



# Promising applications of seedcake of *Jatropha curcas* plants: bioethanol production and bio-sorbent material for dye and heavy metal removal

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## Abstract

*Jatropha curcas* L. as a member of Euphorbiaceae family had many copious applications due to its richness content of oil, cellulose, hemicellulose, and lignin. Therefore, this work was directed to increase the value of the seedcakes of *Jatropha curcas* by their applications in bioethanol production and removal of methylene blue dye (MB) as well as hexavalent chromium Cr (VI) from contaminated wastewater. The seedcakes sources were collected from different irrigation treatments: tap water (1), sewage water (2), sewage water and sludge (3), sewage water and sludge sprayed with tap water, 100, 200, 300, and 400 ppm of chlorocholine chloride (cycocel or abbreviated as CCC), (4), (5), (6), (7), and (8), respectively. Results showed that sewage water only (2) and the combination between sewage water and sludge irrigation (3) produced the highest ethanol quantity (4.59 & 5.15 ml/l, respectively) after the fermentation of the hydrolysates by *Candida tropicalis* Y-26. In respect to CCC spraying, the maximum ethanol concentration (5.57 ml/l) was achieved from *Jatropha curcas* seedcake irrigated with CCC at a concentration of (300 ppm). In the other application, a preliminary screening experiment was demonstrated for the different forms of *Jatropha curcas* seedcake and results revealed that the *Jatropha curcas* seedcake irrigated with sewage water only (2) achieved the highest removal efficiencies of 78.8% and 41% for MB and Cr (VI), respectively. After that, the optimization process stated the optimum parameters; *Jatropha curcas* seedcake dose of 15 and 5 g/l, conc. 50 ppm, pH 7 and 5, and contact time 240 & 120 min were the optimum factors that achieved high (methylene blue (MB)) and hexavalent chromium [Cr (VI)] removal efficiencies, respectively. Langmuir isotherm and the pseudo-first-order model were stated as selected models that explain the adsorption mechanism of MB and Cr by *Jatropha curcas* seedcake.

**Keywords** Bioethanol · Cycocel · *Jatropha curcas* · Hexavalent chromium · Seedcake · Sewage water · Sewage sludge · Methylene blue

## 1 Introduction

*Jatropha curcas* L. is a member of Euphorbiaceae family, which originated in Central America and Mexico and after that were established in other various tropical and subtropical areas. In the past, it was usually used for medicinal purposes, although lately *Jatropha* oil was used

for biodiesel production as it produces around 32–40% valuable oil, which could be supportive in biofuel industry. *Jatropha curcas* was characterized by its high productivity especially when irrigated by wastewater; in addition, it can be cultivated and grown in harsh conditions [1]. Furthermore, seedcake waste is produced from *Jatropha* seeds after the oil extraction process, where one tone of *Jatropha* oil production is approximately equivalent to nearly 3 tons of seedcakes that require a proper disposal [2]. The seedcake is characterized by their considerable percentages of cellulose, hemicellulose, and starch. Accordingly, it has been stimulated and used in the production of bioethanol [3]. Bioethanol production from *Jatropha* lignocellulosic biomass can be achieved via three principal steps: (a) the pretreatment of biomass, (b) the hydrolysis of polysaccharides to induce simple sugars, (c)

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the fermentation of simple sugars for production of ethanol [4, 5]. Additionally, world nowadays faces a significant problem of water pollution due to the rapid industrialization and the discharging of their effluents into waterways. For instance, hexavalent chromium Cr (VI) as a type of heavy metals that is used in many industries was reported by the World Health Organization (WHO) as a carcinogenic form of heavy metals [6]. Exposure to the Cr (VI) for long time leads to a lot of health problems for humans, e.g., nausea, irritant, sever liver and kidney damage, damage to nervous and circulatory tissue, lungs, and digestive tract cancer [7]. Consequently, WHO reported the value of the presence of the chromium in wastewater as not exceed 0.05 ppm [8].

Moreover, color impurity in industrial effluents from dye manufacturing and dye consuming industries threatens the human health and the environment. For example, methylene blue (MB) is a cationic dye that was commonly used as a coloring agent in the textile industries and may help in oncological surgeries. As a result, those industries discharge MB into water streams that in follow cause various problems [9]. Copious conventional chemical and physical treatment methods were applied for heavy metals and dyes removal from the contaminated water, e.g., ion exchange [10] ultra-filtration, coagulation, and chemical oxidation [11]. However, these technologies were widely used; they faced some limitations such as their high cost and their low efficiency at low concentration removal. Hence, finding a low-cost and efficient method for safe disposal of heavy metals and dyes from wastewater such as biosorption becomes an urgent priority [12]. Different biomasses as ecofriendly materials (agricultural wastes, and microorganisms) were applied as bio-sorbent materials for pollutants removal from polluted wastewater [13–17]. For instance, richness of the *Jatropha curcas* seedcake with lignocellulosic biomass has recommended its use as a bio-sorbent material for the removal of dyes and heavy metals from polluted wastewater. Therefore, agriculture wastes can serve as an effective alternative for the usually used costly chemical sorbents and consequently solve the problem of its safe disposal [18]. For instance, *Jatropha curcas* seedcake contains toxic compounds and is not edible, and consequently, there is no worry about food security [2]. Furthermore, most of the used sorbents were applied with particle sizes around 100  $\mu\text{m}$ . However, there is a high need for further smaller sizes, represented by higher surface area, and is still in need [19]. Therefore, this study was directed to collect different *Jatropha curcas* fruits that were irrigated by different water sources as tap water (1), sewage water (2), sewage water and sludge (3), sewage water and sludge sprayed with tap water (4), and different concentrations of CCC (100, 200, 300, and 400 ppm) (5), (6), (7), and (8), respectively. Afterwards, *Jatropha curcas* seeds were used after oil extraction and drying as *Jatropha curcas* seedcakes in many applications to increase their value.

The first application was studying the probability of different *Jatropha curcas* seedcakes in bioethanol production via hydrolysis and fermentation processes. Afterwards, the second application is the studying in the wastewater-pollutants removal (dye and heavy metal removal) after grinding to relatively nano-sizes. Precisely, methylene blue (MB) and chromium (Cr VI) were used in this study. The selected *Jatropha curcas* seedcake will be optimized through two optimization steps, One Factor at A time (OFAT) experiments as a preliminary experiment which after that the selected levels will be further applied in the  $2^3$  general full-factorial design. The experimental data were analyzed by Langmuir and Freundlich isotherms and kinetics studies. At the end, the selected *Jatropha curcas* seedcake will be characterized using FT-IR analysis and DLS.

## 2 Materials and methods

### 2.1 Collection and preparation of *Jatropha curcas* seedcakes

*Jatropha curcas* fruits were collected from plantations with different irrigation treatments at the drinking and sewage water treatment station in Al-gabal Al-Asfar, Cairo, Egypt. The irrigation treatments were tap water (1), sewage water (2), sewage water and sludge (3), sewage water and sludge sprayed with tap water (4), and different concentrations of CCC (100, 200, 300, and 400 ppm) (5), (6), (7), and (8), respectively. The fruits were cracked to remove the shell and the seeds were ground and put in soxhlet extractor for oil extraction by petroleum ether 40:60%. The remaining cake in the thimble was recovered and oven dried. *Jatropha curcas* seedcakes were initially crushed to small pieces using a mortar and soaked overnight in water to remove residual dirt and oil using cheesecloth and floating, respectively. Afterwards, the seedcakes were dried at 60 °C for 48 h to make the weight constant. Finally, they were blended and sieved through a 0.5-mm mesh sieve. The pretreated dried seedcakes were stored at room temperature in covered plastic bottles for the subsequent analyses.

### 2.2 Bioethanol production from *Jatropha curcas* seedcakes

#### 2.2.1 Acid hydrolysis

About 10 g of the different pretreated *Jatropha curcas* seedcakes were mixed separately in an Erlenmeyer flask (250 ml) with 100 ml of 4%  $\text{H}_2\text{SO}_4$ . Each flask was closed to prevent evaporation of acid during autoclaving, for 20 min at 120 °C. The flask was filtered after hydrolysis to remove the unhydrolyzed materials. The filtrate was then neutralized and centrifuged at 10,000 rpm for 10 min. Finally, the

concentrations of total reducing sugars (TRS) in the filtrate (hydrolysate) were determined.

### 2.2.2 Fermentation

The yeast strain *Candida tropicalis* (Y-26) and *Saccharomyces cerevisiae* (Y-39) were used to ferment *Jatropha curcas* seedcake hydrolysates as well as to maximize the ethanol production. These isolates were previously characterized for their ability to ferment various types of sugars (pentose and hexose) for bioethanol production [20]. Yeast suspension for inoculation was prepared by growing the organism on Wickersham's (WH) medium at 30 °C for 48 h in a shaking incubator at 150 rpm. The fermentation process was performed under oxygen-limited conditions in 100-ml blue cap flasks, containing 50 ml of fermentation media: the hydrolysate obtained from the acid hydrolysis of *Jatropha curcas* seedcakes that supplemented with peptone; 10 g/l,  $\text{KH}_2\text{PO}_4$ ; 2 g/l and  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ ; 1 g/l. The pH value for each flask was adjusted to 5.5. After that, the air-locked flasks were sterilized for 20 min at 120 °C. Afterwards, these flasks were eventually inoculated with yeast strain ( $\approx 1 \times 10^5$  cells/ml). The flasks were then incubated at 30 °C for 48 h. In this study, all analyses were performed in triplicate, and the average was recorded.

### 2.2.3 Determination of total reducing sugars

The total reducing sugars (TRS) were estimated by "3,5-dinitro salicylic acid (DNS)" method [21]. Change in color intensity was measured at 540 nm, using UV/VIS/NIR spectrophotometer (Model Jasco V-570). Glucose was used as a standard for the calibration curve.

### 2.2.4 Determination of ethanol concentration

The ethanol concentrations were measured using gas chromatography (6890, Agilent G1530A, USA). The capillary column with a dimension of  $60 \mu\text{m} \times 5.00 \mu\text{m} \times 530 \mu\text{m}$  was used. The helium (gas carrier) was used with a flow rate of 25 ml/min. The oven temperature and the detector were attempt 300 °C.

## 2.3 Application of the *Jatropha curcas* seedcakes in the removal of methylene blue (MB) and hexavalent chromium (Cr VI)

### 2.3.1 The dried seedcake biomass grinding

The dried bio-sorbent materials (*Jatropha curcas* seedcakes) were ground using "Planetary Ball Mill, PM 400 4" grinding stations" to obtain high sorbent surface area.

### 2.3.2 Batch experiments

A preliminary screening experiment was implemented to determine the best treatment, tap water (1), sewage water (2), sewage water and sludge (3), sewage water and sludge sprayed with tap water (4), and different concentrations of CCC (100, 200, 300, and 400 ppm) (5), (6), (7), and (8), respectively, that gives the highest MB and Cr (VI) removal efficiencies. This treatment will be used for further process. Equation (1) was applied to calculate the removal efficiencies through the experiments.

$$\text{Removal efficiency}(\%) = \frac{C_o - C_e}{C_o} \times 100 \quad (1)$$

### 2.3.3 Optimization process

The optimization process for the MB and Cr (VI) removal efficiencies (MBRE and CRE, respectively) was performed through two optimization steps. The first step was achieved with a series of levels of OFAT experiments in which one factor levels were varied, while the other factors were kept constant at a specific level. The factors that were applied for the OFAT experiments were pH with values of 3, 5, 7, 9, and 11; MB and Cr (VI) concentrations of 50, 100, 150, 200, and 300 ppm; and *Jatropha curcas* seedcake doses of 0.25, 0.5, 1.0, and 1.5 g/100 ml. After that, preliminary experiments were used to determine the low/high (−1/+1) levels of each factor were selected to further implemented in the full-factorial design experiment.

Factorial experiments helped to obtain the maximum removal efficiency with minimal number of experiments by studying the interaction effect between the factors. The full-factorial model of 8 runs for every pollutant; MB and Cr (VI). Tables 2 and 3 represents the applied parameters for MB and Cr (VI) removal efficiency improving.

### 2.3.4 Equilibrium studies

The mechanism of MB and Cr (VI) sorption by *Jatropha curcas* seedcake was determined by applying the linear Langmuir and Freundlich isotherms which were calculated according to Langmuir and Freundlich isotherms as previously reported [22, 23].

Langmuir isotherm

$$\begin{aligned} \frac{C_e}{Q_e} &= \frac{C_e}{Q_{\max}} + \frac{1}{bQ_{\max}} \frac{C_e}{Q_e} = \frac{C_e}{Q_{\max}} \\ &+ \frac{1}{b \times Q_{\max}} C_e/Q_e = C_e/Q_{\max} \\ &+ 1/bQ_{\max} \frac{C_e}{Q_e} = \frac{C_e}{Q_{\max}} + \frac{1}{bQ_{\max}} \frac{C_e}{Q_e} \\ &= \frac{C_e}{Q_{\max}} + \frac{1}{bQ_{\max}} \end{aligned} \quad (2)$$

where  $Q_e$   $q_e$  is the amount of dyes/metal adsorbed per unit mass onto the adsorbent material(mg/g),  $Q_{\max}$  is maximum adsorption capacity of the adsorbent material (mg/g), and  $b$  is Langmuir constant that relates to the heat of adsorption (l/mg).

Freundlich isotherm

$$\text{Log}q_e = \text{Log}K_f + \frac{1}{n}\text{Log}C_e \quad (2)$$

where  $q_e$  is the equilibrium adsorption capacity (mg/g),  $C_e$  is the equilibrium concentration of dyes/metal in the solution (mg/l), and  $K_f$  and  $n$  are the Freundlich constants that expressed the adsorption limit and adsorption density, respectively.

### 2.3.5 Kinetics studies

The adsorption process kinetics aimed to the rate of MB and Cr (VI) biosorption by the selected *Jatropha curcas* seedcake, which controls equilibrium time. The kinetics model included the pseudo-first-order and second-order equations [24, 25]. The pseudo-first-order model was evaluated using the following equation

$$\text{Ln}(q_e - q_t) = \text{Ln}q_e - K_1 t \quad (4)$$

where  $q_t$  and  $q_e$  are the measure of dyes/metal adsorbed at time  $t$  and equilibrium (mol  $g^{-1}$ ) respectively and  $k_1$ ( $\text{min}^{-1}$ ) is the rate constant for this model.

Whereas the rate of the pseudo-second-order model was studied using the following equation:

$$t/q_t = t/q_e + \frac{1}{K_2 q_e^2} \quad (5)$$

where  $k_2$  is the rate constant of sorption for this model ( $g \text{ mol}^{-1} \text{ min}^{-1}$ ) and  $q_e$  and  $q_t$  are the quantities of adsorbed dyes/metal at equilibrium and at time  $t$  (mol  $g^{-1}$ ), respectively.

## 2.4 Characterization of the selected *Jatropha curcas* seedcakes

“Fourier transform infrared (FT-IR)” spectroscopy “Nicolet IS-10 spectroscopy” was accustomed in this study to achieve the bio-sorbent (*Jatropha curcas* seedcake) functional groups. The selected *Jatropha curcas* seedcake was applied using KBr disks, within the wavelength range of 400–4000 nm. Afterwards, spectra plot was plotted for the samples. Additionally, *Jatropha curcas* seedcake size was determined using “dynamic light scattering (DLS), Zetasizer Nano Series HT, Nano-25, and Malvern.”

## 3 Results and discussions

### 3.1 Bio ethanol production from *Jatropha curcas* seedcakes

#### 3.1.1 Hydrolysis of *Jatropha* seedcakes

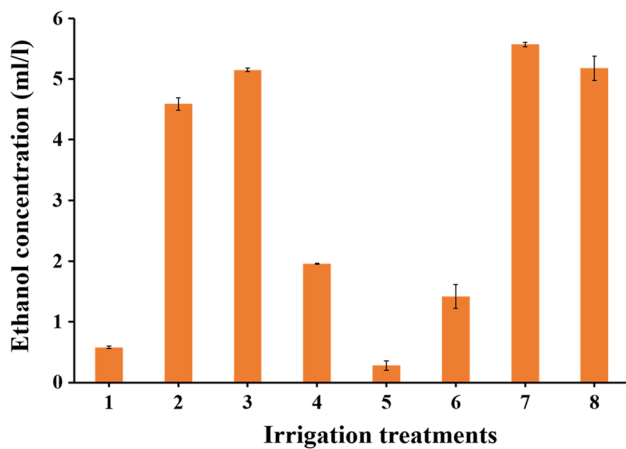
The pretreated *Jatropha curcas* seedcakes with different irrigation treatments were successfully hydrolyzed using 4%  $H_2SO_4$ . Dilute acid hydrolysis by  $H_2SO_4$  is considered the best hydrolysis method in releasing of the TRS from *Jatropha curcas* seedcakes [3, 26]. TRS estimation gives an indication for the total liberated simple sugars after the hydrolyses process of carbohydrate fractions (cellulose and hemicellulose). The results reported in Table 1 showed that *Jatropha curcas* seedcake that irrigated with sewage water significantly increased the value of TRS ( $6.7 \pm 0.3$  g/l). Moreover, adding sludge to sewage water irrigation improved the TRS concentration ( $7.9 \pm 0.1$  g/l) compared to its control ( $1.8 \pm 0.2$  g/l). This could be attributed to the remarkable amount of organic and inorganic minerals in sewage water and sludge, which enhanced and improved the photosynthetic apparatus. In consequence, the carbohydrate fractions were significantly increased and, in follow, TRS concentration increased [27].

Regarding, the hydrolysis of *Jatropha curcas* seedcakes irrigated with CCC, at concentrations of CCC 300 and 400 ppm, gave the highest TRS values ( $9.8 \pm 0.05$  &  $9.6 \pm 0.6$  g/l, respectively). These results might be due to the increase in the levels of total sugars contents at such concentration (300 ppm) of CCC [28]. Similar increased levels of sugar contents under high CCC concentrations were observed in mustard leaves [28] and mulberry leaves [29]. CCC affected positively on the photosynthetic pigments and

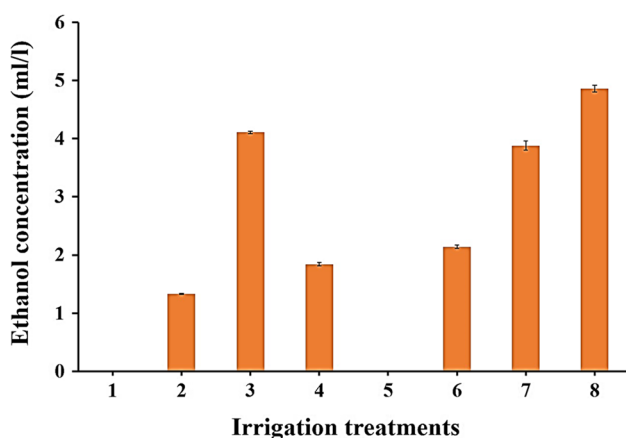
**Table 1** Effect of irrigation with tap water, sewage water, sewage water+sludge and sewage water+sludge sprayed with tap water, 100, 200, 300, and 400 ppm of CCC on total reducing sugars (TRS) produced from acid hydrolysis of *Jatropha* seedcakes by  $H_2SO_4$  (4%)

Treatments	Total reducing sugars (TRS) g/l
Tap water	$1.8 \pm 0.2^g$
Sewage water	$6.7 \pm 0.3^f$
Sewage water + sludge	$7.9 \pm 0.1^c$
Sewage water + sludge sprayed with tap water	$7.7 \pm 0.3^d$
Sewage water + sludge sprayed with different concentrations of CCC (ppm)	100 $1.1 \pm 0.1^h$ 200 $7.3 \pm 0.4^e$ 300 $9.8 \pm 0.05^a$ 400 $9.6 \pm 0.6^b$

Columns with different letters are significantly different at  $p < 0.05$



**Fig. 1** Effect of different irrigation treatments; tap water (1), sewage water (2), sewage water and sludge (3), sewage water and sludge sprayed with tap water (4) and different concentrations of CCC (100 (5), 200 (6), 300 (7), and 400 (8) ppm) on ethanol concentrations from the fermentation of *Jatropha curcas* seedcake hydrolysates by *Candida tropicalis* Y-26. Bars indicate  $\pm$  SE



**Fig. 2** Effect of different irrigation treatments; tap water (1), sewage water (2), sewage water and sludge (3), sewage water and sludge sprayed with tap water (4) and different concentrations of CCC (100 (5), 200 (6), 300 (7), and 400 (8) ppm) on ethanol concentrations from the fermentation of *Jatropha curcas* seedcake hydrolysates by *Saccharomyces cerevisiae* Y-39. Bars indicate  $\pm$  SE

photosynthetic activity which was in a strong relation with the increased levels of carbohydrates and TRS [30].

In contrast, TRS from the hydrolysis of *Jatropha curcas* seedcakes irrigated with low CCC concentrations (100 and 200 ppm) decreased than their control.

### 3.1.2 Fermentation of *Jatropha* seedcake hydrolysate

TRS produced from different *Jatropha* seedcake samples after  $H_2SO_4$  hydrolysis were fermented by two yeast strains for bioethanol production (Figs. 1 and 2). This

study successfully produced bioethanol from *Jatropha* seedcake which in agreement the previously reported study [31].

- Fermentation by *Candida tropicalis* Y-26

The data recorded in Fig. 1 displayed the ethanol concentrations produced from the fermentation of *Jatropha* seedcake hydrolysates by *Candida tropicalis* Y-26. The bioethanol concentrations were significantly advanced with the irrigation treatment with sewage water (4.59 ml/l) either alone (2) or with sludge (3) (5.15 ml/l) in comparison to the control that irrigated with tap water (1). For the CCC spray, the maximum ethanol concentration (5.57 ml/l) was achieved from *Jatropha curcas* seedcake irrigated with CCC at a concentration of (300 ppm) followed by 400 ppm compared to their control. While the reverse was true at low CCC concentrations (100 & 200 ppm).

- Fermentation by *Saccharomyces cerevisiae* Y-39

Compatible to the fermentation of *Jatropha* seedcakes by *Candida tropicalis* Y-26, the fermentation by *Saccharomyces cerevisiae* Y-39 verified that the irrigation treatment with the sewage water (2) and the sewage water and sludge (3), significantly increased the yield of ethanol (1.33 & 4.11 ml/l) than their control. In respect to the irrigation treatment with CCC concentrations, it was noticed that concentrations of 400 followed by 300 ppm of CCC significantly increased ethanol quantity (4.86 & 3.88 ml/l). On the other hand, concentrations of 100 and 200 ppm of CCC displayed a negatively effect on the ethanol production especially at 100 ppm of the CCC which recorded zero ethanol concentration when fermented by *Saccharomyces cerevisiae* Y-39) see Fig. 2).

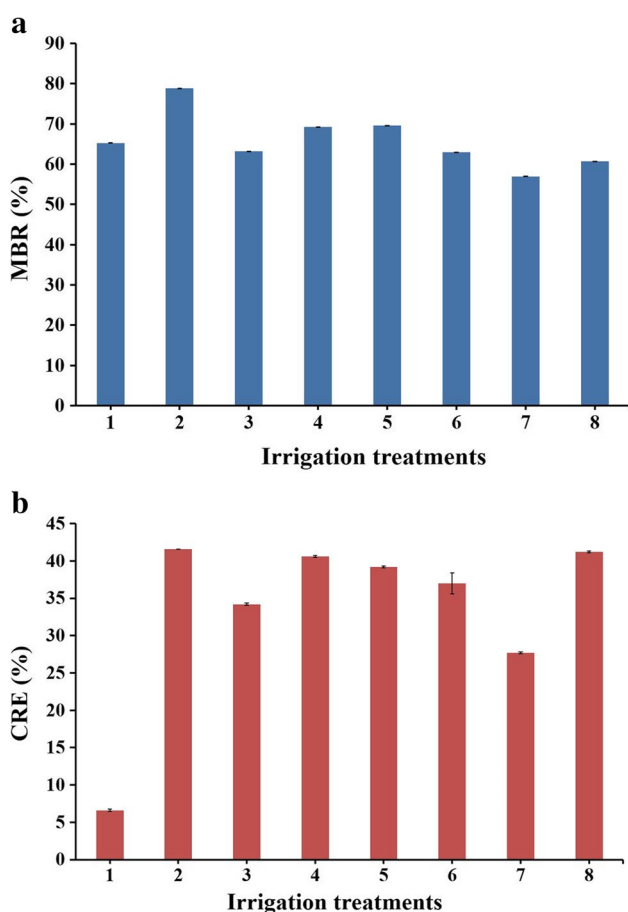
The bioethanol yield from the fermentation of *Jatropha* seedcakes by *Candida tropicalis* Y-26 and *Saccharomyces cerevisiae* Y-39 was higher in high CCC concentrations 300 or 400 ppm than other irrigation treatment. In addition, the highest ethanol concentrations were achieved from *Jatropha curcas* seedcake fermented by *Candida tropicalis* Y-26 than *Saccharomyces cerevisiae*.

It has been reported that the bioethanol yields relayed on the selection of ideal substrate and microbial strain [32]. Accordingly, *Candida tropicalis* was used as the best microbial strain for fermentation and ethanol production from different lignocellulosic wastes [33–35]. Furthermore, Baki and Bello [31] demonstrated that *Jatropha curcas* is the most suitable seedcakes for sugars and ethanol production than other cakes such as *Racinus communis*, *Azadirachta indica*, and *Lagenaria siceraria* seedcakes.

## 3.2 Application of the *Jatropha curcas* seedcakes in the removal and recovery of methylene blue (MB) and chromium (Cr VI)

### 3.2.1 Preliminary screening experiment

The preliminary screening experiment was implemented to determine the best treatment that gives the highest MB and Cr (VI) removal efficiencies. Preliminary screening experiment stated that applying 10 g/l dose of *Jatropha curcas* seedcakes irrigated with sewage water (2) achieved the maximum MBRE of (78.8%) and CRE (41%), at 50 ppm concentration, and 45, 180-min contact time, respectively (see Fig. 3a,b).



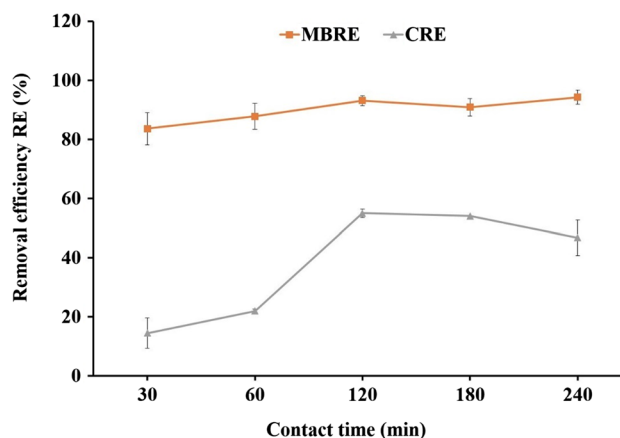
**Fig. 3** **a** Methylene blue (MB) removal efficiency of the selected *Jatropha curcas* seedcakes that irrigated by sewage water (2), sewage water and sludge (3), sewage water and sludge sprayed with tap water (4) and different concentrations of CCC (100 (5), 200 (6), 300 (7), and 400 (8) ppm) compared to control tap water (1). Bars indicate  $\pm$  SE. **b** Hexavalent chromium Cr (VI) removal efficiency of the selected *Jatropha curcas* seedcakes that irrigated by sewage water (2), sewage water and sludge (3), sewage water and sludge sprayed with tap water (4), and different concentrations of CCC (100 (5), 200 (6), 300 (7), and 400 (8) ppm) compared to control tap water (1). Bars indicate  $\pm$  SE

### 3.2.2 OFAT experiments

Preliminary experiments stated the effectiveness of treat no. 2 (*Jatropha curcas* seedcakes irrigated with sewage water), and after that, the treat was optimized to give high MB and Cr (VI) removal efficiency. Various factors: time, pH, *Jatropha curcas* seedcake dose, MB, and Cr (VI) concentrations, were studied through the two steps optimization to achieve higher removal capacity of *Jatropha curcas* seedcakes and the results can be discussed as follows:

**Effect of contact time** The results (Fig. 4) exhibited that the MB and Cr (VI) removal efficiencies (MBRE & CRE) were increased with increasing contact time. The optimum contact time was 4 h with the maximal removal efficiency of 94.26% for MBRE. The maximum CRE of 54.01% was achieved after 2-h contact time. These results indicated that the contact surface *Jatropha curcas* seedcake was fully saturated after 4 h and 2 h for MBRE and CRE, respectively. Additional increase in the contact time has displayed no effect with a relation to the availability of active sites and the equilibrium [36, 37].

**Effect of pH** The data presented in Fig. 5 indicated that the removal efficiencies of the *Jatropha curcas* seedcake were increased from 3 to 7 and from 3 to 5 towards MB and Cr (VI), respectively. For instance, maximum MBRE of 42.26% was obtained at pH 7, whereas higher CRE of 65.08% was observed at pH 5. It has been previously reported that the removal efficiencies were effectively increased by further increase in the pH values [38, 39]. At low pH values, the higher proton ( $H^+$ ) concentration was increased in the solution. This proton competes with metal ions by making relative bonds with the bio-sorbent active sites [40]. While by increasing the pH values, the number of protons decreases



**Fig. 4** Effect of contact time intervals on the MBRE and CRE by the selected *Jatropha curcas* seedcake. Bars indicate  $\pm$  SE

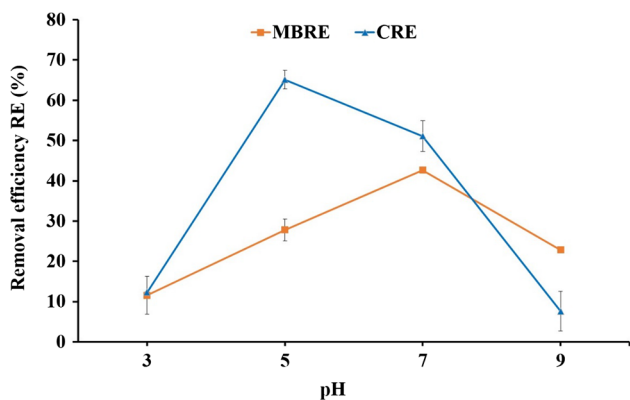


Fig. 5 Effect of different pH on the MBRE and CRE by the selected *Jatropha curcas* seedcake. Bars indicate ± SE

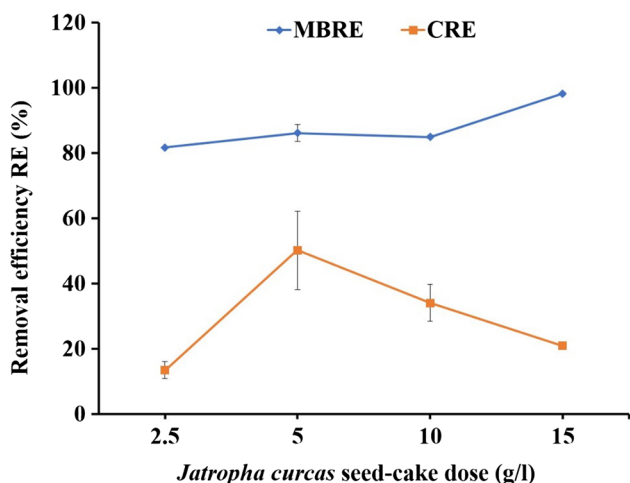


Fig. 6 Effect of different the selected *Jatropha curcas* seedcake doses on MBRE and CRE. Bars indicate ± SE

and the competition between the protons and Cr (VI) ion or MB on the active sites of the bio-sorbent materials decreases.

**Effect of *Jatropha curcas* seedcake dose** Results (Fig. 6) displayed that 15 g/l was the most effective dose of the *Jatropha curcas* seedcake for the MBRE of 98.2%. On the other hand, 5 g/l of *Jatropha curcas* seedcake dose was the optimum dose to achieve the highest CRE of 50.18%. Further increase in the *Jatropha curcas* seedcake dose decreased the efficiency of the removal. The adsorption capacity increases with increasing the bio-sorbent doses were previously reported [41]. The interpretation for this attempt was the higher bio-sorbent dose offered a greater surface area which leads to increasing in the removal efficiencies [41].

**Effect of methylene blue and hexavalent chromium Cr (VI) concentration** Results in Fig. 7 demonstrated a slight decrease in the MBRE and CRE by increasing the MB and Cr

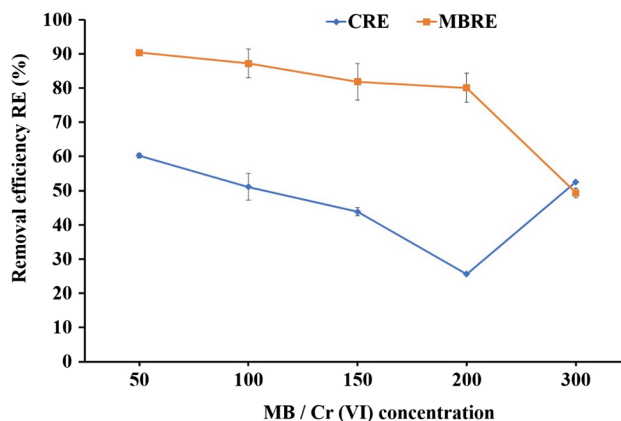


Fig. 7 Effect of different MB and Cr (VI) concentrations on MBRE and CRE by the selected *Jatropha curcas* seedcake. Bars indicate ± SE

Table 2 Coded factors and the selected levels used in the full-factorial optimization mode of MB

Factor	Unit	Symbol	Statistical code	Values of coded levels	
				Low (-1)	High (+1)
Time	Min	Time (min)	A	30	60
Dose	g/l	Dose (g/l)	B	2.5	15
MB conc	Ppm	Conc. (ppm)	C	50	300

Table 3 Coded factors and the selected levels used in the full-factorial optimization mode of Cr (VI)

Factor	Unit	Symbol	Statistical code	Values of coded levels	
				Low (-1)	High (+1)
Time	min	Time (min)	A	30	180
Dose	g/l	Dose (g/l)	B	2.5	5
Cr (VI) conc	ppm	Conc.(ppm)	C	50	200

(VI) concentrations from 50 to 200 ppm. After that, a sharp decrease was noticed in the CRE after 200 ppm, whereas the MBRE increased up to nearly 60%. This result may be attributed to the higher pollutant concentrations that leads to form collisions between the ions and the bio-sorbent material, which hence decrease the adsorption capacity [41, 42].

3.2.3 Full-factorial optimization model

Low and high levels of each factor were chosen from the optimum level (see Tables 2 and 3) and applied in the full-factorial design model. The full-factorial design experiments with the MBRE and the CRE were reported in Tables 4 and 5. The main and interaction effects, the Pareto chart, and the normal probability plot were elucidated as follows:

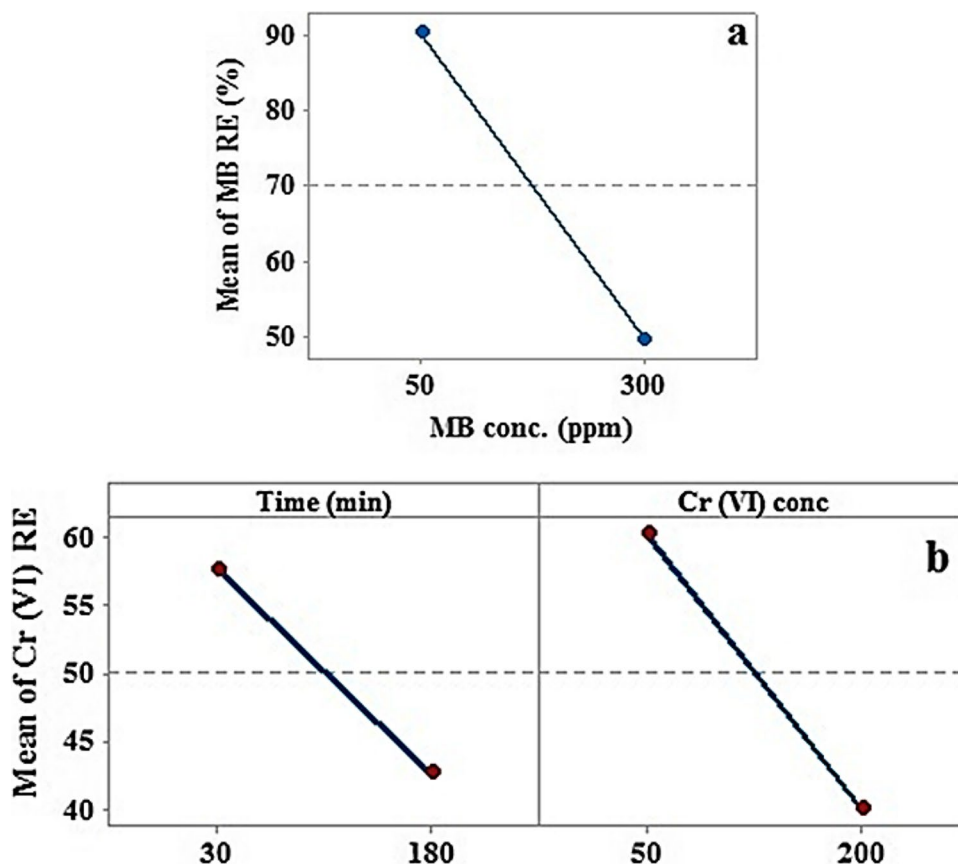
**Table 4** The design matrix and the results of the 2<sup>3</sup> full-factorial design of MB

Std. order	Run order	Pt type	Blocks	Time (min)	Dose (g/l)	MB conc. (ppm)	MBRE (%)	FITS1	RES11
3	1	1	1	30	15	50	90.13	90.53	-0.40
6	2	1	1	60	2.5	300	53.53	49.46	4.07
1	3	1	1	30	2.5	50	84.78	90.53	-5.75
4	4	1	1	30	15	300	87.63	49.46	38.16
7	5	1	1	60	15	50	94.23	90.53	3.70
2	6	1	1	30	2.5	300	50.38	49.46	0.92
8	7	1	1	60	15	300	6.29	49.46	-43.16
5	8	1	1	60	2.5	50	92.99	90.53	2.45

**Table 5** The design matrix and the results of the 2<sup>3</sup> full-factorial design of Cr (VI)

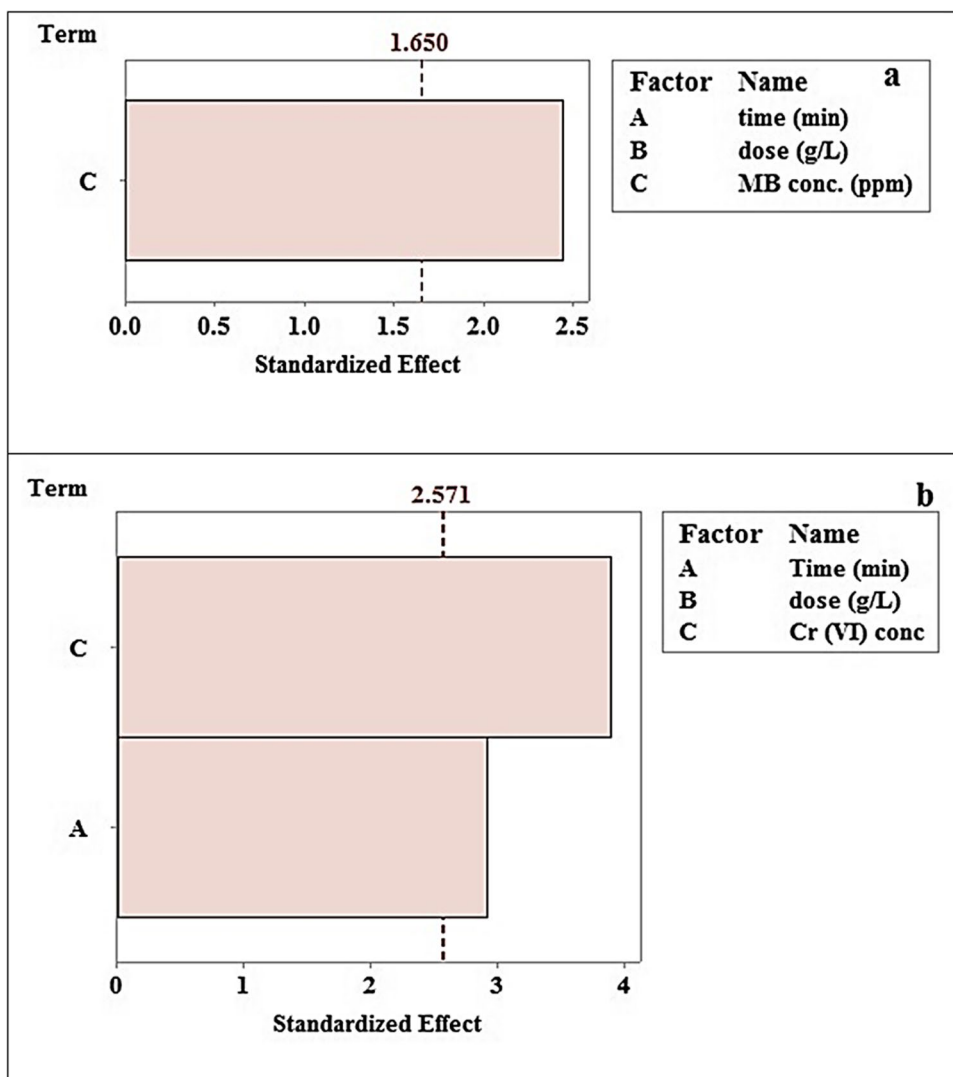
Std. order	Run order	Pt type	Blocks	Time (min)	Dose (g/l)	Cr (VI) conc. (ppm)	Cr (VI) RE (%)	FITS1	RES11
3	1	1	1	30	5	50	59.06	67.84	-8.77
6	2	1	1	180	2.5	200	29.02	32.39	-3.36
7	3	1	1	180	5	50	58.01	52.66	5.34
5	4	1	1	180	2.5	50	49.04	52.66	-3.61
1	5	1	1	30	2.5	50	74.89	67.84	7.04
4	6	1	1	30	5	200	55.04	47.57	7.47
2	7	1	1	30	2.5	200	41.83	47.57	-5.73
8	8	1	1	180	5	200	34.02	32.39	1.63

**Fig. 8 a, b.** Main effects plot for the MB removal efficiency and Cr (VI) removal efficiency at the low/high levels for the selected *Jatropha curcas* seedcake





**Fig. 9** Pareto chart of the main and interaction effects for the selected *Jatropha curcas* seedcake



**Main and interaction effects** The main effects are defined as the effects that study the levels deviation of each factor from the high level of the factor to its low level. The effect is positive when the deviation increases from the low level to the high level, whereas the effect is negative when the deviation decreases from the high level to the low level of the factor [43, 44]. Figure 8a and b showed the main effects of the MB and Cr (VI) removal process. Results demonstrated that only the MB concentration induced a negativity effect on the MB removal efficiency, while time and Cr (VI) concentration showed negative effects on the Cr (VI) removal efficiency. Nevertheless, the interaction effect aims

to study the interaction that occurs between two factors and their effect on the removal process, and this type of effect was not noticed through this process.

**Pareto chart** Pareto chart is another method for the main and interaction effects expression where it depended on the Student *t*-test to estimate the significance values of the effects [45, 46]. Figure 9a and b displayed the Pareto chart of both MB and Cr (VI) removal efficiencies where horizontal columns expressed significant of the values of the main and interaction effects. The vertical lines that were known

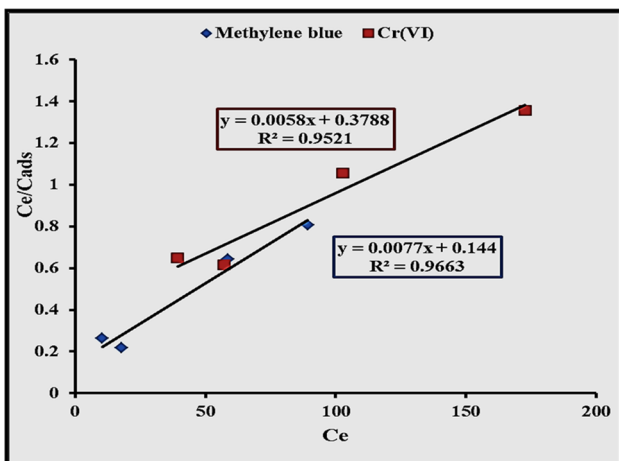
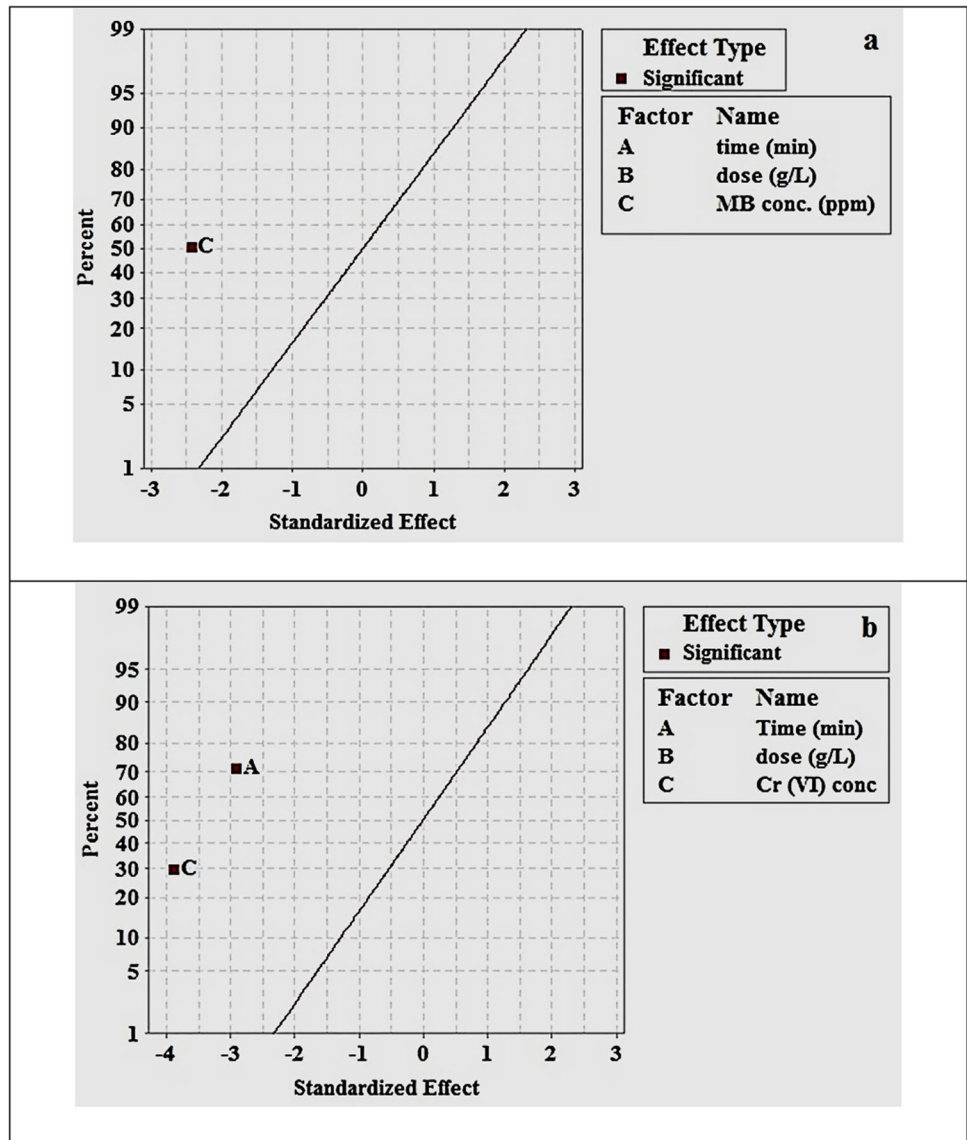
**Table 6** Coded co-efficient of MB

Term	Effect	Coef	SE Coef	T-value	P-value
Constant		70.00	8.40	8.33	0.000
MB conc. (ppm)	-41.07	-20.54	8.40	-2.44	0.050

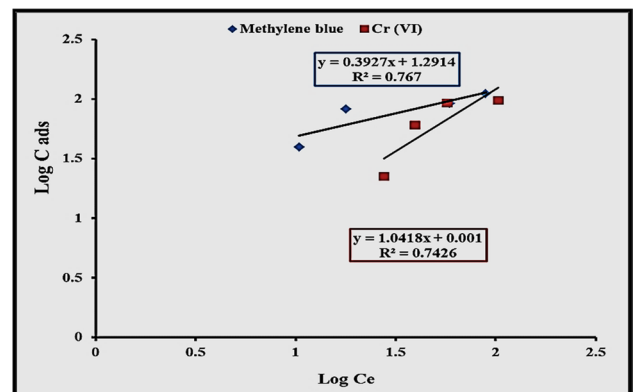
**Table 7** Coded co-efficient of Cr (VI)

Term	Effect	Coef	SE Coef	T-value	P-value
Constant		50.12	2.60	19.25	0.000
Time (min)	-15.18	-7.59	2.60	-2.92	0.033
Cr (VI) conc	-20.27	-10.14	2.60	-3.89	0.011

**Fig. 10** Normal probability plot of the main and interaction effects for the selected *Jatropha curcas* seedcake



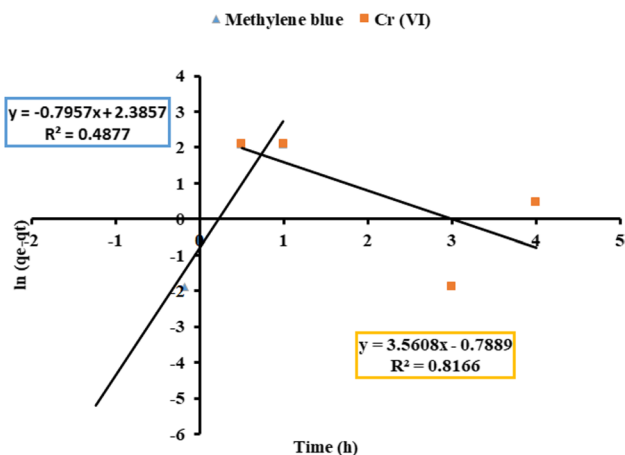
**Fig. 11** Langmuir isotherm linearized plot for the MB and Cr (VI) adsorption by the selected *Jatropha curcas* seedcake



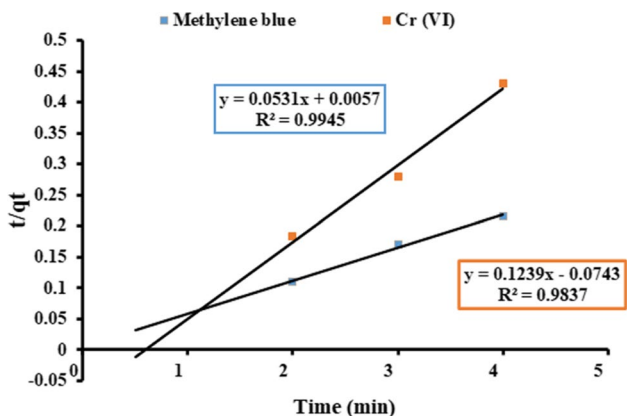
**Fig. 12** Freundlich isotherm plot for MB and Cr (VI) adsorption by the selected *Jatropha curcas* seedcake

**Table 8** Isotherm parameters for both MB and Cr (VI) adsorption

	Langmuir			Freundlich		
	$R^2$	$Q_{max}$	$b$	$R^2$	$k$	$n$
MB	0.96	129.8701	0.053472	0.767	19.5614	2.546473
Cr (VI)	0.95	172.4138	0.015312	0.7426	1.002305	0.960615



**Fig. 13** Pseudo-first-order reaction of the selected *Jatropha curcas* seedcake



**Fig. 14** Pseudo-second-order reaction of the selected *Jatropha curcas* seedcake

also by the reference lines indicated the  $t$ -values which were 1.650 and 2.57 for both MB and Cr (VI), respectively. The MB concentration was the only factor that had a significance

**Table 9** Kinetics parameters for both MB and Cr (VI) adsorption

	1st-order kinetics				2nd-order kinetics			
	$R^2$	$K$	$Q_e$ calculated	$Q_e$ experimental	$R^2$	$K$	$Q_e$ calculated	$Q_e$ experimental
MB	0.4877	1.8325	305.9849	18.5	0.994	0.51347	18.48429	18.5
Cr (VI)	0.8166	-8.201	6.150352	10.9	0.983	0.20661	8.071025	10.9

effect on the MB removal efficiency, whereas the Cr (VI) concentration and the time were the significant factors that affected the Cr (VI) removal efficiency. These results confirmed the previous obtained results from the main effects plot, further see Tables 6 and 7.

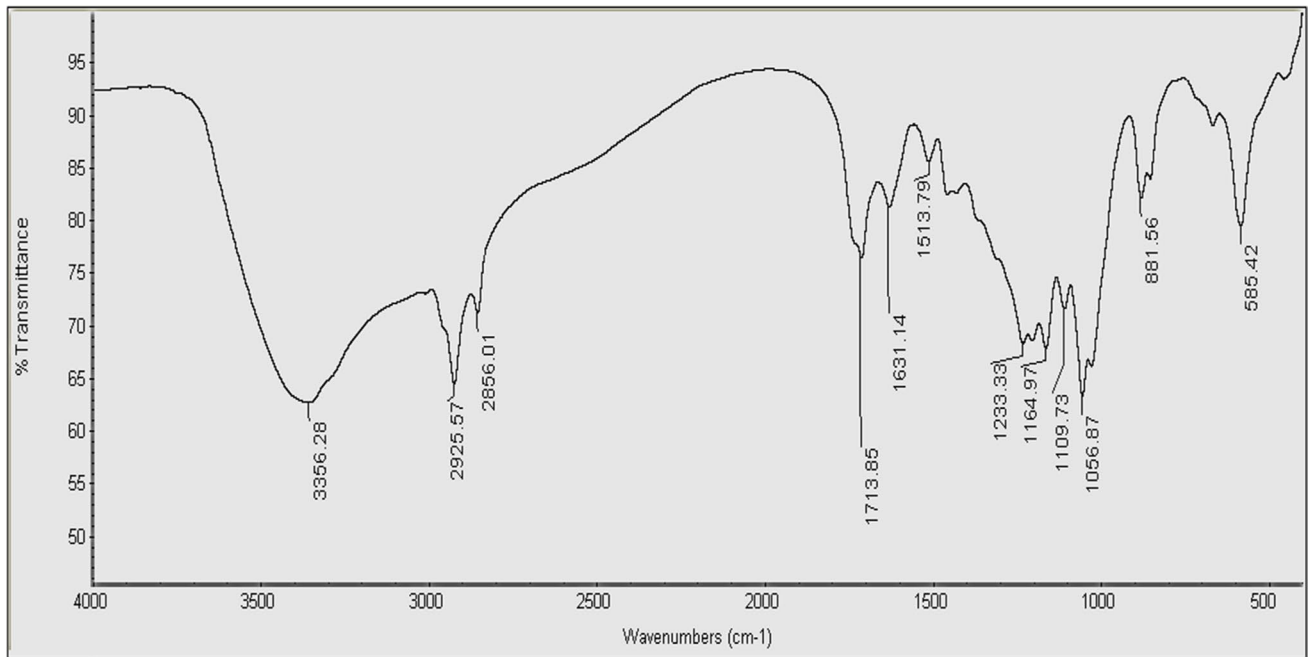
**Normal probability plot** Normal probability plot was displayed to achieve the real and chance effects which confirm its significance [47]. Figure 10a and b exhibited the normal plot for the MB and Cr (VI) removal efficiencies, respectively. From the plots, it was noticed that the MB concentration was the only significant real factor that affected the MB removal efficiency. Furthermore, the Cr (VI) concentration and the time were the real factors in the state of the Cr (VI) removal process.

### 3.2.4 Equilibrium studies

The two linear Langmuir and Freundlich adsorption isotherms were applied to display the adsorption mechanism. The linear plots of the Langmuir and Freundlich isotherms were afforded in Figs. 11 and 12 and the isotherm parameters were shown in Table 8. High noticeable  $R^2$  of Langmuir adsorption isotherm exhibited that Langmuir isotherm is a well-fitted model for this equilibrium. Furthermore, the results exhibited that the adsorption proceed via a monolayer adsorption [48–50]. Moreover, these results could be related to the adsorbent surface homogenous nature. It can be noticed that MB and Cr (VI) were adsorbed completely to the *Jatropha curcas* seedcake surface “active sites” until no further adsorption achieve [51].

### 3.2.5 Kinetics studies

Kinetics analysis studying is a fundamental step to assess the selected *Jatropha curcas* seedcake affinity towards MB and Cr (VI) removal. Figures 13 and 14 showed the kinetics studies of MB and Cr (VI) removal by applying *Jatropha*



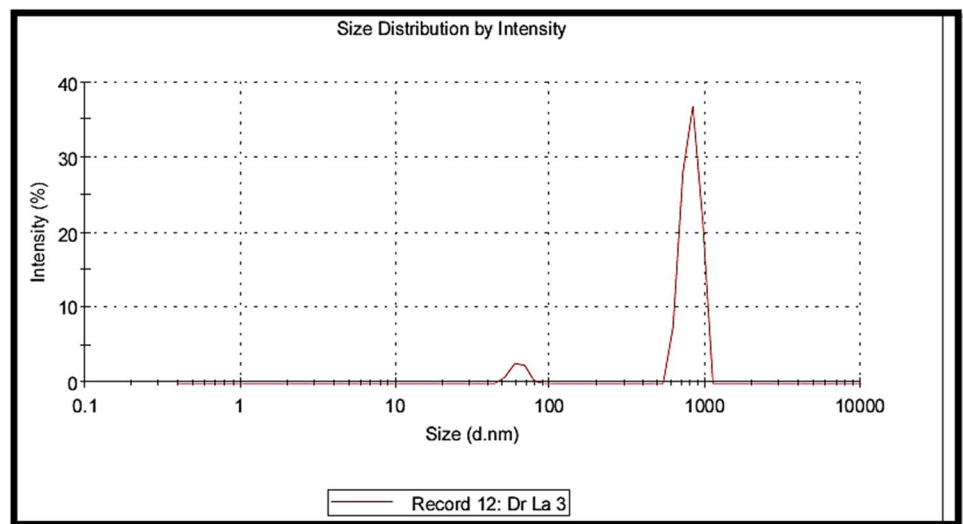
**Fig. 15** FT-IR spectra of the selected *Jatropha curcas* seedcake

*curcas* seedcake via pseudo-first-order and second-order linear plots. Additionally, Table 9 shows the various calculated parameters of both pseudo-first-order and second-order kinetics. The correlation co-efficient ( $R^2$  value) and the nearest results of both calculated and experimental  $Q_e$  indicated that the model of pseudo-second order fitted more to the experimental data than the pseudo-second order. Moreover, the pseudo-first-order model  $q_e$  calculated value fitted well with the  $q_e$  experimental value. In consequence, it can be concluded that first-order kinetics fits better with the achieved results.

### 3.2.6 Characterization of the selected *Jatropha curcas* seedcake

FT-IR spectra of the selected *Jatropha curcas* seedcake were studied to determine the distinguishing functional groups. That is why MB and Cr (VI) were adsorbed and removed (Fig. 15). The IR spectra of the selected *Jatropha curcas* seedcake displayed O–H stretching and aliphatic C–H (asymmetric and symmetric stretching) absorptions at 3356, 2925, and 2856  $\text{cm}^{-1}$ , respectively. Furthermore, the absorption at 1713  $\text{cm}^{-1}$  was due to C=O stretching. The absorption at 1631  $\text{cm}^{-1}$  represented asymmetric

**Fig. 16** DLS result of particle size of the selected *Jatropha curcas* seedcake



stretching band of unsaturated alkene: C=C bond. Furthermore, the C-O stretching of aliphatic ester was allocated at  $1233\text{ cm}^{-1}$ . The strong absorption at  $1164$  and  $1109\text{ cm}^{-1}$  originated from secondary and primary alcoholic C-O stretching, respectively. The absorbance band at  $881\text{ cm}^{-1}$  may be attributed to bending of C-H and/or C=C alkene groups. Finally, the absorption band at  $585\text{ cm}^{-1}$  may be attributed to stretching of halo compound as C-Cl or C-Br groups. Based on these results, the different reactive functional groups of the selected *Jatropha curcas* seedcake was responsible for the highest MBRE and CRE [52].

Dynamic light scattering (DLS) demonstrated the *Jatropha curcas* seedcake particle size distribution (see Fig. 16) which was stated with an average size of about 900 nm and trace of nanoparticles were a range of 60 nm.

## 4 Conclusion

This survey has successfully applied the *Jatropha curcas* seedcakes in bioethanol production and in the removal of cationic dye methylene blue (MB) and hexavalent chromium Cr (VI) from wastewater relying on the seedcake's possession of remarkable amounts of lignocellulosic biomass and highly adsorption functional groups. The lignocellulosic biomass was improved by irrigating the plants with sewage water, sewage water and sludge, and sewage water and sludge sprayed with tap water, 100, 200, 300, and 400 ppm of CCC. The seedcakes were acid hydrolyzed and the resultant hydrolysates were used in fermentation process by two yeast strains (*Candida tropicalis* Y-26 and *Saccharomyces cerevisiae* Y-39). Based on the achieved results, the irrigation by sewage water either alone or with addition of sludge achieved higher TRS content and bioethanol production than control plants irrigated by tap water. Belonging to CCC spraying, it was found that there was a direct relationship, i.e., higher concentrations (300 & 400 ppm) produced the highest TRS and bioethanol quantity. A preliminary screening experiment was done to select the seedcake achieving the best MB and Cr (VI) removal efficiencies, MBRE and CRE, respectively. The seedcakes irrigated by sewage water were the most efficient in MBRE and CRE with 78.8% and 41%, respectively. "One Factor at A time (OFAT) experiments and a  $2^3$  general full-factorial design" were done for the selected seedcakes. The maximum MBRE and CRE were achieved with the optimum parameters: 15 and 2.5 g/l of bio-sorbent dose (BM), conc. 50 ppm, and contact time 60 and 30 min for MBRE and CRE, respectively. The kinetic results proposed that the adsorption of MB and Cr (VI) fits well with the pseudo-first-order model. Moreover, the adsorption nature was found to be fitted more towards Langmuir which stated a monolayer adsorption nature of the bio-sorbent surface.

Further studies could be performed on the optimization experiments to the seedcakes to detect the optimal factors (concentration, temperature, time) for maximal bioethanol production. Additionally, using different ways of hydrolysis (enzymatic and enzymatic/chemical) to obtain the maximum quantity of TRS and in follow of bioethanol production will be evaluated.

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**Author contribution** All authors provided critical feedback and helped shape the research, analysis, and manuscript equally.

All authors discussed the results and commented on the manuscript equally.

Zahraa S. Taha carried out the experimental work, wrote the manuscript, performed, the analytic calculations.

Ahmed Labena conceived of the presented idea, carried out the experiment, performed the analytic calculations, aided in interpreting the results, and worked on the manuscript.

Hekmat R. Madian conceived of the presented idea, carried out the experiment, performed the analytic calculations, wrote the manuscript, aided in interpreting the results, and worked on the manuscript.

Hala S. Ahmed conceived of the presented idea, helped supervise the work, discussed the results, and commented on the manuscript,

H.M Hassan conceived of the presented idea, carried out the experiment, aided in interpreting the results, and worked on the manuscript.

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## Declarations

**Competing interests** The authors declare no competing interests.

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