2, and increased with increasing pH value [49].

3.4. Effect of contact time and initial Cd concentration

The biosorption experiments were carried out in the Cd concentration range of 1–300 mg/L at neutral pH value of 6.8, contact times from 3 to 120 min, and 2 g/L biosorbent. As seen in Fig. 4, Cd biosorption rate decreased as the initial Cd concentration was raised. In the present adsorptive reaction, the Cd removal rate varied directly with the concentration of adsorbate (Cd). The capacity of the biosorbent got exhausted with increasing the initial Cd concentration. In the beginning and first minutes (3 min), biosorption rate was fast but after that almost remained constant. Constant biosorption rate with increasing contact time showed that equilibrium is reached, which is due to overlapping of the active areas on the Mosp at higher concentration of Cd and thus decreasing the net surface area. Jain et al. [34] and Abdel-Aty et al. [50] reported similar results in the Cd removal from aqueous solutions using sunflower waste carbon and alga *Anabaena sphaerica*, respectively. Abdel-Aty et al. reported that the biosorption of Cd ions was increased with subsequent increasing the biosorbent dose and almost became constant at higher dosage of 0.2 g/100 mL for Cd [50].

3.5. Effect of the biosorbent dose

The effect of biosorbent dosage on the removal of Cd was studied using various biosorbent doses of Mosp in the range of 0.1–10 g/L (Fig. 5). As presented in Fig. 5, the results showed that by increasing biosorbent dosage from 0.1 to 5 g/L the removal efficiency of Cd increased from 55% to 100%. This can be explained based on the fact that the higher the biosorbent dose in the aqueous solution, the greater the availability of exchangeable sites for Cd ion. Babu et al. [49] reported similar results for the removal of Cd by natural biosorbent.

3.6. Biosorption isotherm

In order to quantify the sorption capacity of Mosp in the removal of Cd from aqueous solutions, two usually used isotherms including Freundlich and Langmuir have been adopted [51]:

The Freundlich isotherm can be written as:

$$q_e = k_f c_e^{1/n} \tag{3}$$

and written in linear form as [52]:

$$\log q_e = \log k_F + \frac{1}{n}\log C_e \tag{4}$$

The plot of $\log C_e$ (the equilibrium concentration of Cd in solution [mg/L] after biosorption) vs. log q_e (the mass of Cd biosorbed per unit weight of the biosorbent [mg/g]) gives a straight line with a slope of $1/n$ (the Freundlich intensity parameter) and $\log k_F$ (the Freundlich capacity factor and a measure of adsorption capacity) as the intercept of $\log q_e(x/m)$ at $\log C_e = 0$.

The Langmuir adsorption isotherm is defined as:

$$q_e = \frac{q_b C_e}{1+bC_e} \tag{5}$$

and can be rewritten as [52]:

$$\frac{C_e}{q_e} = \frac{1}{bq_{\max}} + \frac{1}{q_{\max}}C_e \tag{6}$$

where q_e is the mass of Cd per unit mass of biosorbent (mg/g); $q_{\max}$ is the monolayer sorption capacity; and b is the Langmuir

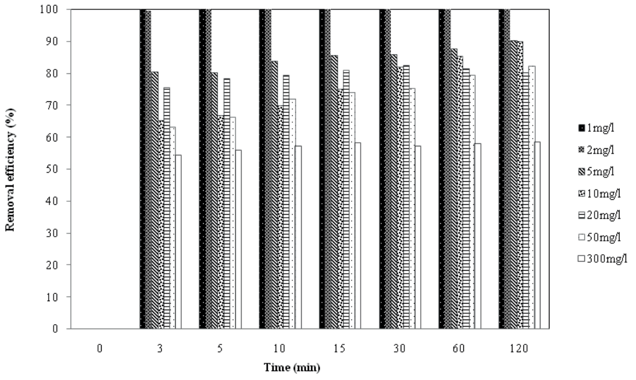

Fig. 4. Effect of initial Cd concentration on biosorption of Cd onto Mosp (pH = 6.8, biosorbent dose = 2 g/L).

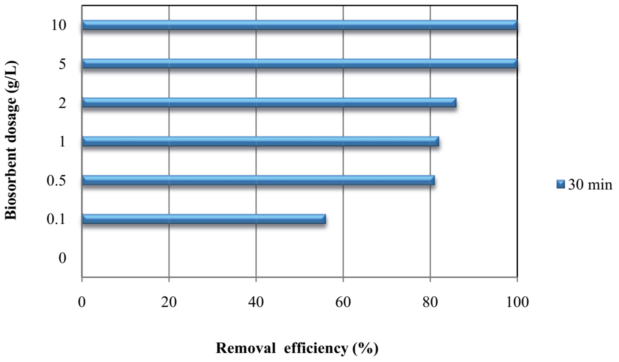

Fig. 5. Effect of biosorbent dose on the biosorption of Cd by Mosp (pH = 6.8, Cd concentration = 5 mg/L).

constant related to the free energy of sorption equilibrium concentration of Cd in solution (mg/L) after biosorption. Maximum biosorption capacities of different biosorbents for cadmium are shown in Table 2.

From the plots between (C_e/q_e) and C_e, we can calculate the slope ($1/q_{\max}$) and the intercept ($1/b$). As seen in Figs. 6 and 7, Freundlich model is better fitted than Langmuir model. Freundlich isotherm assumes the heterogeneous adsorption surface and the possibility of multilayer adsorption.

3.7. Biosorption kinetics and modeling

The biosorption kinetic is important in the aqueous solution treatment, as it presents valuable insights into reaction and mechanisms of biosorption reactions. The experimental sorption kinetics were defined by using pseudo-first-order and pseudo-second-order kinetics. These kinetics can be characterized in their non-linear forms, as follows:

Table 2
Maximum biosorption capacities of different biosorbents for cadmium

$q_{\max}$ (mg/g)	Biosorbent	Reference
126.55	Mosp	In the present study
15.65	Untreated coffee grounds	[53]
6.94	Ulmus leaves	[54]
285.7	Green coconut shell	[55]
48.78	Loquat leaves	[56]
8.58	Rice husks	[57]
46.1	Sugar beet pulp	[58]

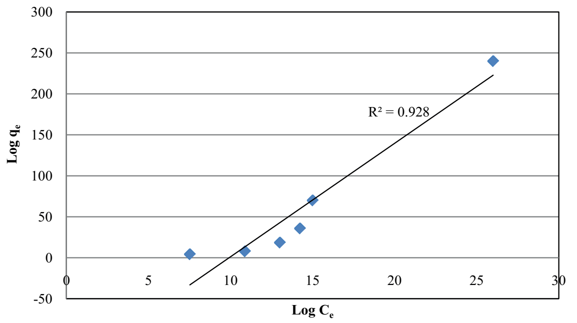

Fig. 6. Freundlich isotherm investigation of Cd biosorption by Mosp.

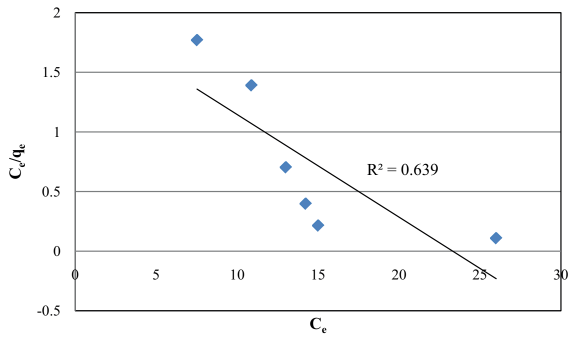

Fig. 7. Langmuir isotherm investigation of Cd biosorption by Mosp.

Pseudo-first-order model:

$$\log(q_e - q_t) = \log q_e \frac{K_{1,ads}}{2.303} t \tag{7}$$

Pseudo-second-order model:

$$\frac{t}{q} = \frac{1}{q_e^{\,2} K_{2,ads}} + \frac{1}{q_e} t \tag{8}$$

where q_e is the mass of solute sorbed at equilibrium (mg/g); q_t the mass of solute sorbed at time t (mg/g); K_1 the first-order equilibrium rate constant (g/g/min); and K_2 is

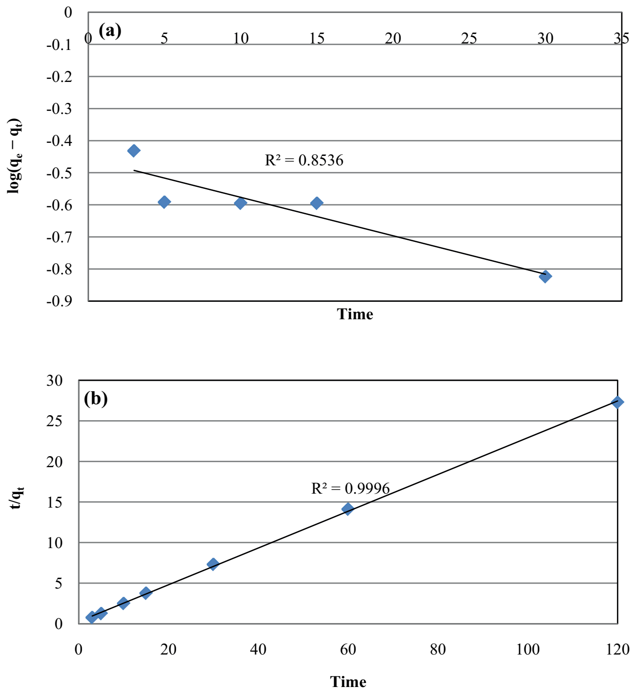

Fig. 8. (a) Pseudo-first-order model and (b) pseudo-second-order model of Cd biosorption by Mosp.

Table 3
Biosorption kinetic parameters of Cd onto Mosp

Model	Parameter	Value
First-order kinetic	q_e (mg/g)	0.4566
	K_1 (1/min)	0.027
	R^2	0.8536
Second-order kinetic	q_e (mg/g)	0.2805
	K_2 (1/min)	0.18
	R^2	0.9996

the second-order equilibrium rate constant (g/g/min). The first-order equilibrium rate constant (K_1) for Cd biosorption was calculated from the slop of the linear plot of log(q_e − q_t) vs. time. In the case of order equilibrium rate constant (K_2), kinetic data were plotted between t/q_t against time. As shown in Fig. 8, it can be concluded that Cd sorption onto Mosp activated carbon appears to be pseudo-second order. Sorption kinetic parameters for Cd biosorption onto Mosp are shown in Table 3.

4. Conclusion

This study showed that Mosp is an excellent biodsorbent for the removal of Cd from aqueous solutions. The highest removal efficiency was 100% and was reached only after a few minutes contact time in the optimum operation condition. The removal efficiency was depended on the initial Cd concentration, pH, activation temperature, contact time, and the biosorbent dosage. Finally, it can be concluded that the Mosp can be used as an effective with considerable biosorption capacity, renewable, cost benefit, and environmental friendly biosorbent for the Cd removal from aqueous solution especially industrial effluent containing high level of Cd.

Acknowledgment

The authors are grateful to the Bushehr University of Medical Sciences for their financial support.

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