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Production of Highly Valued Vegetable Juice by Osmotic Distillation Technique: Optimization of Process Parameters

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Osmotic distillation (OD) was investigated for the concentration of vegetable juice, in sync with the increasing consumer interest towards natural and healthy foods. Celery juice first underwent a filtration step for removal of suspended solids, then a concentration step by OD. An OD plant was set up with a hollow fibre membrane module, where a concentrated solution of $CaCl_2$ (60% w/v) was used as extraction brine circulating in counter current with the celery juice. Preliminary tests were performed to identify the optimal process parameters (i.e., flow rate, volumes, etc.). Celery juice was concentrated from 3.7 to 22.4 °Brix after 200 min of processing. The content of beneficial compounds and antioxidant activity were preserved after the OD process obtaining a concentration of the solution without loss of phenolic compounds.

1. Introduction

The global market for fruit and vegetable juices has increased in the last years due to new trends to consume natural and healthy beverages. Consumers' attention is towards the maintenance of health and well-being, and hence they are more conscious of the intake of bioactive compounds obtained from vegetable matrices. In this framework, the juice sector is currently demonstrating a positive and dynamic growth, in sync with the choices of manufacturers for healthier ingredients in their juices together with the increase in purchasing power (Priyadarshini, 2018).

Celery (*Apiumgraveolens* L.) is a member of the family Apiaceae (synonymous with Umbellifereae). It is native of the Mediterranean area, cultivated in the lowland area of Italy later extending to other countries and by trading it diffused in India in the early 1900 up to nowadays where this plant is normally grown in the whole world (Khalil et al., 2015). The celery, in all its parts (stems, leaves and seeds), has been used since antiquity for the treatment of various illnesses due to anti-inflammatory properties (stems), for diuretic use or relaxing properties in tea. The main use is a popular flavouring agentin several food preparations (soups, salads) and as aromatic herb and spice in aromatherapy and in the mixture of cocktail drinks (Sellami et al., 2012). Celery is a rich source of vitamin C and various minerals (calcium, magnesium, potassium and sodium); the pleasantsmelling compounds (i.e. essential oils and volatile compounds) are largely confined to the green leaves of the plant (Khalil et al., 2015). Natural beverages made by celery and/or mixing it with other vegetables are highly valuable products rich in phytochemical compounds (i.e. phenolics), resulting also in a strong electrolyte replacement drink for the high level of minerals.

For the concentration of juices, membrane processes, especially osmotic distillation (OD), has gained a growing interest being non-thermal processes. Significant advantages in terms of improved product quality, the maximum achievable concentration of juices, lowering ethanol content in alcoholic beverages, low fouling index and low energy consumption have been demonstrated in OD and membrane distillation, compared to other pressure-driven membrane technologies such as microfiltration, ultrafiltration, nanofiltration and reverse osmosis (Cassano et al., 2020). These latter even if are consolidated systems for clarification, concentration, and purification in the food production show some typical drawbacks (i.e.concentration polarization, membrane fouling, and constraints on the maximum achievable concentration) that limit their application in liquid food concentration.

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The OD process is driven by a vapour pressure difference between porous hydrophobic membrane surfaces, through which only water vapour molecules can pass. Therefore, concentration polarization is not a limiting factor and high solid contents can be achieved.
The osmotic solution used in juice concentration by OD should be no-toxic, no-corrosive, and should have a high osmotic activity to maintain a lower vapour pressure and to maximize the driving force.
To our knowledge, the recent applications of OD were on wine and beer dealcoholisation (Corona et al., 2019; Russo et al. 2019; Liguori et al. 2019) and concentration of several fruit juices (Cassano et al., 2020; Alves et al., 2006; Cissé et al., 2005), and just for a vegetable (broccoli) juice (Yilmaz et al., 2018). In the juices concentration, hollow fibre membrane contactors are preferred over flat sheet membranes due to their high specific area per unit volume, easy scale-up and low manufacturing cost (Cassano et al., 2020).
The aim of this study is the identification of the optimal OD process parameters (i.e. flow rate, volume ratio of feed and brine extraction) for the production of concentrated celery juice with a high amount of natural bioactive compounds. An OD pilot plant using a hollow fibres membrane contactor (Liqui-Cel) has been set up. Finally, the effects of the OD on some nutritional and healthy parameters of celery juice has been also evaluated.

2. Materials and methods

2.1 Materials

Celery (*Apiumgraveolens*L.) was purchased at the local market. Leaves and core were removed and the stems were initially washed, drained, and cut into pieces. The juice was extracted by squeezing the celery stems using an electric extractor (HR1869/80, Philips). The extracted juice was centrifuged at 8000 rpm for 5 min and filtered by Whatman filter paper to remove suspended solids. Subsequently, the obtained juice was treated by the OD plant.

2.2 Lab-scale plant and experimental conditions

The lab-scale plant is equipped with a membrane module, pumps, flowmeters, a manometer to check feed pressure at the inlet of the module, a thermostatic water bath to regulate the stream's temperature at 20°C and K-thermocouples to checkthe fluid temperature. The membrane module (1x5.5 Liqui-Cel) has the following characteristics: polypropylene membrane, 1800 cm^2surface area, 42 μm thickness and 14 cm length, 40% porosity, 0.03 μm membrane pore diameter. It consists of 2300 fibres with dimensions: 11.5 cm length, 220 μm inner diameter and 300 μm outer diameter.
The extraction brine solution (S) is calcium chloride dihydrate at 60% (w/v) which is fed in the tube side of the membrane in a counter-current mode to the feed (f) (celery juice) which flowsoutside the bundle of fibres. The streams are recycled during the trials. A schematic view of the experimental set-up is presented in Fig.1.
Preliminary tests were performed at two flow rate of celery juice: 6 and 12 L/h. The following combinations were evaluated: $Q_f = Q_s$; $Q_f = 2Q_s$; $2Q_f = Q_s$. At the best condition in terms of final soluble solids concentration in feed, the volumes ratio $V_f/V_s = 1/1$ and $V_f/V_s = 1/2$ were tested.
In all the tests, the feed volume was 280 mL, the overall process time was 200 min.

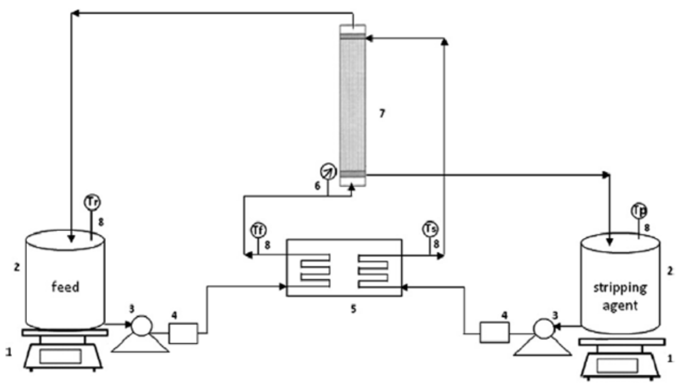

Figure 1: Experimental setup of OD plant: (1) stirrer, (2) feed and stripping tanks, (3) pumps, (4) flow meter, (5) thermostatic bath, (6) pressure gauge, (7)membrane module, (8) thermocouples in feed (Tf), stripping (Ts), retentate (Tr) and permeate streams (Tp)

2.3 Chemical and Physical Characterization

Total soluble solids (TSS) content was measured by using a hand type refractometer (Atago, Co., Tokyo, Japan). Colour measurements were performed using the CIELab colour space ($L^*a^*b^*$) with CR-300 colorimeter (Konica Minolta, Inc., Osaka, Japan). Total colour differences (ΔE) were also calculated periodically at 80, 120, 160 and 200 min of the process using initial juice as reference, according to the following equation:

$$\Delta E = [(\Delta a^*)^2 + (\Delta b^*)^2 + (\Delta L^*)^2] \quad (1)$$

The total phenols content was determined by Folin-Ciocalteau method (Singleton & Rossi, 1965): the juice (initial and concentrated samples) (1 mL) was mixed with 15 mL Na_2CO_3 (20%) and 7.5 mL Folin-Ciocalteau reagent and the final volume was made up to 100 mL with distilled water. After 2 h of reaction at room temperature, in the dark, the absorbance at 765 nm was determined.Gallic acid as reference compound was used for calibration curveand results were expressed as gallic acid equivalent (GAE)mg/L juice(Di Matteo et al., 2012).

DPPH radical scavenging activity of different samples of juice was determined according to the method of Brand-Williams et al. (1995) with minor changes (Liguori et al., 2018; 2020; Apicella et al., 2019): juice sample (0.1 ml) was added to 3.9 ml of 6 $*10^{-5}$mol/L DPPH methanol solution. The absorbance at 515 nm was measured after the solution had been allowed to stand in the dark for 40 min. A blank experiment was also carried out applying the same procedure to a solution without the test material and the absorbance was recorded. The antioxidant activity (AA) was expressed as percentage inhibition of DPPH calculated according to the following equation:

$$(\%)\ \text{inhibition of DPPH} = 100- [(A_{blank} - A_{sample})/ A_{blank}] \quad (2)$$

2.4 Statistical analysis

All the analyses were performed in triplicates. Results were reported as mean and standard deviation and data were subjected to analysis of variance (ANOVA). The significance of differences ($p < 0.05$) among samples was determined by Student’s t-test. Statistical analysis was performed using Analysis Lab software.

3. Results and discussion

Preliminary experiments were performed by changing the feed and stripper flow rates ratio in order to characterize hydrodynamic conditions in the hollow fibre membrane module used in this work. In details, Q_f = Q_s was set equal to 12 L/h; then Q_f = 6L/h and Q_s = 12 L/h ($2Q_f = Q_s$) were set and the opposite Q_f = 12 L/h and Q_s = 6 L/h (Q_f=2 Q_s). In all these experiments, the celery juice was the feed and the solution of calcium chloride dihydrate at 60% (w/v) the stripper.

The evolution of experimental transmembrane flux of water in feed was reported in Figure 1a. As expected, water flux decreasedas function of the timeincreased. In particular, during the first 100 min of the process, different water flux values were registered among the three operative conditions applied.

In details, when both streams circulated at the same flow rate (Q_f=Q_s= 12 L/h) the water flux was about 0.38 kg/m^{2}hwhile at the end of OD process reached a value of 0.03 kg/m^{2}h. In these conditions, the juice reached a final volume concentration of 80% with a TSS content of 16.1°Brix respect to the initial value of 3.5 °Brix.In the other cases ($2Q_f$=Q_s and Q_f=$2Q_s$), the water flux during the OD process was lower allowing to reach a final juice concentration of about 70% with a TSS content of 12.8 and 10.2° Brix, respectively. In these latter conditions, a lower juice concentration was achieved probably due to a lower efficacy of exchange between both sides of the membrane.

On the basis of the previous results, the flow rate of both streams was set equal and to 12 L/h. Different volume ratios were investigated in further tests.

The evolution of TSS and the volume reduction factor (VRF) in the juice were reported in Figure 1b. The results referred to two different volume ratios for the streams volumes ratio V_s=V_f and V_s=$2V_f$.

When V_s=$2V_f$ was used, a higher value of VRF was reached (0.83) with respect to V_f=V_s(VFR=0.74). The higher volume reduction measured when the volume of stripper was twice that of juice, with respect to V_f=V_s was due to the higher waterflux from feed to the extraction brine solution (Russo et al., 2013), which caused a stronger concentration in TSS of the juice equal to 22.4°Brix (V_s=$2V_f$) with respect to 16.1°Brix (V_f=V_s).

In the best process conditions (Q_f=Q_s; V_s=$2V_f$), the concentration level reached in celery juice was about 80%.Similar results were reported by other authors, who applied the OD process for fruit juice concentration. Cassano et al. (2007) obtained a cactus pear juice concentration of 82%after 250 min of process with the same membrane module configuration, used in this study,but with a bigger (1.4 m^2) membrane surface area. Valdes et al. (2005) reached a concentration of noni juice of 75% after 60 min with ahollow fibre membrane with 0.58 m^2 of surface.

In our knowledge, only the paperby Yilmaz et al. (2018)applied the OD process to a vegetable juice. In particular, broccoli juice was concentrated of 83%by a capillary membrane module after 340 min. Once identified the best conditions (Q_f=Q_s; V_s=2V_f), the main bioactive properties of the celery juicebefore and during the process (at 80, 120, 160 and 200 min) were analysed.

The initial celery juice had a total phenols content of 141.1 mg/L juice. As shown in Figure 2a, after 120 min of the concentration process, corresponding to 9.7 °Brix of TSS in juice, no significant differencesin total phenolswere found with respect to the initial content. Then, an increasing of TSS to 14 and 22.4 °Brix after 160 and 200 min respectively was observed. Correspondingly, an increase of total phenols concentration was found up to182.2 mg GAE/L in the final juicewith respect to the initial value of 141.1 mg GAE/L. The antioxidant activity measured in initial juice was 2.2%. During the OD process, as well as for the total phenols trend, the increase in TSS content of the juice resulted in a significant increase in the antioxidant activity (Figure 2b). Similar results were obtained by Valdes et al. (2009) who found an increasing concentration of phenolic substances during the OD concentration of noni juice on semi-industrial scale. Acorrelation coefficient of 0.9031between total phenols and AA during the OD concentration process was found. This result can be justified by a different concentration factor registered for antioxidant activity (about 7) and that measured for phenolic compounds (about 1.3)

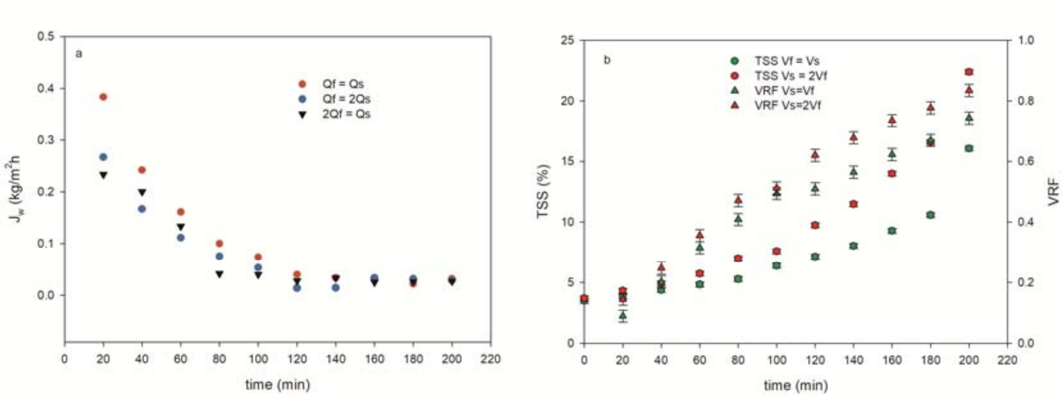

Figure 1: Experimental data of transmembrane flux of water a) at different flow rate ratios: Q_f = Q_s; Q_f = 2Q_s; 2Q_f = Q_s; and *b) total soluble solids (TSS) and volume reduction factor (VRF):* V_f/V_s= 1/1 and V_f/V_s= ½ *as a function of the time during OD of celery juice.*

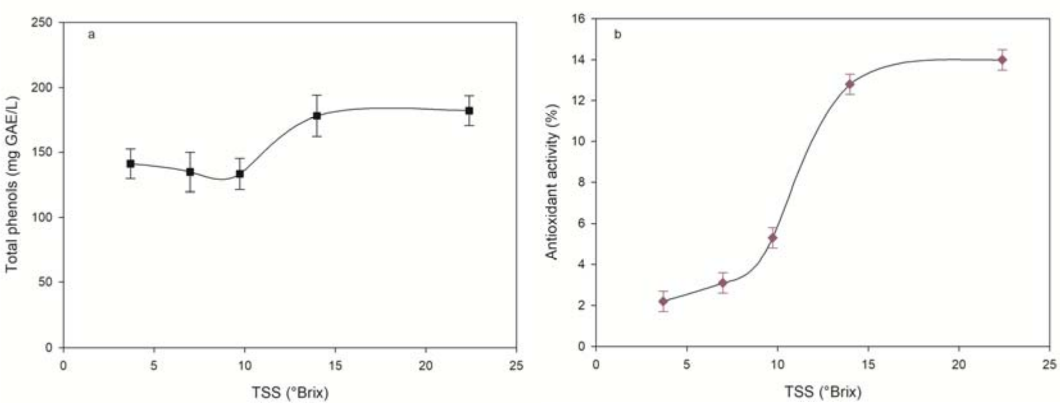

Figure 2: Content and evolution of a) total phenols ; b) Antioxidant activity in the celery juice as a function of the TSS during the OD concentration process.

The colour parameters of the celery juice during the OD processwere reported in Table 1.

The results highlighted significant changes during the concentration process. In details, it was observed that celery juice turned dark, its colour progressively intensified, confirmed by the hue angle index and the total difference with respect to the initial juice due to the concentration process. The same trend was also detected in the concentration of noni juice by Valdes et al. (2009).The appearance of a dark colour could be also a consequence of oxidation phenomena of phenolic compounds which can occur due to the operating conditions of the lab-scale process.

Table 1: Colour parameters of juice samples during OD concentration process.

time (min)	L*	a*	b*	h	ΔE
0	32.06±0.11[a]	2.97±0.43[ac]	3.27±0.26[a]	47.9±3.4[a]	0.0[a]
80	32.30±0.01[b]	2.33±0.04[b]	4.09±0.05[b]	60.4±0.1[b]	1.1±0.1[b]
120	31.68±0.03[c]	2.65±0.21[c]	3.53±0.20[a]	53.2±3.7[a]	0.4±0.3[c]
160	30.27±0.08[d]	3.44±0.04[a]	3.34±0.49[a]	44.0±2.6[a]	3.6±0.3[d]
200	28.75±0.38[e]	2.82±0.02[c]	2.53±0.14[c]	41.8±0.7[c]	11.5±0.6[e]

4. Conclusions

The application of osmotic distillation process for the concentration of celery juice was evaluated. The influence of some process parameters (i.e. feed and brine flow rate and volume ratios) was investigated in order to identify the optimum conditions. At the best identified conditions (Q_f=Q_s; V_s=2V_f), a 6-fold concentration of total soluble solids was achieved with a change of TSS from 3.7 up to 22.4 °Brix. The results in antioxidant activity and polyphenols showed an improvement in the health quality of the celery juice due to the concentration of total phenols and an increase of radical scavenging activity. Changes in colour were registered due to both the concentration process and probably to the recirculation of stream which can influence the enzymatic oxidation/browning of the juice. The preliminary results obtained in this work showed that the concentration of vegetable juice by osmotic distillation could be a promising alternative to obtain high-quality juice and the feasibility of the operation may be assured by the high value of the juice.

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