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CACTUS JUICE PREPARATIONS AS BIOFLOCCULANT: PROPERTIES, CHARACTERISTICS AND APPLICATION

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Abstract

In this study, the flocculating properties of different cactus juice preparations were assessed using synthetic and real industrial and leachate wastewaters. It was found that the crude CJ had a flocculating activity (FA) of 89 ± 2.2% which is significantly higher than that of a powdery sample derived from a 60°C oven-dried CJ sample and which was about 37.8 ± 2.5%. Interestingly, both crude and dried samples showed the ability to flocculate kaolin suspensions within a wide pH range from 3 to 11. The flocculating activity was found to increase with the presence of divalent and trivalent cations. The enhancement is more significant in the case of the 60°C oven-dried sample. Crude bioflocculant showed to be thermally stable. Both crude and dried CJ removed COD in petrochemical and leachate wastewaters at efficiencies of 72±2.5% (with crude CJ), 69±3.0% (with dried CJ), 88±2.5% (with crude CJ) and 82±2.7% (with dried CJ), respectively. Flocculation reduced also the SS by 85±3.0% (with crude CJ), 75±1.5% (with dried CJ), 91±2.0% (with crude CJ) and 85±1.7% (with dried CJ), respectively for petrochemical and leachate wastewaters. The removal efficiency of an emerging pollutant (naproxen) from the kaolin suspension, did not exceed 32%. Physico-chemical analysis revealed that the bioflocculant is mainly composed of polysaccharide. Nevertheless, the elemental composition and the microstructure of the samples, obtained by SEM, may explain the variability of the flocculating activity of the CJ preparation.

Keywords: bioflocculant, cactus juice, emerging pollutant, flocculating activity, naproxen, opuntia

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1. Introduction

1.1. Background of actor-network theory (ANT)

Currently, various methods are used to treat industrial and municipal wastewaters and leachates from municipal solid wastes (MSW). The choice of the process depends on the wastewater characteristics, on the treatment objectives and on the economics of the process. The use of chemical treatment processes (including the chemical precipitation, coagulation-flocculation, chemical oxidation etc.) is one of the most common and well-known approaches. For efficiency and economic reasons, the conventional coagulation-flocculation process (using various chemicals such as salts of iron, aluminum, lime, and polyelectrolytes etc.) is world widely used, showing higher treatment efficiency in terms of organic matter, solids material and heavy metal removals (Abdel-Shafy and Emam, 1991; Ginos et al., 2006; Rodrigues et al., 2017; Szpak et al., 1996; Tatsi et al., 2003; Watanabe et al., 1993). This process is well used for industrial and municipal wastewaters treatment offering a higher resistance to toxic loadings and to higher quantities of organics, ease of operation, energy saving etc. (Lee et al., 2014). However, it was reported

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by many authors that residual synthetic coagulants and flocculants resulted in treated waters and sludges may induce health or ecological problems (Flaten, 2001; Ward et al., 2006).

Therefore, it is very important to point out the toxicity of synthetic coagulants and flocculants before using them to treat wastewaters (Aguilar, 2005). Safe and available alternative polymers should be considered when applying the coagulation-flocculation process. In this perspective, various natural materials of biological origin have been evaluated for their coagulation-flocculation properties. These potential polymers have to be accessible, economical, nontoxic and eco-friendly. They are derived from seeds, leaves, crustaceans, pieces of bark or sap as well as roots and fruits of plants (bean, Moringa, maize, chitosan etc.) (Abirami et al., 2010; Antov et al., 2010; Arjunan et al., 2012; Babineau et al., 2008; Diaz et al., 1999; Giuliano et al., 2017; Hassan et al., 2009; Katayon et al., 2006; Mangale et al., 2012; Ndabigengesere et al., 1995; Pritchard et al., 2009, 2010; Shyam and Kalwania, 2014; Zhang et al., 2006). However, the evaluation of the natural flocculants was generally limited to chemical oxygen demand (COD), nitrogen, phosphorus, suspended solids (SS) and turbidity removals and no studies were devoted to their ability to remove emerging pollutants present in wastewaters (Al-Hamadani et al., 2011; Khiari et al., 2010).

Emerging pollutants (Sauvé and Desrosiers, 2014), which represent an increasing environmental problem related to the human activities, include a wide range of chemicals such as pesticides, personal care and household products, pharmaceuticals, etc. For example, an acidic drug such as naproxen is a pharmaceutical product detected in municipal wastewaters at a concentration up to 4.6 μg/L (Daughton and Ternes, 1999). It was demonstrated that chronic toxicity, endocrine disruption and pathogen resistance are related to low naproxen concentration.

Therefore, the multi-generational exposure of naproxen can cause harmful effects for the aquatic system (Halling-Sörensen et al., 1998). Moreover, the presence of such a pollutant among others limit the reutilization of treated wastewater, which is not in agreement with a total sustainable water cycle management strategy (Muñoz et al., 2009).Recently, our research team has demonstrated that CJ can be used as an eco-friendly flocculant replacing polyacrylamide, for industrial wastewater treatment (Sellami et al., 2014).

This promising process will help to eliminate the troubles related to the use of synthetic polymers and ensure higher quality for treated water. However, more investigations are needed. Hence, the objectives of this study were to optimize the coagulation-flocculation parameters taking into consideration the chemical characteristics, as well as evaluating the stability and the flocculating properties of different forms of cactus juice. The efficiency of this natural material will be confirmed for petrochemical and MSW leachate wastewaters and the ability of CJ to remove naproxen will be also determined.

2. Experimental

2.1. Cactuses sampling and preparation

Cactuses (*Opuntia ficus indica*), were sampled in Sfax locality (Tunisia). Pant identification was carried out in the LBGEL laboratory (ENIS, Sfax, Tunisia). Samples were ground with a grinder, and filtered using a gauze compress. This operation produced a green liquid called cactus juice (CJ), which was stored at 4ºC until used. A sample of the crude CJ was oven dried at 60ºC to obtain a powder form. Another sample was allowed to settle for 24 hours, and then the supernatant and pellet were separated and lyophilized. The obtained powder was stored at ambient temperature until use.

2.2. Measurement of the flocculating activity

To determine the flocculating activity (FA), the coagulation-flocculation experiments were conducted using the jar-test procedure (Phipps & Bird model PB-700, equipped with six beakers of 1 L volume) in the presence of a kaolin clay suspension at 4 g/L, according to the method of Guo et al. (2014) with modification. Bioflocculant samples were added to the beakers at the flocculation stage. The jar-test procedure consisted of rapid stirring for a period of 3 min at 150 rpm followed by 17 min slow stirring at 60 rpm. After a settling period of 30 min, supernatant was taken from the upper phase. A control was prepared by substituting the bioflocculant with distilled water and the experiment was conducted under similar conditions. F_A (as %) was then calculated using the following formula (Guo et al., 2014) (Eq. 1):

$$F_A = \left(\frac{A-B}{A}\right)\times 100 \tag{1}$$

where *A* is the optical density at 550 nm of the control and *B* the optical density at 550 nm of the sample from the experiments using the bioflocculant.

2.3. Determination of the optimum bioflocculant doses

The same jar-test procedure as described above was used to determine the optimum dose of the bioflocculant for the clarification of kaolin clay suspension (at 4 g/L) at around neutral pH (Guo et al., 2014). Experiments were conducted at room temperature (22±1°C). Different concentrations of the bioflocculant were used. After settling, the upper phase was sampled to measure the optical density and determine the F_A as described above.

2.4. Thermal stability test

Crude CJ samples were incubated in water baths fixed at 50 and 100ºC (for 30 min). Then, the residual FA was determined using the same jar-test procedure as described above.

2.5. Effects of cations on cactus juice flocculating activity

Various solutions (1%) of monovalent (KCl, NaCl), divalent ($MnCl_2$, $MgCl_2$, $CaCl_2$, $FeSO_4$) and trivalent ($FeCl_3$) cations were added separately at a rate of 10 mL/L to the kaolin raw solution samples. A control without cation addition was also conducted. Then, the FA was determined using the same jar-test procedure as described above.

2.6. Effect of pH on cactus juice flocculating activity

The effect of pH on FA of the crude, 60°C dried and lyophilized samples of CJ was examined. The pH of the kaolin solution was varied in the range of 3 to 12 using either 1.0 N HCl or NaOH solutions.

2.7. Effect of the cactus juice preparation on the flocculating activity

The FA of crude, dried and lyophilized samples of CJ was determined as described above.

2.8. Chemical analysis of cactus juice

CJ samples were analyzed in accordance with AOAC (1995) methods and as described by Sellami et al. (2014).

2.9. Scanning electron microscopy observations

The morphology and the elemental composition of CJ samples were determined by a scanning electron microscope (FEG-SEM S-4700, HITACHI) equipped with energy dispersion spectrometer (EDS).

2.10. Treatment assays using cactus juice as bioflocculant

Coagulation–flocculation experiments using the jar-test procedure were conducted for petrochemical wastewater, landfill leachate and also a kaolin suspension (4 g/L) supplemented with naproxen (5 µg/L). Petrochemical wastewater and leachate samples were collected respectively from a company treating petroleum wastewater and a controlled discharge solid waste disposal near the Sfax region (Tunisia).

Dried and crude CJ samples used as bioflocculants (at optimum doses) were added to the beakers at the flocculation stage. After a settling period of 30 min, supernatant was taken from the upper phase and treatment efficiencies were evaluated by calculating suspended solids (SS) and chemical oxygen demand (COD) removals. Also, naproxen removal was determined from the kaolin solution. SS and COD were determined according to Standard Methods (APHA, 1992). pH was measured with a pH-meter (model 420A, Orion, USA).

Naproxen-containing samples were filtered through a 0.2 µm filter prior to analysis. Samples were analyzed using an Acquity – Xevo TQ MS (Waters Corp.) UPLC/MS-MS system. The LC parameters were mobile phase A H_2O + 0.2% formic acid, mobile phase B methanol:acetonitrile (80:20 v/v)+ 0,2% formic acid, column temperature 40°C and 5 µL injection volume. The MS parameters were ESI+ as the ionization mode of the source, capillary and cone voltages 2.5 kV and 20 V respectively and MRM (multiple reaction monitoring) as the detection method for quantification. Parent / daughter m/z were 231.1 / 115 with collision energy (CE) of 40 eV, and 231.1 / 185.1 with a CE of 10 eV.

2.11. Data analysis

All experiments were conducted in three to four replicates and statistical analyses were performed using ANOVA analysis. Differences were considered significant at $P<0.05$.

3. Results and discussion

3.1. Determination of the optimum doses

The jar-test experiments were performed using CJ (crude and dried sample at 60ºC) as bioflocculant with doses varying between 0.056 and 0.729 g/L (g/dry weight of CJ) and between 0.1 and 0.6 g/L (g/dry weight of CJ) respectively for crude and dried samples (Fig. 1). In the presence of the crude sample, the FA increased with an increase of both CJ dosages until reaching an optimal value of 0.28 g/L giving a FA of 89±2.2%. Above this optimum, the flocculating efficiency remains almost constant.

In case of the dried CJ, the optimum dose was 0.4 g/L giving a FA of almost 37.8±2.5%. This value decreased while increasing the dried CJ concentration above the optimum dose. The decline may be the result of kaolin particles re-suspension beyond this optimum concentration. The high concentrations of the bioflocculant might confer positive charges on the particle surface (a positive zeta potential) allowing re-dispersion of the particles as reported for polyelectrolytes (Kemmer, 1988). The crude sample shows a high FA compared to the dried CJ. This could be explained by the importance of hydrogen bonds in enhancing the flocculation process. The drying technique may break the hydrogen bonds and consequently reduce the FA.

Generally, dosage is an important parameter to be considered during the determination of optimum conditions for the performance of coagulation-flocculation process and an inadequate dosage or an over-dosage may reduce the flocculating performance (Hassan et al., 2009). Therefore, the determination of the optimum flocculant dose may achieve better performance of the treatment process and simultaneously reduce the process cost.

3.2. Thermal stability test

Thermal stability of the crude CJ was examined at 50 and 100ºC for 30 minute period and compared to the untreated sample (Fig. 2). FA values were 89.7±2.8% and 88.3±1.7% respectively for 50 and 100ºC treatment. These values were not statistically different (*p*-value > 0.05) from that obtained by the untreated CJ sample (89±2.2%). Therefore, the CJ was considered to be thermally stable. The high FA and thermal stability may be attributed to the nature of the bioflocculant which is composed mainly of polysaccharide. It is worth noting that similar remarks were reported for bioflocculants produced by microorganisms (Cosa and Sekelwa, 2014).

In this perspective, it has been reported that bioflocculants with protein or peptide backbone in the structure were generally sensitive to heat, while those made of sugars were thermostable (Salehizadeh and Shojaosadati, 2001). The biochemical composition of CJ (Table 1) confirmed the presence of polysaccharide in CJ as a main ingredient however proteins are lower and were under 10% w/dw as reported by Sellami et al. (2014).

The high flocculation capacity of CJ may be attributed to the complex of carbohydrate contained in CJ as l-arainose, d-galactose, l-rhamnose, d-xylose and galacturonic acid which have a great water retention capacity (Swati and Govindan, 2005; Vijayaraghavan et al., 2011).

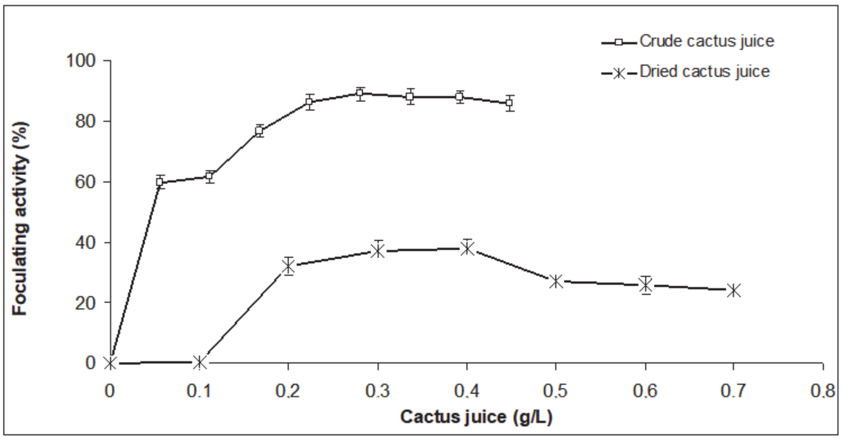


Fig. 1. Determination of the optimum doses for liquid and dried CJ at 60ºC

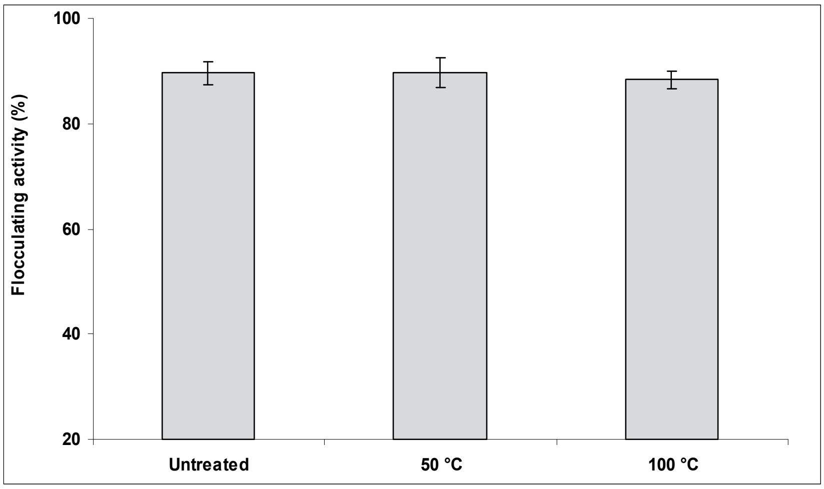


Fig. 2. Thermal effect on the FA of crude CJ

Table 1. Biochemical composition of CJ (% w/dw)

	Pellet From CJ	***Supernatant from CJ***	***CJ***
Proteins	9.45 ± 0.07	5.69 ± 0.37	8.35 ± 0.15
Lipids	4.35 ± 0.59	1.53 ± 0.17	2.01 ± 0.26
Ash	2.58 ± 0.18	2.76 ± 0.16	2.58 ± 0.17
Carbohydrate	83.63 ± 0.69	90.02 ± 0.38	87.06 ± 0.06

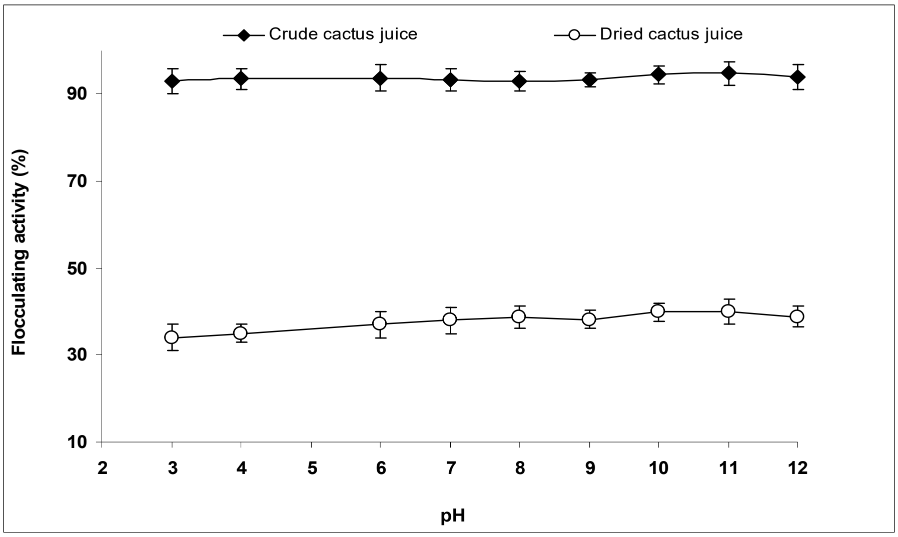


Fig. 3. Effect of the pH on the FA of crude and dried CJ

3.3. Effect of pH

The effect of pH on the FA of CJ within a range of 3.0 to 12.0 indicated that both crude and dried samples showed the ability to flocculate kaolin suspension within a wide range of pH (Fig. 3), while CJ showed an acidic pH value of 4.30. A slight enhancement of the FA was observed at pH 10 and 11. Generally, pH plays a decisive role in the flocculating process. Wang et al. (2011) stated that pH affects the stability of suspended particles and the formation of flocs. For example, Guo et al. (2013) showed the importance of a higher pH for flocculating process during the use of a bioflocculant from activated sludge. This may be explained by the biofloculant nature (a protein is the main backbone of this microbial bioflocculant obtained from activated sludge) (Guo et al., 2013). However, an exopolysaccharide produced by *Bacillus* spp. UPMB13 was demonstrated to have a relatively wide pH tolerance ranging from slightly acidic to slightly alkaline conditions (Zulkeflee et al., 2012).

3.4. Effect of cations

Effects of salts on the FA of CJ are shown in Fig. 4. For both crude and dried CJ, the FA was enhanced in the presence of divalent cations (Mg^{2+}, Mn^{2+}, Ca^{2+} and Fe^{2+}) and the trivalent ion (Fe^{3+}). Generally, these cations neutralize and stabilize the negative charges of the functional groups of kaolin particles and CJ.

As reported by many researchers, cations are usually used in the flocculation process in order to enhance the flocculating efficiency and increase the absorption of the flocculant onto suspended particles, allowing the decrease of the negative charges of the particles and of the bioflocculant (Deng et al., 2003; Zhang et al., 2012). Various cations show significant effects on different bioflocculants. According to Zhang et al. (2012), various divalent and trivalent cations such as Ca^{2+}, Al^{3+} and Mg^{2+} enhanced FA, but not K^+ and Na^+ (Wang et al., 2011). As reported by Sellami et al. (2014), the IR spectrum of the polymer proved the presence of the carboxyl group which may serve as binding sites for cations.

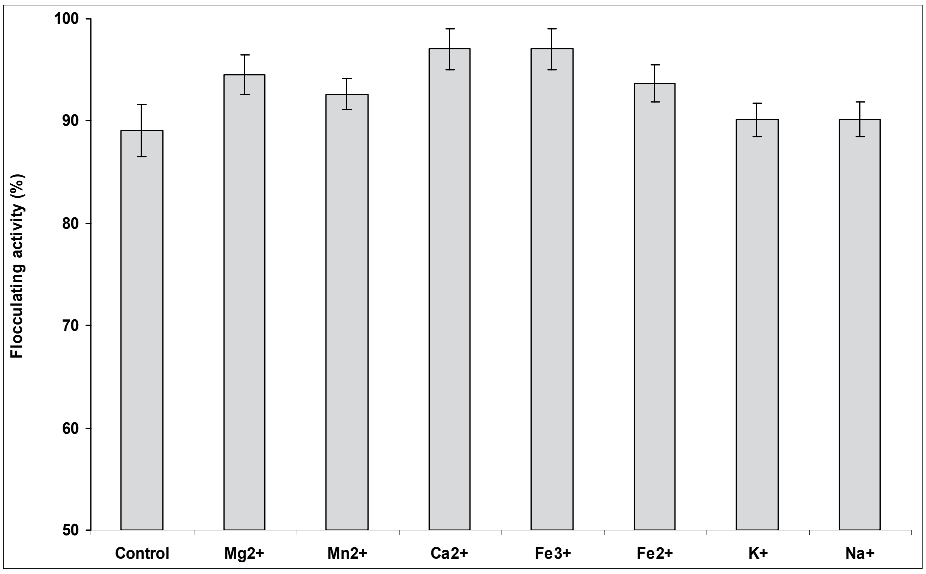
3.5. Effect of the CJ preparation on the flocculating activity

Fig. 5 shows the FA of crude, dried and lyophilized samples of CJ. The crude CJ and the lyophilized CJ supernatant gave the highest FA (89.6 ± 2.2% and 86.7 ± 2.5%, respectively). However, the lyophilized JC pellets showed a flocculating activity of 73.4 ± 2.6%. Compared to lyophilizing process, drying at 60°C reduced significantly the FA of the CJ (37.8 ± 2.5% and 80. 9 ± 3% respectively for the dried and lyophilized crude CJ).

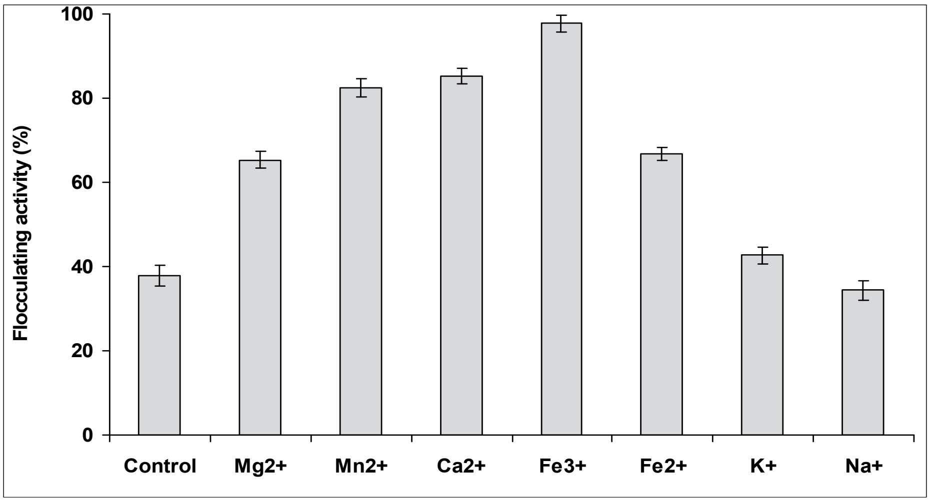
The fact that the crude CJ allows the highest FA may be explained by the importance of hydrogen bonds which are considered as belonging to the preferred groups for the flocculation process, and the dewatering process by either lyophilizing or drying which may break the hydrogen bonds and consequently reduce the FA. Moreover the prolonged drying process may have altered the CJ flocculating activities.

3.6. Applications of cactus juice for pollutant removals

The efficiency of crude and dried CJ to remove pollutants was evaluated for kaolin suspension supplemented with naproxen and for two real wastewater samples (petrochemical wastewater and leachate from a controlled discharge solid waste disposal). Treatment results are presented in Table 2. For kaolin suspension, naproxen removal rates were 22 ± 4.6% and 32 ± 6.8% obtained with crude and dried CJ at optimum doses of 0.28 and 0.40 g/L, respectively.



(a)



(b)

Fig. 4. Effect of cations on FA (a) crude CJ; (b) dried CJ at 60ºC

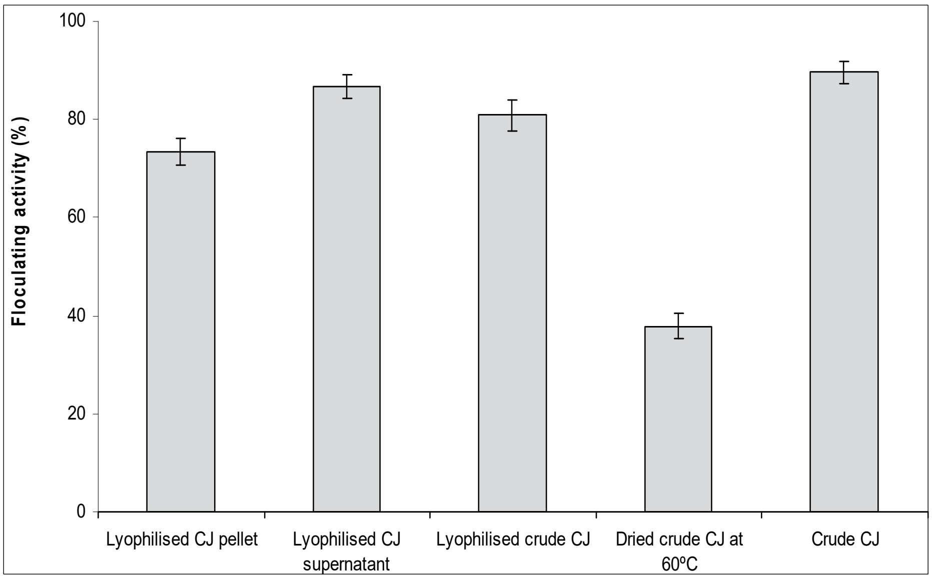


Fig. 5. Effect of the CJ preparation on the FA

Table 2. Treatment assays with CJ

	Kaolin solution containing naproxen[3]	***Leachate from controlled discharge [2]***	***Petrochemical wastewater[1]***
Treatment with crude CJ			
Optimum crude CJ dose (g/L)	0.280	0.081	0.081
COD removal (%)	-	88±2.5	72±2.5
SS removal (%)	-	91±2.0	85±3.0
Naproxen removal (%)	22 ± 4.6	-	-
Treatment with dried CJ			
Optimum dried CJ dose (g/L)	0.400	0.180	0.180
COD removal (%)	-	82±2.7	69±3.0
SS removal (%)	-	85±1.7	75±1.5
Naproxen removal (%)	32.8 ± 6.8	-	-

Note: [1]effluent with initial COD = 45 g/L, initial SS = 0.29 g/L and pH = 9.23; [2]effluent with initial COD =92 g/L, initial SS =0.37 g/L and pH = 9.97; [3]kaolin 4 g/L and naproxen 5 µg/L

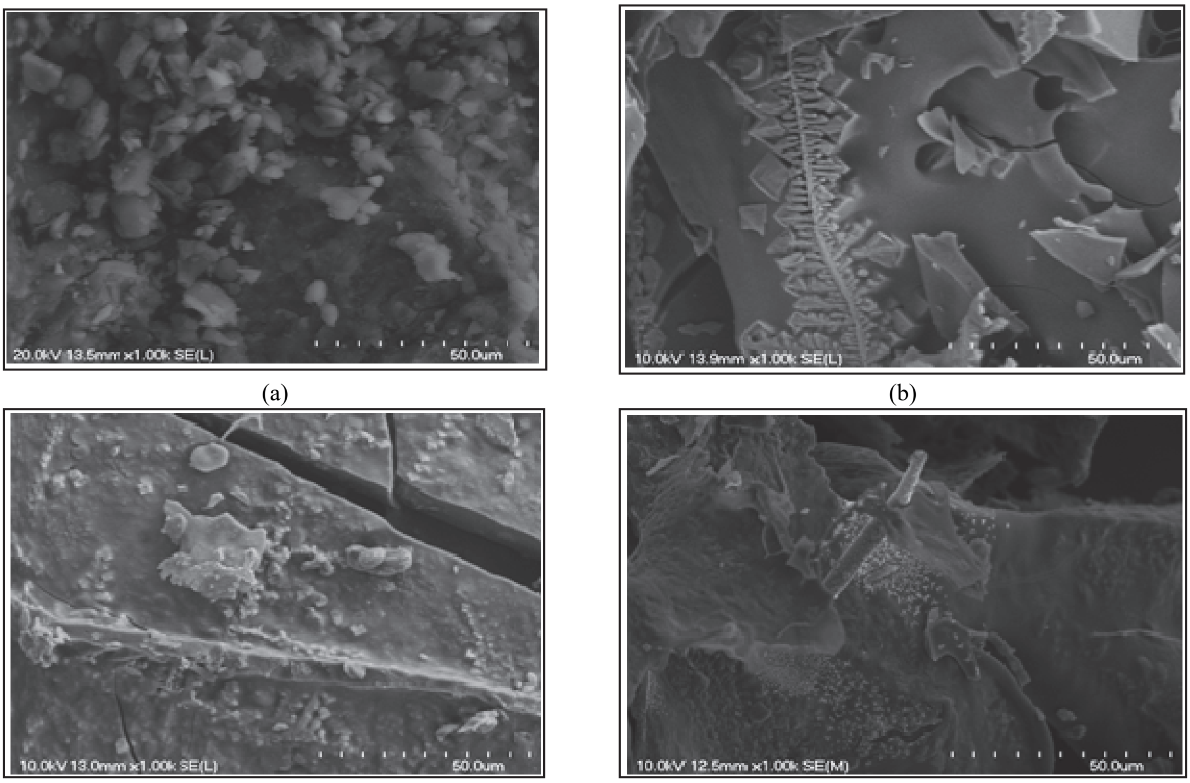


Fig. 6. SEM of CJ: (a) dried crude CJ; (b) lyophilised crude CJ; (c) lyophilized supernatant from crude CJ; (d) lyophilised pellet from crude CJ

For the petrochemical wastewater, removal efficiencies of SS and COD were 85 ± 3.0% and 72 ± 2.5%, respectively at the optimum dose of 0.081 g/L for the crude CJ. Using the dried CJ sample (optimum dose of 0.18 g/L), SS and COD removals were lower with 75 ± 1.5% and 69 ± 3.0% of removal rates, respectively. The application of CJ as bioflocculant for the leachate wastewater showed higher removal efficiency compared to the petrochemical wastewater (91 ± 2.0% for SS and 88 ± 2.5% for COD obtained with the crude CJ and 85 ± 1.7% for SS and 82 ± 2.7% for COD obtained with the dried CJ). The variability in the efficiencies of CJ for the two types of wastewater may be related to their original characteristics as reported for the synthetic polyelectrolytes used for the treatment of various wastewaters (Goloba et al., 2005; Zhu et al., 2011). Results variability was also observed for various biofloculants by other researchers. For examples, Al-Hamadani et al. (2011) used dried and grinded isabgol as bioflocculant for the treatment of leachate at an optimum dose of 0.4 g/L. In this case, SS and COD removals of 96 and 64% were observed. Suopajärvi et al. (2013) obtained a COD removal of 60% when using 5 mg/L of cellulose as bioflocculant in the treatment of municipal wastewater. Also 2.68 g/L of bioflocculant extracted from *Pseudomonas aeruginosa* strain was used in the treatment of petroleum wastewater and a COD removal of 25% was obtained (Pathak et al., 2014).

Table 3. Elemental composition of CJ (wt %)

Element	*Lyophilized crude CJ*	*Lyophilized supernatant from crude CJ*	*Lyophilized pellet from crude CJ*	*Dried crude CJ*
C	48.31 ± 0.10	45.37 ± 0.69	53.83 ± 1.63	49.24 ± 1.51
O	32.89 ± 0.32	34.19 ± 3.74	36.99 ± 1.35	35.24 ± 1.11
Mg	0.91 ± 0.02	1.32 ± 0.06	0.52 ± 0.17	0.83 ± 0.17
Al	0.12 ± 0.00	0.26 ± 0.07	0.55 ± 0.06	0.30 ± 0.03
P	0.13 ± 0.01	0.17 ± 0.01	0.09 ± 0.01	0.12 ± 0.04
S	0.37 ± 0.02	0.38 ± 0.06	0.22 ± 0.04	0.32 ± 0.15
Cl	7.97 ± 0.09	9.09 ± 2.35	3.43 ± 1.16	5.86 ± 0.76
K	6.39 ± 0.04	6.31 ± 1.79	2.67 ± 1.05	4.50 ± 0.72
Ca	2.75 ± 0.08	2.82 ± 0.48	1.48 ± 0.56	3.57 ± 1.28
Cu	0.13 ± 0.01	0.02 ± 0.01	0.1 ± 0.04	0.1 ± 0.00

3.7. Scanning electron microscopy observation of cactus juice

SEM observations of the cactus juice's surface were carried out to explore the effects of different preparation and drying at 60ºC on the microstructure changes of the samples (Fig. 6). Extensive surface shrinkage was found on CJ dried at 60ºC, which may have a significant effect on the microstructure of the sample. However, lyophilizing (low processing temperature and the absence of air) led to relatively uniform size and shape with smooth particle surface. This process prevents deterioration due to oxidation or chemical modification of the product. Generally, natural product preparations cause various changes in the microstructure of the samples and lead to products with different properties.

The EDX analysis of CJ in Table 3 shows its major elemental composition (% w/w). Table 3 reveals a slight variation in elemental content between CJ samples (crude, dried and lyophilized samples). The weight percentage of each element varied depending on the preparation process. The elemental compositions are indicative of the presence of polysaccharide the main compound of the CJ bioflocculant. Nevertheless, it is very important to indicate that the variation of the elemental composition and the microstructure of the samples may explain the variability of the flocculating activity of the CJ preparation.

4. Conclusions

To our knowledge this is the first study evaluating the flocculant properties of CJ under different formulations. The crude, dried and lyophilized CJ showed remarkable FA. The bioflocculant was thermally stable. High SS and COD removals for petrochemical and leachate wastewaters have been obtained.

The capability of CJ for naproxen removal, an emerging pollutant, was also observed for kaolin suspension. The flocculation stability at wide range of pH and the enhancement of the FA in the presence of divalent and trivalent cations make CJ an attractive candidate for the treatment of different wastewaters.

Abbreviations

ANOVA: analysis of variance;
CE: collision energy;
CJ: cactus juice;
COD: chemical oxygen demand;
EDS: energy dispersion spectrometer;
F_A: flocculation activity;
IR: infrared;
LC: liquid chromatography;
MS: mass spectrometry;
MSW: municipal solid waste;
SS: suspended solids.

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