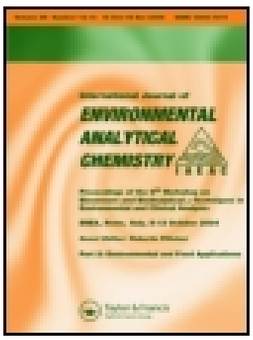

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ARTICLE

Effect of a natural coagulant extract from *Opuntia ficus-indica* cladode on electrocoagulation-electroflotation water treatment process

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ABSTRACT

Opuntia ficus-indica: (OFI) cladode mucilage was extracted using conventional extraction (CE) and ultrasound-assisted extraction (UAE), to test its effect on the efficiency of electrocoagulation-electroflotation (EC-EF) water treatment process. The mucilage was identified by biochemical assays, by scanning electron microscope analysis (SEM), and by Attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR). The results showed that the UAE enhanced mucilage extraction yield. At 10 min of extraction time, the extraction yield attained 60.77 ± 1.07% at 60% of the amplitude sonication, while CE allows only 36.71 ± 1.07% after 60 min at 80 ^{0}C. EC-EF mucilage-assisted process enhanced turbidity removal efficiency by 30% at 15 min of treatment time. Mucilage identification showed a bio-material, of a polysaccharide nature, with several functional groups through SEM, biochemical essays and ATR-FTIR, respectively. The addition of 5 mg/L OFI mucilage to EC-EF allowed 89.47% of EC-EF efficiency at 25 min.

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Opuntia ficus-indica; cladode mucilage; biocoagulant; ultrasound-assisted extraction; electrocoagulation-electroflotation

1. Introduction

Opuntia ficus-indica (OFI) belongs to the family of *Cactaceae*, commonly known under the name of prickly pear. It is a native plant of Mexico that grows currently in different countries of the world [1]. It is a suitable plant for sustainable agriculture especially in sub-arid regions, due to its resistance to drought, its effect against desertification, and its use in feeding man and cattle. The plantation of Algerian cactus covers about 28 000 ha in 2015, in recent years studies have shown that 1 ha is able to produce up to 100 t of fresh cladodes per year with low rainfall [2]. In order to meet the growing global demand for pectin, recent studies have explored various plant products and by-products including

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cladodes of *Opuntia ficus-indica* for pectin extraction [3]. It is known that food technologists are always looking for alternative sources of soluble fibres (gums, mucilage, pectin, hemicelluloses or polysaccharides) with new-improved functional properties [4]. Polysaccharides were used empirically to adjust and improve the rheological properties of some products [5]. OFI mucilage as a pectic polysaccharide is considered as a potential source for industrial hydro-colloids, multiple uses have been found for this component as a food thickener, food emulsifier, natural super-plasticiser in the mortar and as a food product [2,6]. It is necessary to develop an appropriate, selective and profitable system for environmental-friendly extraction technologies [7,8]. The use of ultrasound to extract compounds could be a promising technique. Its application reduces the use of organic solvents and allows the extraction of different compounds, such as proteins, pigments, phenols, flavours, sugars and oils [9–14]. Many studies conducted on the extraction of high-quality bioactive polysaccharides from plant materials used the ultrasound technology. These studies showed that this technique is an effective extraction method with many advantages such as extraction yield enhancement, reduction in solvents use, and reduction in extraction time [15]. Ultrasound-assisted extraction of polysaccharides reveals not only an enhancement of the extraction of the compounds, but also preserves the structure and molecular properties [16].

On the other hand, industrial growth and changes in manufacturing processes have led to the problematic of wastewater discharges into the environment. Many traditional and/or new treatment processes have been modified and developed to eliminate the releases of various chemical substances (copper, lead …) using low-cost natural coagulants [17–20]. Electrocoagulation-electroflotation (EC-EF) is a known technology for its great potential to purify wastewater, it has the ability to eliminate a broad spectrum of pollutants and presents itself as a new alternative [21]. Inorganic coagulants/flocculants most commonly used in EC-EF technology are aluminium and iron [22]. Their uses are accompanied by a significant consumption of chemicals including the addition of chemical adjuvants leading to the production of large volumes of toxic sludge. Nowadays, there is a growing interest in the use, environmentally friendly biomaterials of low cost to remove synthetic pollutants [20,23]. Several researchers reported the potential of *Opuntia ficus-indica* mucilage to replace iron or aluminium coagulants in the coagulation-flocculation standard jar-tests [23–25]. Moreover, a synergetic effect of cactus cladode powder with aluminium chloride gave better results than cactus or aluminium chloride used exclusively [25]. Currently, from the industrial point of view, Al and Fe are highly accessible around the world. However, the use of *Opuntia ficus-indica* as a natural coagulant may strengthen the treatment without notable additional cost. OFI cladode mucilage characterisation showed that mucilage is a water-soluble pectin-like polysaccharide of high molecular weight composed of several polysaccharides: neutral sugars, as L-arabinose, D-galactose, L-rhamnose, D-xylose, and polygalacturonic acid with varying proportions [2,4,26]. These polysaccharides act as adsorbents in water treatment [27]. Mucilage is a slimy fluid when the cladode is cut, it has the ability to precipitate ions and particles from aqueous solutions through surface-active mucilage properties [4]. The efficacy of OFI cladode juice in improving the elimination performance of turbidity by EC-EF process was reported, allowing 15% of the improvement after RSM optimisation [28]. Also, the association of OFI mucilage with EC-EF allowed 100% copper removal under certain conditions of pH, conductivity, agitation rate and current density [29]. Higher EC-EF turbidity

removal (93.14 ± 1.31%) was obtained under optimal conditions of voltage, pH conductivity and mucilage concentration (2.5 mg/L) by contrast to 62.02 ± 1.45% in EC-EF without mucilage [30].

Nevertheless, to the best of our knowledge, combination of an eco-process for natural coagulant extraction; ultrasound-assisted extraction of *Opuntia ficus-indica* (OFI) mucilage as a bio-coagulant has not been previously reported, as well as its UAE extraction from OFI. This is also the first study on OFI cladode mucilage structure by SEM and FTIR-ATR analysis. This study aims to establish a comparison of mucilage extraction performance using conventional extraction (CE) and ultrasound-assisted extraction (UAE), the valorization of *Opuntia ficus-indica* cladode as an agricultural by-product in wastewater treatment through examining its effect on EC-EF process efficiency, and an identification approach that confirm the adsorbent and polysaccharide nature of this biocoagulant.

2. Experimental

2.1. Sample collection and plant preparation

Thornless cladodes of *Opuntia ficus-indica* were harvested at Bir Essalem (Bejaia, northeast of Algeria) during the month of March 2018. The species belongs to the genus *Opuntia*, its identification is based on the morphology and physical characteristics of cladodes (dimensions, number of spines and areoles), flowers, and fruits colour. The species *Opuntia ficus-indica* is a plant without spines [31]. The cladodes of OFI were kept cold at 4°C and one of these cladodes was used after a day of harvest. The cladode was cleaned by removing thorns and black spots, followed by rinsing with tap water and distilled water. Measures of dimensions of the cladode were noted (length 430 ± 3 mm, width 196 ± 25 mm and thickness 23 ± 5 mm), a number of areoles and weight of the cladode are 155 ± 2.5 and 1133.83 ± 42 g, respectively. The cladode was cut into strips, and the outer epidermis (cuticle) was gently removed [27], using a kitchen knife. Then, cladodes were mixed with three volumes of water during 50 s using a domestic mixer [6,32,33]. The obtained ground material was kept in 500 mL beaker and stored at 4°C for mucilage extraction.

2.2. Conventional extraction (CE)

Conventional extraction was carried out by maceration of the ground material in distilled water and agitation on stirring plate (Velps Scientifica, France) at varied extraction times (60, 120, and 180 min). This operation was carried out at two different temperatures: room temperature (20–22°C) and 80°C. The mucilage was precipitated using ethanol (96%) (v/v).

2.3. Ultrasound-assisted extraction (UAE)

Mucilage extraction was carried by subjecting the cladode mixture to ultrasound waves by a sonicator probe (Bioblock Scientific-Vebra Cell75115). Two parameters, time and amplitude, have been studied in order to improve extraction yields. The sonicator was set 60% of the amplitude for extraction performance at different times (1, 3, 5, 10, 20, 30, 40

and 50 min). Then, amplitude value was varied (20 and 100%) at fixed extraction time. The probe pulsation value was equal to 5 s at each test.

After both extractions (CE and UAE), the mixture was filtered using a domestic strainer to remove the pulp; then, the recovered filtrate is subjected to mucilage precipitation by adding ethanol 96% (v/v) at a ratio of 1:3; then, the mixture was stored at 4°C overnight [34].

Mucilage filtrate was recovered by centrifugation at 5000 rpm for 5 min at 4°C (Sigma 2–16 PK Model, Osterode AM Harzs, Germany) followed by drying in a ventilated oven (Nuve-FN 400P, Turkey) for 1 h at 40°C in order to remove ethanol and lyophilisation using a Lyophiliser (ALPHA 1–2 LD plus, Germany). The dry extract was weighed for yield calculation. The extraction procedures of mucilage from OFI cladode are shown in Figure 1. Before mucilage addition to the EC-EF treatment process, the extract was dissolved in a solution of NaOH (1 M) at room temperature [29].

2.4. Determination of total dry matter (moisture determination)

Total dry matter of OFI cladode was carried out using the oven-drying method. 100 g of cladode were placed in an oven at a temperature of 103 ± 2°C until complete dryness. After that, the results were expressed in percent (%, w/w) as follows (equation (1)):

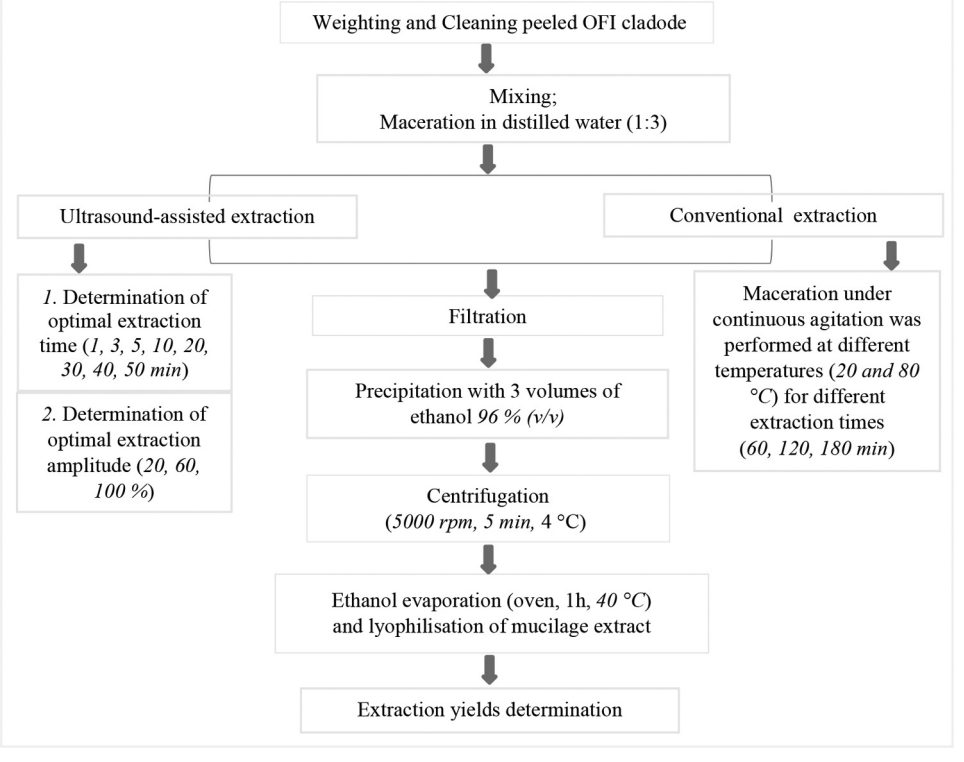

Figure 1. Extractions procedures for mucilage isolation from *Opuntia ficus-indica* (OFI) cladodes.

$$M\ (\%) = \frac{W_i - W_d}{W_i}.100 \quad (1)$$

where M is moisture, W_i is the initial weight of the sample, W_d is the weight of the sample after drying.

2.5. Calculation of extraction yield

The extraction yield (EY(%)) was calculated as the percentage of the weight (g) of the dried lyophilised mucilage extract obtained from 100 g of raw material, on the dry matter determined in the initial fresh material [16], as explained below (equation (2)):

$$EY(\%) = \frac{weight\ of\ the\ lyophilized\ extract}{weight\ of\ the\ dehydrated\ initial\ sample}.100 \quad (2)$$

Therefore, the weight of the dehydrated original sample refers to the dry matter determined in the original fresh sample (100 g used for extraction).

2.6. Determination of pHpzc (pH point of zero charge)

The pH at which the charge of the solid surface is zero is referred to as the point of zero charge (pHpzc). The pHpzc of a biomaterial helps to understand the coagulation flocculation mechanism. The determination of the mucilage pHpzc was carried as follows [35]: 50 mL of 0.01 M NaCl solutions were placed in closed Erlenmeyer flasks. The pH of each solution of each flask was adjusted to values of 2.25, 4.19, 5.94, 8.27, 9.8 and 10.84 by adding HCl or NaOH solutions (0.1 M). These values correspond to Initial pH and were considered to be approximately equal to pH 2, 4, 6, 8, 10 and 11, respectively. Then, 0.15 g of mucilage powder sample was added to NaCl solutions. The Erlenmeyer flasks were gently stirred at room temperature and the final pH values were measured after 48 h. The pHpzc is the point where the curve final pH versus initial pH crosses the line $pH_{initial} = pH_{final}$ [35,36].

2.7. Colourimetric biochemical assays

Lyophilised mucilage extract was dissolved in distilled water and gently stirred for 24 h to obtain a final concentration of 1 g/L. The pH of the mucilage was measured from this 1 g/L overnight mucilage solution. Specific volumes of the solution were taken depending on the biochemical dosage [37,38], as explained below.

2.7.1. Determination of neutral sugars

The content of neutral sugars was determined using phenol sulphuric acid method of [39]. 200 µL of aqueous phenol solution (5%) are added to a previously prepared mucilage solution (200 µL). The mixture is homogenised using a vortex, then 1 mL of concentrated sulphuric acid (97%) is rapidly introduced in the reactional mixture. The solution is left in a water bath at 100°C for 5 min. Tubes are cooled and placed in the dark for 30 min. A yellow colouration appears. The absorbance of optical densities is performed at 480 nm (λ_{max}). Sugar dosages were established using a glucose standard solution (50, 80 and 100 µg/mL).

2.7.2. Determination of uronic acids

Uronic acids were determined using the method described by [40]. Sodium tetraborate solution (0,125 M) is prepared in concentrated sulphuric acid, then 1.2 mL of this solution is added to 200 µL of mucilage solution. The mixing is homogenised by a vortex, and placed in ice (≃ – 5°C). Then, tubes are placed in a water bath of 100°C for 5 min. After cooling in an ice bath, 20 µL of a solution of mHDP (meta-hydroxydiphenyl) is added, the mHDP solution of 0.15% was prepared in a sodium hydroxide solution of NaOH at 0.5%. After agitation, pink colouration raises progressively. Absorbance is read at 520 nm (λ_{max}). A standard calibration range is prepared with galacturonic acid at concentrations of 25, 50, 100, 200 and 400 µg/mL.

2.7.3. Determination of reducing sugars

The quantification of reducing sugars was carried out using PAHBAH (p-hydroxybenzoic acid hydrazide) method described by [41]. 750 µL of a solution of PAHBAH (5%) are added to a mucilage solution of 250 µL. The PAHBAH solution of 5% is prepared in HCl (0.5 M) and freshly diluted by NaOH (0.5 M) to 1/5. The mixture is homogenised and placed in a water bath (100°C) during 5 min, and then it was cooled under tap water. The absorbance is read at 410 nm (λ_{max}). Xylose standard calibration curve is prepared at concentrations of 10, 20, 30, 40 and 50 µg/mL to establish the reducing sugar concentrations.

2.7.4. Determination of proteins

Proteins dosage was made according to the method of [42]. Three solutions were prepared. Solution A is composed of 4 g/L of NaOH, 20 g/L of sodium carbonate and 0.2 g/L of potassium sodium tartrate. Solution B is composed of copper sulphate at 5 g/L. Solution C, is prepared extemporaneously with 50 mL of solution A and 1 mL of solution B. The calibration range is a solution of bovine serum albumin (BSA) of 15 to 75 µg/mL. Then, 1 mL of the sample and 3 mL of solution C are mixed. The tubes were allowed to stand 15 min at room temperature and exposed to light. Folin-Ciocalteu reagent is added (0.3 mL), the tubes were shaken and placed in dark for 30 min. Absorbance is read at 750 nm (λ_{max}) against the blank. The blank, in all tests, is a solution of distilled water.

2.8. ATR-Fourier Transform Infrared (FTIR) spectroscopy

Attenuated Total Reflectance (ATR) is today the most widely used FTIR sampling tool. The lyophilised mucilage fraction was directly pressed onto a ZnSe crystal support. The IR beam reflects from the internal surface of the crystal and creates an evanescent wave, which projects orthogonally into the sample. Some of the energy of the evanescent wave is absorbed by the sample and the reflected radiation is returned to the detector. The blank was a clean sample-free crystal. Spectra were recorded in the absorbance mode at room temperature, in the mid-infrared region of 400 to 4000 cm^{-1}, using a Thermo Nicolet spectrometer (AVATAR 370 FT-IR) coupled to a computer loaded with OMNIC software.

2.9. Scanning Electronic Microscopy (SEM) analysis

SEM (JSM 6490LV, JEOL society, Japon) was used for the observation of morphological characterisation of the lyophilised mucilage powder after extraction. Sample particles

were fixed on the specific carbon film support. Then, all samples were coated with gold/palladium (Au/Pd) layer at 5 Volt during 5 min in a metallizer (Fine Coat – Ion Sputter JFC 1100, JEOl society, Japon). Their shape and surface characteristics were observed using secondary electron detector (SED) with a low vacuum mode.

2.10. Electrocoagulation-electroflotation (EC-EF) Unit and wastewater Preparation

The experimental EC-EF unit is shown in (Figure 2). It consists of an electrochemical reactor of 4 L, with two rectangular aluminium electrodes (27 mm x 17 mm x 1 mm) corresponding to a surface area of 4,59 cm^2. The electrodes were installed vertically in the middle of the reactor and are connected to a DC power supply (Statron TYP 3217, Germany) providing 0–30 V (0–10 A). The distance between the electrodes is equal to 1 cm [43]. The synthetic wastewater was prepared at an initial concentration of 300 mg/L using silica gel (Woelm Pharma) to simulate highly polluted industrial wastewaters [28,44]. A silica gel synthetic wastewater solution (2000 mL) was poured then purified in the electrolytic cell after switching on the generator. The samples were taken at a distance of 4.5 cm above the bottom of the reactor; this operation is carried out at regular intervals from 5 to 40 min of treatment time. The same initial parameter values are set namely: pH of the solution of 7.7, generator amperage of 0.2 A (current density *J* of 43.6 mA/cm^2), and conductivity (*k*) of 1.4 mS/cm. The high current density value (43.6 mA/cm^2) using a set of small electrodes was chosen in order to exhibit a strong effect on EC-EF. At high current density, the extent of anodic dissolution of aluminium rises, leading to a greater amount of precipitate to remove pollutants. In addition, bubble generation rate increases and the

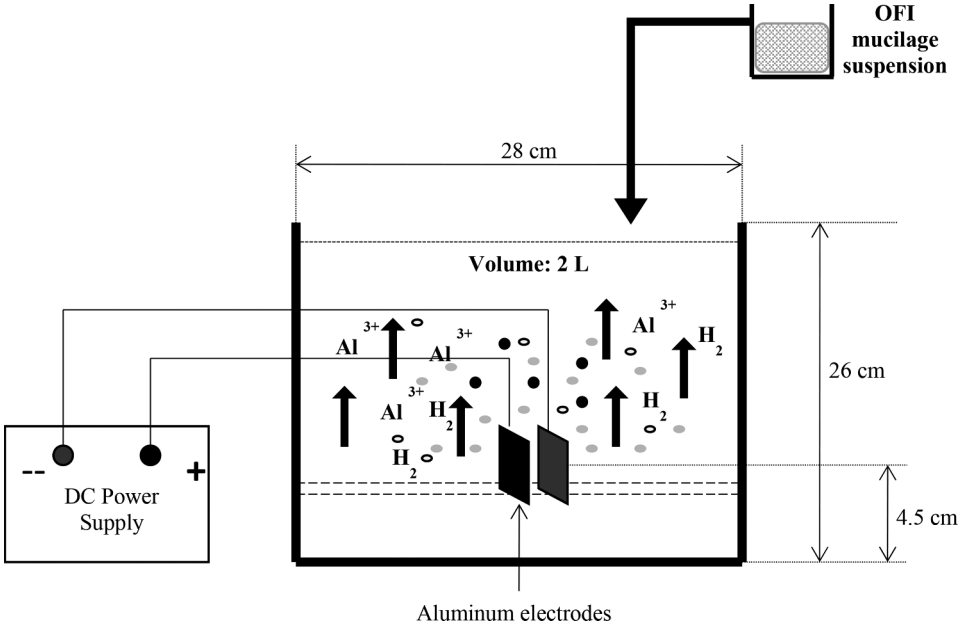

Figure 2. Electrocoagulation-electroflotation (EC-EF) experimental unit [28].

bubble size decreases with increasing current density. pH and conductivity of solutions during treatment were measured using an EC211 pH metre and an EC 215 conductivity metre (Hanna instruments, Italy), respectively.

In order to analyse its effect on EC-EF treatment efficiency, mucilage was added at different concentrations (2.5, 5, 7.5 and 15 mg/L). The same process is repeated under the same conditions.

2.11. Turbidity measurement

Samples were analysed by measuring turbidity, determined using UV-VIS spectrophotometer (Spectro scan 50, Japan). The concentration of the silica gel was estimated from a maximum wavelength of 740 nm (λ_{max}). Calculation of turbidity removal efficiency (TR %) after electrochemical treatment, assisted or not with OFI mucilage, was performed using the following formula (equation (3)):

$$TR\ \% = \frac{(C_0 - C)}{C_0}.100 \tag{3}$$

C: silica gel concentrations (mg/L) at different treatment times.
C_0: initial silica gel concentration (mg/L).

2.12. Statistical analysis

All the data are the average of three tests. An analysis of the variance (ANOVA) followed by Tukey's HSD test were applied using JMP software to display significant differences between varied parameters in conventional or ultrasound-assisted extractions, and between removal efficiencies of EC-EF processes (assisted or not with OFI mucilage). The degree of significance of the data is taken at the probability of $p < 0.05$.

3. Results and discussion

The moisture content of OFI cladodes was equal to 95.18 ± 0.14%. Young Algerian cladodes contain high levels of water (91–93%) [45], and varied moisture percentages from 5 to 95% were summarised in [19]. The pH of the cladode juice obtained after grinding was 4.86. This result is close to that found by [45]. These variations can probably be explained by the nature of the soil type, the variations of the climatic conditions, as well as the maturation stage.

3.1. Conventional Extraction at room temperature

Figure 3 shows the yield of conventional OFI mucilage extraction at room temperature (20–22°C). We observed an increase in extraction yield, which reaches the value of 45.83 ± 0.80% after 180 min. However, lower extraction yields were noted with values of 36.16 and 38.03% at extraction times of 60 and 120 min, respectively. Thus, the statistical analysis shows a significant difference ($p < 0.05$) with different significant letters (a > b). It can be deduced that the increase in mucilage yield depends on the increase in

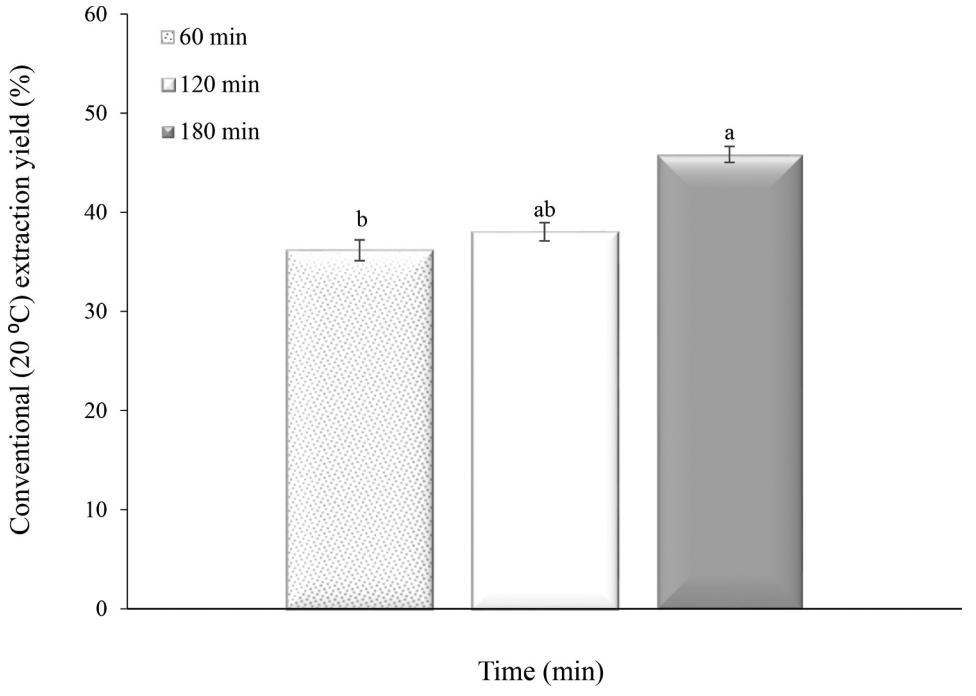

Figure 3. Effect of time (60, 120 and 180 min) on the conventional extraction yield of OFI mucilage at room temperature (20°C). Values with different letters (a, b) are significantly different (Tukey, $p < 0.05$) for each treatment.

extraction time, where an improvement in efficiency, of 9.67%, was observed between 60 and 180 min.

3.2. Conventional extraction at temperature of 80°C

The yield of the mucilage conventional extraction at a temperature of 80°C is shown in Figure 4. It can be seen that the optimal extraction yield value is equal to 54.16%, which was obtained at an extraction time of 180 min. Lower values of 36.71 ± 1.07 and 36.66 ± 1.04% were obtained at 60 and 120 min, respectively. Statistical analysis reveals that there is a significant difference ($p < 0.05$) concerning mucilage extraction yield of 60, 120 and 180 min. It can be deduced from Figures 3 and 4 that the increase in temperature from 20 to 80°C has a positive effect on the extraction yield; conventional extraction yield was improved after 180 min by 8.33% (optimal value 54.16 ± 1.54% at 80°C) in comparison to the optimal value of 45.83% obtained at room temperature.

3.3. Ultrasound-assisted extraction (UAE) of OFI mucilage

Ultrasonic-assisted extraction was carried out to improve conventional extraction yield. Two parameters were optimised: the extraction time (min) and the amplitude (%), both

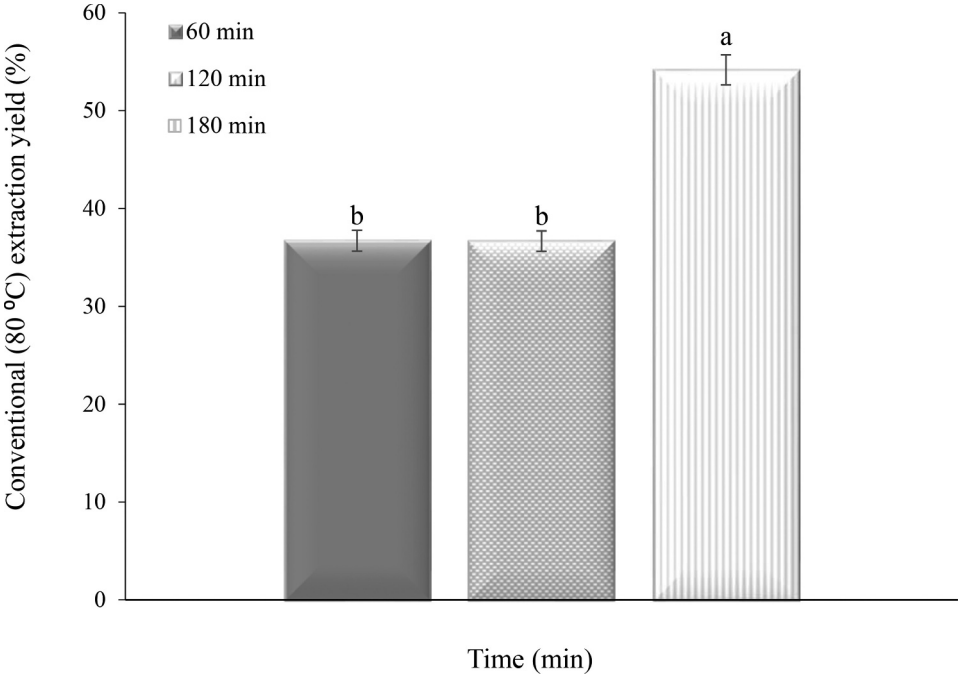

Figure 4. Effect of time (60, 120 and 180 min) on the conventional extraction yield of OFI mucilage at temperature of 80°C. Values with different letters (a, b) are significantly (Tukey, $p < 0.05$) different for each treatment.

parameters are related to the operating conditions of the sonicator and may significantly affect the ultrasound effect. In addition, this could minimise the process of energy cost.

3.3.1. Effect of extraction time

To optimise this parameter for UAE, different extraction times were tested: 1, 3, 5, 10, 20, 30, 40 and 50 min. Figure 5 shows the different yields of mucilage extracted by UAE and temperature values observed at different extraction times. Two other parameters were kept constant during these extractions; amplitude (60%) and the sample-solvent ratio (1:3)

It is clear from Figure 5 that the kinetic of mucilage extraction efficiency increases dramatically until reaching a maximum value at the extraction time of 10 min. Thus, from the 1 min, a value of 21.66 ± 1.53% with a temperature of 23 ± 1.20°C were observed, then the extraction yield progresses with the processing time until reaching its maximum value of 60.77%, for an optimum time of 10 min, where a temperature of 71 ± 1.13°C was noted. Beyond 10 min mucilage yield decreases to 35.09 ± 1.02% at a duration of 50 min as the last extraction time. Statistical analysis reveals that there was a significant difference ($p < 0.05$) when comparing different extraction times at that of 10 min. The optimal value of amplitude varies according to the plant material, the extracted compound source, and extraction conditions. For UAE of *Citrus limon* peels phenolic compounds, an optimal amplitude value of 77.8% was obtained among amplitude range of 20–100% [14]. For UAE of *Pistacia lentiscus* leaves phenolic

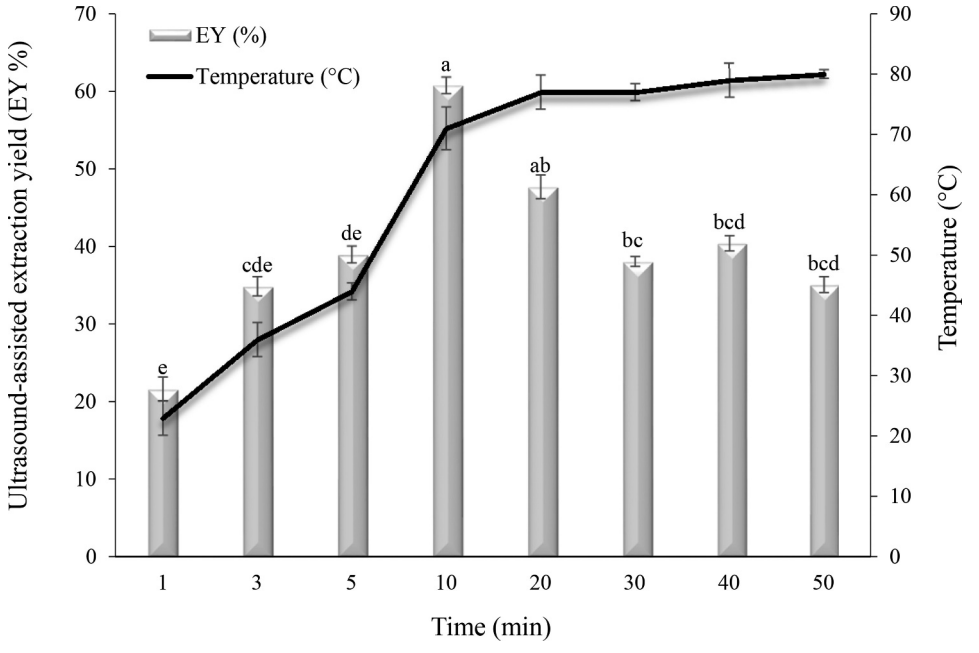

Figure 5. Effect of time on OFI mucilage Ultrasound-assisted extraction yield and on temperature (°C) at amplitude of 60%. Values with different letters (a, b, c, d and e) are significantly different (Tukey, $p < 0.05$) for each treatment.

compounds, an optimal amplitude value of 33.82% was obtained among amplitude range of 21.8–78.2% [12].

3.3.2. *Effect of extraction amplitude*

Different amplitudes (20, 60 and 100%) were varied for each extraction experiment where extraction time and sample-solvent ratio were kept at 10 min and 1:3, respectively. At the end of each extraction, the temperature of the vegetable matrix suspension was immediately measured. UAE yield of OFI mucilage at different amplitudes of 20, 60 and 100% is illustrated in Figure 6. We showed that the highest ultrasound-extraction yield is equivalent to 60.37% at an amplitude of 60%, where a temperature of 71 ± 1.13°C was noted. Beyond this amplitude (at 100%) extraction yield decreases to 46.03% and temperature reaches 77 ± 1.4°C. Amplitude value of 60% was considered as optimal for extraction performance. Statistical analysis shows significant difference ($p < 0.05$) between the different amplitudes (a > b > c).

From the results of Figures 3–5, the efficiency of OFI mucilage extraction by conventional method (at 20 and 80°C) was compared to that of ultrasound-assisted extraction (UAE). The condition parameters of both methods and the obtained results are summarised in Table 1.

In a recent study, OFI mucilage extraction was optimised for conventional and microwave-assisted extractions (MAE) [2]. Similarly, it was found that the extraction yield increases with treatment times (60, 120 and 180 min) for conventional extraction at 20°C (Table 1). However, the increase in temperature (80°C) improves the efficiency of up to

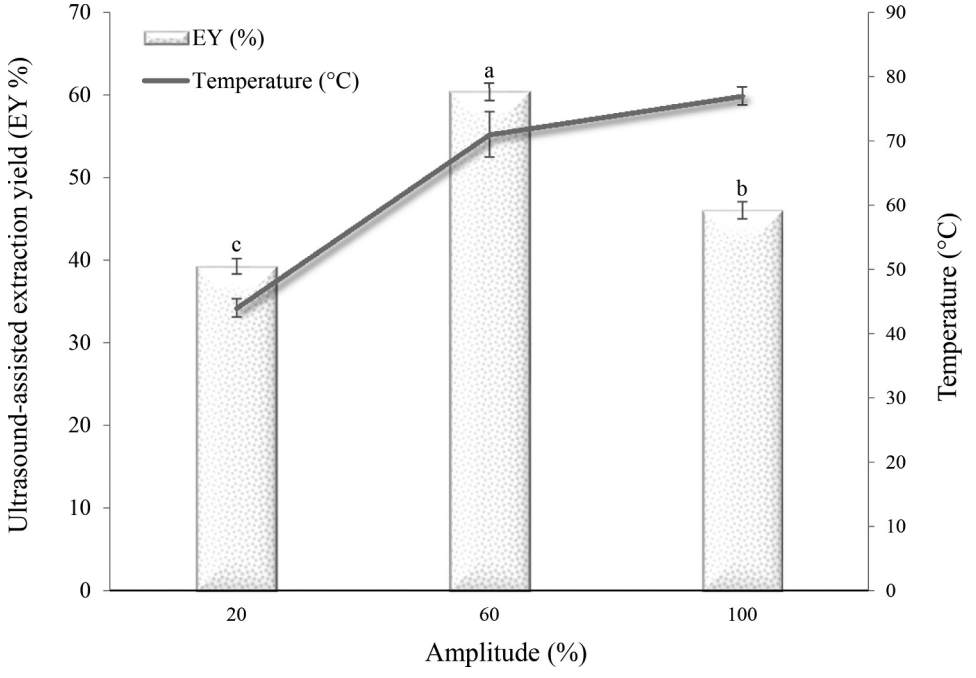

Figure 6. Effect of amplitude on OFI mucilage Ultrasound-assisted extraction yield and on temperature (°C) at extraction time of 10 min. Values with different letters (a, b, and c) are significantly different (Tukey, $p < 0.05$) for each type of treatment.

Table 1. Extraction yields of OFI peeled cladode mucilage from conventional extraction (CE) (at 20 and 80°C) and ultrasound-assisted extraction (UAE) (amplitude 20, 60 and 100%) at different extraction times. Values with different letters (a, b, c, d, and e) are significantly different (Tukey, $p < 0.05$) for each type of treatment.

Extraction method	Extraction time (min)	Extraction yield (%)
Conventional extraction (°C)		
20	60	36.16 ± 1.04 ^{b}
	120	38.03 ± 0.91 ^{ab}
	180	45.83 ± 0.80 ^{a}
80	60	36.71 ± 1.07 ^{c}
	120	36.66 ± 1.04 ^{b}
	180	54.16 ± 1.54 ^{a}
UAE (%)		
60	1	21.66 ± 1.53 ^{e}
	3	34.86 ± 1.23 ^{cde}
	5	38.95 ± 1.08 ^{de}
	10	60.77 ± 1.07 ^{a}
	20	47.69 ± 1.52 ^{ab}
	30	38.07 ± 0.64 ^{bc}
	40	40.40 ± 0.99 ^{bcd}
	50	35.09 ± 1.02 ^{bcd}
20	10	39.25 ± 0.93 ^{c}
100	10	46.03 ± 1.03 ^{b}

180 min. While their maximum yield is reached at 120 min, this can be due to the extraction process type, sample collection sites, environmental conditions or the plant harvest season.

Generally, the temperature has a positive effect on the extraction of phenolic compoundsfrom plant sources [46,47]. Studies have reported that mucilage increases exponentially with this parameter, where the effect of high temperatures on seeds decreases mucilage viscosity and produces less sticky seeds, therefore, mucilage can be easily released in the solution [48]. It has been reported by [49] that the increase in the extraction temperature could also change the structure of the vegetable matrix, and therefore facilitate the extraction process. The effect of time on yield was more noticeable in conventional extraction, maximum yield values were reached at 180 min (Table 1). In UAE, the effect of time was more distinct at lower temperatures (from 1 to 10 min) (Figure 5), and the yield reaches almost equilibrium after 10 min, while prolonging time decreases mucilage extraction. A similar trend was observed for flax gum [50], krueo pectin Ma Noy (*Cissampelos pareira*) [51], and polysaccharides from Inshell fruit sterculia (Buddha nuts) [52]. In addition, for Chinese yam polysaccharides extraction, an excessive extraction time induces polysaccharides gelatinisation, which hinders the extraction [53]. This study shows that the ultrasound-assisted extraction gave important extraction yields in comparison to those obtained by conventional method for both tested temperatures. This was noted by an improvement in yield by 6.21%, at the amplitude of 60% which gave the best yield result (60.77 ± 1.07%) in 10 min (Figure 5), compared to the conventional extraction where the yield was equal to 54.16% after 180 min at 80°C (Figure 4). It is important to note that prolonged contact time of solvent and plant material can lead to the degradation of active ingredients. Long-term ultrasonic irradiation can damage the quality of heat-sensitive materials [54].

Thus, ultrasound-assisted extraction of OFI mucilage allows a considerable gain of time. This is due to the cavitation effect produced by sound waves in liquid solutions, where the sono-chemical effects are reflected by performance improvements particularly with regard to certain solid-liquid extractions [55]. Another study explains that ultrasonic waves form tiny bubble specimens subjected to rapid adiabatic compression and expansion, cause a local increase of temperature and pressure in the medium [56,57]. Increasing the temperature in the medium has a direct impact on cavitation threshold, which will cause the collapse of the cavitary nucleus, therefore disturbs the tissues which improves the mass transfer to the solution [58]. When the temperature increases from 40 to 60°C during ultrasonic extraction, the viscosity and the density of the extracts decrease, which facilitates the penetration of the solvent more deeply in the sample matrix [55]. As the solvent moves deeper, its area of exposure increases which results in higher extraction efficiency.

The decrease of the extraction yield, beyond 60% at 100% amplitude (Figure 6), was explained by the fact that high amplitude could rise ultrasonic intensity. It is important to note that high amplitude values can lead to rapid deterioration of the ultrasonic transducer, resulting in liquid agitation instead of cavitation and poor transmission of ultrasound through liquid media. However, the amplitude must be increased when working on high viscosity liquids such as oils [7,59].This is because as the viscosity of the sample increases so does the resistance of the sample to the movement of the ultrasonic device [59].

It was reported that the high amplitude of ultrasonic waves in a liquid medium is accompanied by temperature increase that reduces cavitation during ultrasound extraction. This will counteract their effect [60], and decreasing performance extraction is due to the fact that the high-temperature values increase the steam pressure, which seeps into the bubble cavity, so many cavitation bubbles collapse less violently and reduce the effect of sonication [59,61].

3.4. Effect of initial concentration of mucilage on EC-EF process

EC-EF mechanism has been discussed by a number of authors [21]. The EC-EF process is based on three fundamental reactions following an electrical current, when Aluminium electrodes are used the following reactions occur (equations (4), (5) and (6)):

Oxidation of the Al metal at the anode

$$Al \rightarrow Al^{3+} + 3e^{-} \tag{4}$$

Water electrolysis at the cathode

$$3H_2O + 3e^{-} \rightarrow 3/2H_2 + OH^{-} \tag{5}$$

Hydrolysis of Aluminium ions:

$$Al^3 + 3H_2O \rightarrow Al(OH)_3 + 3H^{+} \tag{6}$$

It is well known that an oxidation reaction occurs at the anode that generates at a suitable pH a wide range of coagulants and metal hydroxides which destabilise and aggregate the particles in suspension by precipitation and adsorption of dissolved pollutants (electrocoagulation). Subsequently, the hydrogen gas released from the cathode by reduction reaction would help float the flocculated particles, this leads to electroflotation. The EC-EF process by these mechanisms (electrocoagulation and electroflotation) produces *in situ* coagulants ($Al(OH)_3$) by electrical dissolution of aluminium ions. While electroflotation allows flotation of the formed flocs through hydrogen gas formation at the cathode.

To study the effect of OFI mucilage on the efficiency of turbidity removal, different concentrations of OFI mucilage (2.5 to 15 mg/L) were tested in combination with EC-EF. The following initial conditions were used: silica gel initial concentration C_0 = 300 mg/L, distance between electrodes d = 1 cm, initial pH 7.7, conductivity κ = 1.4 mS/cm, current density j= 43.6 mA/cm^2. Turbidity removal efficiency was calculated according to equation 3.The obtained results are shown in Figure 7, where turbidity removal efficiency is illustrated in function of time (0, 1, 5, 10, 15, 20, 25, 30, 35 and 40 min) and mucilage concentration. It was found that turbidity almost decreases with increase of time. In addition, we observed that the highest values of turbidity were obtained with 5 mg/L of mucilage through the different processing time. The best turbidity removal efficiency was obtained with mucilage concentration of 5 mg/L at 25 min. The negative turbidity removal values found at 5 min can be explained by the polymerisation of dissolved aluminium in the form of aluminium hydroxide thus increasing the turbidity at the beginning of the treatment [28,44,62]. Beyond 5 min, removal efficiency increases in function of time where a maximum

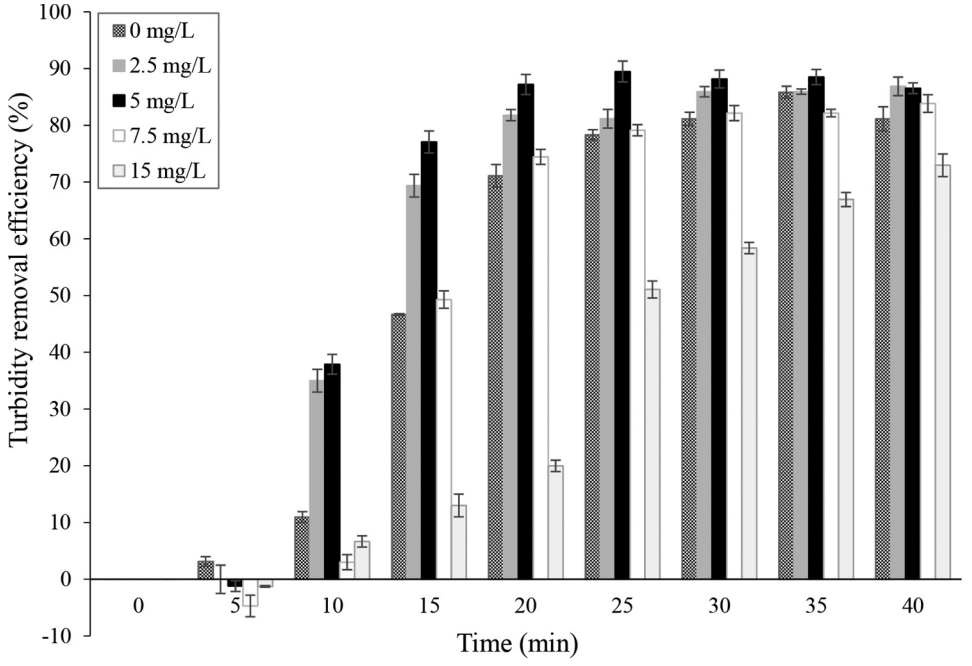

Figure 7. Effect of OFI mucilage dosage on the turbidity removal efficiency. Initial silica gel concentration C_0 = 300 mg/L, inter-electrode distance d = 1 cm, initial pH 7.7, initial conductivity κ = 1.4 mS/cm, current density j= 43.6 mA.

value of 89.47% was reached at 25 min with mucilage concentration of 5 mg/L, after that it is almost constant for all tested mucilage concentrations. The additional increase in mucilage concentrations above 5 mg/L does not further improve the removal efficiency (Figure 7), and a considerable decrease is noted with 15 mg/L of mucilage, this is probably due to the accumulation of mucilage polymers preventing interaction with suspended particles and saturation of the interaction sites [28]. However, the residual turbidity at 25 min was equal to 31.55 mg/L (Figure 7) when using 5 mg/L of mucilage; this means that the initial turbidity (300 mg/L) was reduced by approximately 10 fold.

Figure 8 shows the effect of mucilage addition (5 mg/mL) on the EC-EF turbidity removal efficiency. According to the statistical analysis, there is a significant difference between both experiments assisted or not with OFI mucilage from 10 min of treatment time. At 10 and 20 min, we obtained improvements of 27 and 16%, respectively. However, at 15 min significant improvement of 30% was noted, this allows a great saving of time. It was noted that pH of the solution tends to neutrality at the end of each treatment (data not shown); thus, mucilage addition had no effect on pH change in an EC-EF process as noted before [28,29].

According to these results, we note that EC-EF assisted with OFI mucilage allows turbidity removal of 77.06% at 15 min and a maximal value of 89.47% at 25 min, while in mucilage-free EC-EF the removal is equal to 88.50% after 35 min. This result is similar to that of [28] where 15% of improvement was observed when using OFI cladode juice.

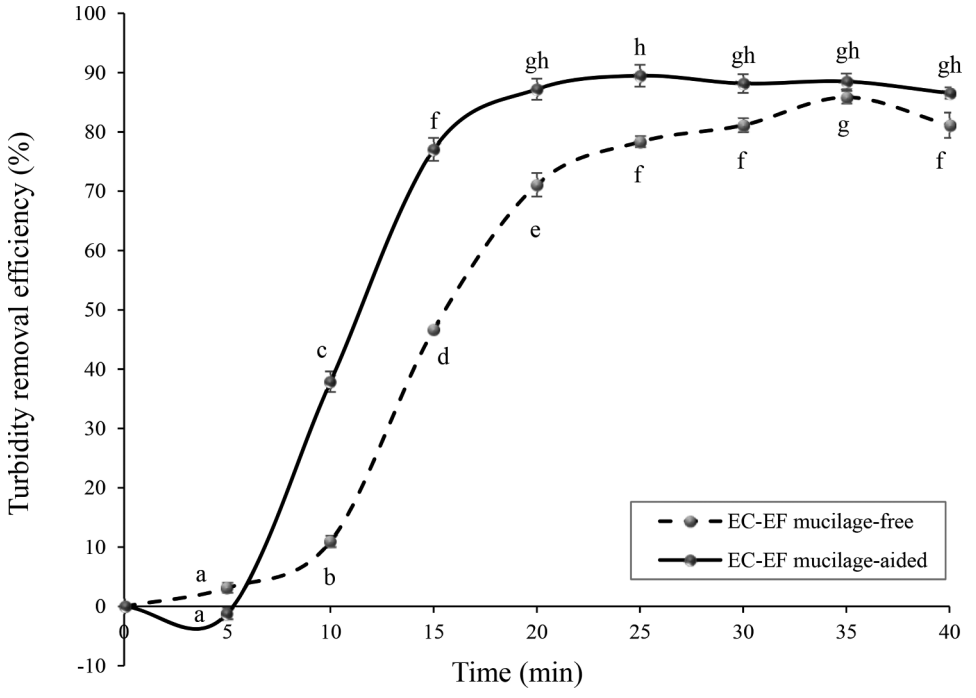

Figure 8. Effect of mucilage (5 mg/mL) on the improvement of the EC-EF process efficiency. Values with different letters (a, b, c, d, e, f, g and h) are significantly different (Tukey, $p < 0.05$) for each treatment.

The decrease of turbidity removal efficiency in both cases at different treatment times is explained by the dissolution of aluminium electrodes (equations (4), (5), (6)). Ionic species complexes (Al metal hydroxides) act as coagulants, which adsorb suspended particles and neutralise particles colloidal material, and leads to emulsion destabilisation and aggregates formation. Then flocs become easily eliminated [63,64]. According to [65], charged hydroxy-cationic gelatin complexes can effectively remove pollutants by adsorption; otherwise, in the vicinity of the cathode, the reduction of water occurs causing the formation of gaseous hydrogen. This release of gas occurs in the form of bubbles [66], and contributes to the mixing of the medium, these bubbles adhere to the formed solids in solution (adsorbent-pollutant), producing flocculated particles after electrocoagulation, and promote their flotation [29,63].

The improvement in turbidity removal efficiency in the presence of mucilage (Figure 8) may be related to an increase in the surface bio-coagulant binding sites, which promote pollutant or preformed aggregates adsorption and thus floc formation [67]. Other studies suggest that galacturonic acid of OFI mucilage may be the active component that provides coagulation activity by a mechanism that works mainly by adsorption [27]. A study on Okra mucilage (*Abelmoschus esculentus*), proves that the presence of hydroxyl groups (-OH) act as active sites for colloidal particle adsorption with mucilage [68].

Another more recent study reported by [29] stated that mucilage constituents could interact directly with pollutants (copper) in solution before copper adsorption

on EC-EF aluminium hydroxides, which is probably due to the porous structure of mucilage. However, the reverse mechanism could also occur, in this study it could be suggested that the adsorption of suspended molecules (silica gel) on the surface of the aluminium species could facilitate their attachment to OFI mucilage; consequently, this would increase the efficiency of coagulation as well as the elimination of turbidity.

OFI mucilage has the ability to reduce turbidity and therefore interacts with solid suspensions in contrast to synthetic coagulants as metal salts [69] that work by charge neutralisation. The use of zeta potential measurement and scanning electron microscopy images showing flocs formation, showed that the coagulation mechanisms of *Opuntia* spp. works mainly by adsorption mechanism. This process can be achieved by electrolytes of *Opuntia* spp., particularly divalent cations [27]. Mucilage contains about 20% of charged sugars with potential to interact with ionic species [70]. On the other hand, it is found that the cactus powder did not cause any significant effect on the final pH of treated water compared to chemical coagulants [71].

Note that small amount (5 mg/L) of OFI mucilage was required to improve EC-EF efficiency in the treatment of 2 L of solution. Only 0,005 to 0.015 g/L of Cladode powder of *Opuntia ficus-indica*. was effective to treat 0–125 NTU of synthetic wastewater [27].

It should be pointed out that the major objective of the paper is to study the benefits of using green chemistry (OFI cladode natural coagulant) to improve the efficiency of Electrocoagulation-electroflotation, and thus to be in consistency with sustainable development and biotechnologies applications. In this context, we opted for the use of ultrasound, as an eco-process, for the extraction of this natural coagulant. Up to now, previous works showed that the way of extraction does not really affect the intensification properties of OFI mucilage as an additive for different applications [2,9,30,32,72].

3.5. Mucilage identification

The final product is a relatively stable friable powder of white to off-white colour, with a pH of 6.14. The pH of point zero charge (pHpzc) is equal to 8 (Figure 9). The surface charge depends on pH: when the pH of the biomaterial in its aqueous environment, in our case mucilage in the treatment solution, is lower than the pHpzc, the surface of the mucilage of cactus is positively charged. While it is negatively charged when pH is higher than the pHpzc value [35,73]. *Opuntia ficus-indica* is a source of natural polyelectrolytes with negative surface charges, it has the ability to precipitate ions and particles from aqueous solutions through surface-active properties [4,27]. It was proposed for turbidity removal using zeta potential measurements and flocs electron microscopy images that the coagulation mechanisms of *Opuntia* spp. proceeds through adsorption. This process can be achieved by natural *Opuntia* spp. electrolytes (divalent cations), and bridging in which turbid particles are bound to a polymer-like material from *Opuntia* spp [27].

3.5.1. Colourimetric test (Biochemical assays)

The results of the colourimetric assays determined through spectrophotometric methods are expressed in µg/mg of extract (Table 2). The obtained values range from ≈ 11 to

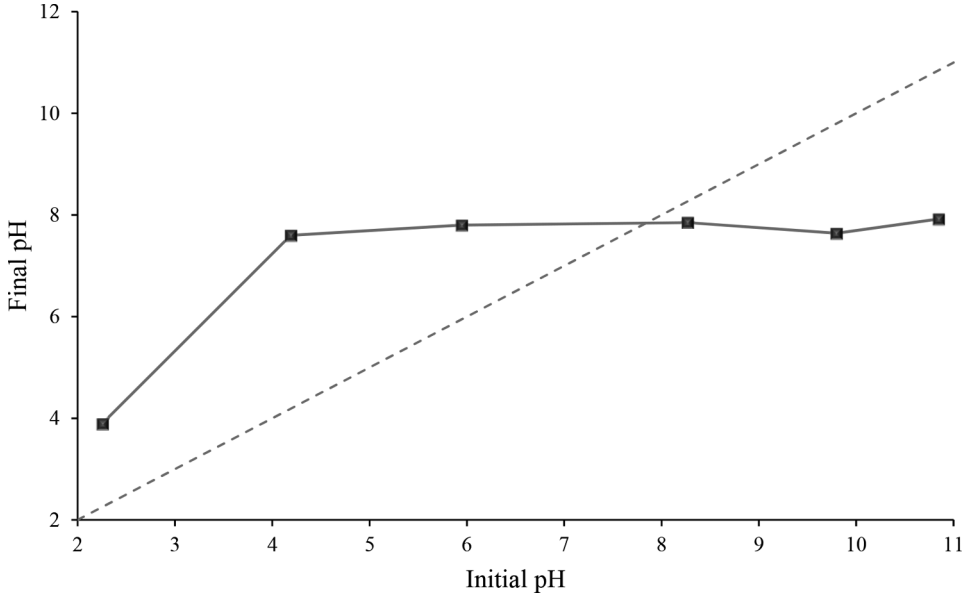

Figure 9. Determination of pH of point zero charge (pHpzc).

Table 2. Neutral and reducing sugar, uronic acid and protein contents of lyophilised OFI mucilage.

Parameter	Average value (µg/mg)
Neutral sugars	544.71 ± 4.00
Uronic acids	51.57 ± 1.33
Reducing sugars	10.55 ± 2.81
Proteins	64.00 ± 5.00

550 µg/mg. The values of neutral sugars, uronic acids and proteins are slightly lower than that obtained by [2]. However, neutral sugars were also found in the highest proportions. Mucilage is composed of 55 neutral sugars, mostly L-arabinose, D-galactose, L-rhamnose, and D-xylose and polygalacturonic acids which are involved in the mechanism of coagulation [4,74]. This explains the very low level of reducing sugars. The likely reason why proteins were not removed during extraction is probably due to the extraction method, mainly that ethanol was used for mucilage precipitation [75]. Thermal effects naturally match the requirements for the disruption process of tissues and, consequently, could be used to induce cell rupture for efficient extraction of useful components in plant tissues.

3.5.2. ATR-Fourier Transform Infrared (FTIR) spectroscopy

Characterisation using FTIR spectroscopy often results in the identification of functional groups and the modes of their attachment to polymer backbone [76]. The ATR-FTIR spectra of lyophilised OFI mucilage in the range 400–4000 cm^{-1} were taken to obtain information on the nature of functional groups at the surface of the natural coagulant

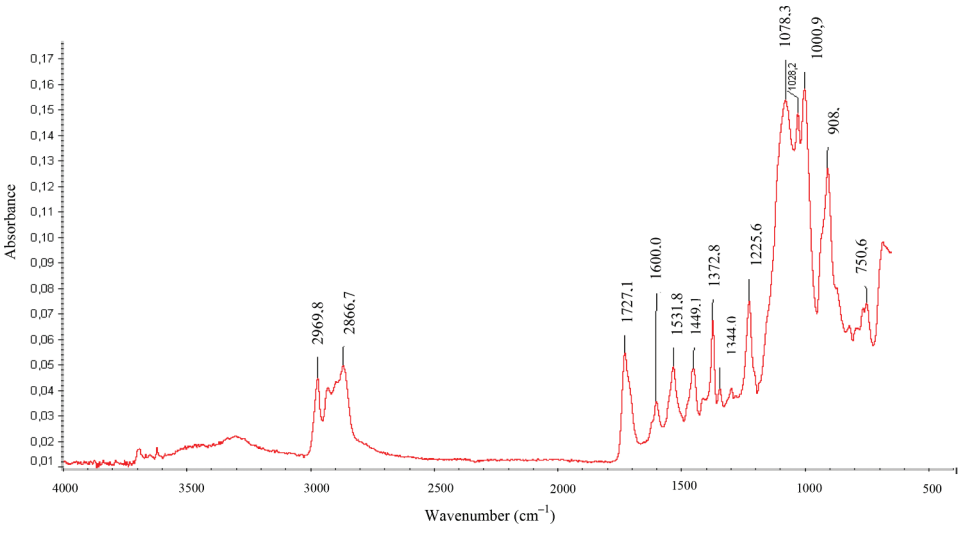

Figure 10. FTIR spectra of lyophilised OFI mucilage.

(Figure 10). The spectra show three characteristic regions: First 3200–3700 cm^{-1} region corresponds to O–H stretching bands envelope [77], or due to the over-lapping of O–H and N–H stretching vibrations [73,78]. The region between 3300 and 3400 cm^{-1} results from the presence of hydroxyl (-OH) groups [32,79], and – CO groups [76]. Second, 2500–3000 cm^{-1} corresponds to C-H (methyl) stretching bands [26,32], the pronounced bands observed at 2969 and 2866 cm^{-1} correspond to C–H stretching vibration [32,79], and to -CH_2 stretching (methylene groups) [19]. Third, the wavenumbers between 700 and 1800 cm^{-1} represent the fingerprint region envelope for carbohydrates [76,77].

In addition, the peak observed at 1727 cm^{-1} corresponds to carbonyl C = O stretch (carboxyl groups) [32,73]. Bands in the region of 1400–1450 cm^{-1} and 1600–1650 cm^{-1} for free carboxyl group [26], the band at 1430 cm^{-1} was assigned to – OH stretching (phenols) and – C = O stretching (carboxylates) [80]. Band at 1618 cm^{-1} was attributed to C = C ring (alkenes) [19] and to galacturonic acid [26], and that at 1574.2 cm^{-1} was related to the NH deformation of amines [73].

The intensity of the bands at 1100 and 1018 cm^{-1} correlated with the uronic acid content of pectic polysaccharides [81]. Bands at 1054, 1085 and 1154 cm^{-1} were assigned to – C–OH, – C–C–, and – C–O – vibration mode, respectively [32]. Other authors assigned the peak at 1072 cm^{-1} to – C–O–C – and – OH stretching (polysaccharides) [20]. The peak at 1027 cm^{-1} was assigned to R–CH_2–OH: Glucose units on biopolymers, according to [82]. However, it was reported that the band of 1080 cm^{-1} corresponds to P-OH stretching vibration (phosphates) [73]. The observed stretching vibration band at 1372.8 cm^{-1} is due to the asymmetric stretching of the carboxylic COO— double bond of deprotonated carboxylate functional groups [23]. The absorption peaks around 1225.6 cm^{-1} are indicative of P = O [73]. Peaks in the region of wavenumbers lower than 1000 cm^{-1} were attributed to aromatic groups [83]. The band at 908,2 cm^{-1} was attributed to S = O stretching vibration, and bands lower than 700 cm^{-1} to halogenated stretching [73]. A band at 1658 cm^{-1} corresponds to proteins [84].

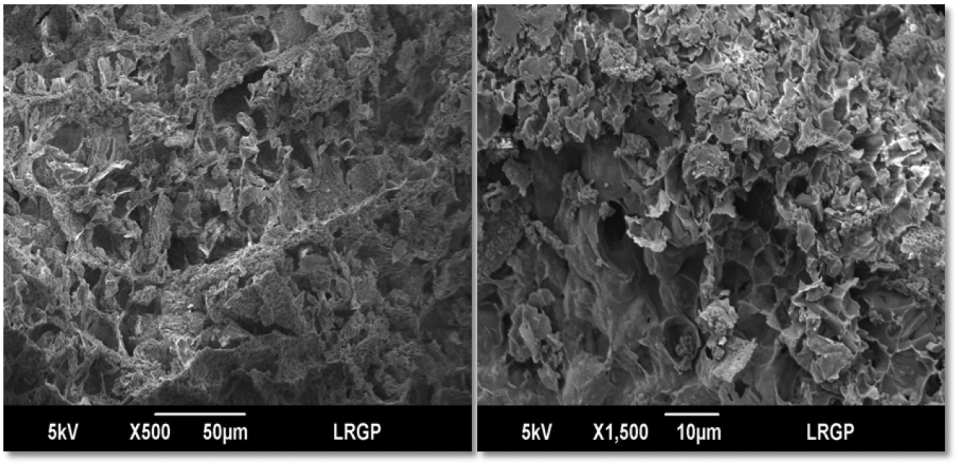

Figure 11. SEM images of lyophilised OFI mucilage at different magnifications.

The functional groups present in the mucilage have been identified in OFI FTIR spectra to be involved in pollutant adsorption [19]. FTIR analysis showed that carboxylic acid groups are certainly responsible for cationic metal (Cd, Pb) binding by dried cactus biosorbent [20]. ATR-FTIR result indicates that OFI mucilage contains a variety of functional groups such as carboxyl, hydroxyl, sulphate, phosphate, aldehydes, ketones and other charged groups [78].

3.5.3. SEM analysis of OFI mucilage

The structure of the lyophilised mucilage extract was observed by SEM (Figure 11). The images show extremely porous structure. This is a characteristic structure of adsorbents that show a three-dimensional network structure [85]. Their physical properties allow them to capture and store water and moisture in their microscopic holes, which are arranged in layers, pores and channels. The adsorption consists in the attachment of an element on the surface of the internal cavities of a porous material, or imprisonment in carbon nanotubes. The structural aspect of *Opuntia* spp. SEM images of parenchymal cells (100 µm), are highly similar to OFI mucilage SEM aspect in the work of [86]. Mucilage is produced mainly in parenchyma. Parenchyma cell walls consist of cellulose microfibrils associated together in strips, which are intertwined with each other in a relatively uncoordinated network. Stable and not flocculants suspensions in water have been obtained, which could present very interesting rheological properties [86]. Scanning electron microscope of arabinoxylan polysaccharides show a porous structure of a gel network with bacterial cells entrapped inside, suggesting that these gels can be potential candidates for the entrapment of probiotics [87].

4. Conclusion

The cladode of *Opuntia ficus-indica* was used for its richness in polysaccharide. Polymer polysaccharide demand is important on the industrial scale, especially in the agro-food

field; however, there is a growing interest for these polymers in wastewater treatment processes. These biocoagulants are meeting a great interest and had been widely used in purification, separation, and recovery processes. The goal of our work was to enhance OFI cladode mucilage extraction yield, and to promote its use as a natural coagulant in combination to EC-EF.

It was found that ultrasound-assisted extraction gave better extraction yields in comparison to conventional extraction. The results obtained with the conventional method at a temperature of 20°C reveal that mucilage extraction yield increases with time (36.16 ± 1.04% after 60 min and 45.83 ± 0.83% after 180 min). At 80°C, the extraction yield was improved (54.16 ± 1.54% after 180 min). However, ultrasound-assisted extraction at amplitude of 60% demonstrates that the yield increased with time until reaching optimal value at 10 min with 60.77 ± 1.07% of extraction yield. Thus, ultrasound-assisted extraction improved extraction yield by 6.21%.

OFI mucilage was tested on the treatment of a synthetic wastewater in combination with the electrocoagulation-electroflotation (EC-EF) process. The results obtained showed an optimal concentration of mucilage of 5 mg/L that allows a maximum turbidity removal efficiency of 89.47% at 25 min. It was also noted that in the presence of mucilage, 10 min of treatment were sufficient to obtain an improvement of 27% in comparison to the conventional process (mucilage-free EC-EF). This allows saving time and a considerable gain of energy. These results suggest an opportunity for the use of OFI mucilage as a natural coagulant in order to avoid the addition of other chemical adjuvants to EC-EF process.

Biochemical analysis confirms that OFI mucilage is composed in majority of sugars. SEM and ATR-FTIR analysis of OFI mucilage showed a similar morphology to that of adsorbents, and its richness in various functional groups that are probably involved with water impurities interactions, respectively.

Several studies highlight the benefits and applicability of OFI mucilage as a biocoagulant in wastewater treatment industries. However, in the context of Biotechnology and sustainable development, the combination of a natural coagulant for water treatment extracted by an eco-process encourages future application on the laboratory and on the industrial scale. In addition, other methodological approaches are necessary to examine the particular mucilage composition.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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