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Osmotic and membrane distillation for the concentration of tomato juice: Effects on quality and safety characteristics

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ABSTRACT

The aim of this study was to evaluate the utilization of osmotic distillation, membrane distillation, and coupled operation of these systems as an alternative to the conventional thermal evaporation (CTE) technique for the concentration of tomato juice. Some physicochemical characteristics of the products obtained by different techniques were determined. The samples concentrated by membrane systems were more advantageous by means of many parameters investigated, especially for color, hydroxymethylfurfural (HMF) and furan formation. HMF and furan contents were increased up to three to four times after CTE while there was no statistical change after membrane concentrations. Ascorbic acid and dehydroascorbic acid levels were decreased significantly after all concentration operations. Nonetheless, as total vitamin C content, membrane systems were more advantageous compared to CTE. Sensorial evaluation also showed that, except consistence, products obtained by membrane techniques gained higher scores than thermally concentrated products.

Industrial Relevance: The quality of tomato paste is dependent on process conditions used to convert the tomato pulp into paste. Conventional thermal evaporation may result in the deterioration of product quality by damaging heat sensitive tomato juice components as well as inducing color changes. Moreover, some mutagenic and carcinogenic compounds such as furan and HMF may be formed. Concentration of tomato juice using membrane systems can be proposed as the promising alternative to the CTE since most of the characteristics, especially color, are preserved, and HMF and furan formations are reduced significantly by these processes.

1. Introduction

Tomato is widely consumed either fresh or after processing into various products. Regular consumption of these products is associated with reduced risks of chronic diseases such as certain cancer types and coronary heart disease (Çapanoğlu, Beekwilder, Boyacioglu, Hall, & de Vos, 2008; Podsedek, Sosnowska, & Anders, 2003; Vallverdu-Queralt et al., 2013). In order to obtain more stability as well as reducing the packaging, transport and storage costs, tomato is processed into concentrated products such as tomato paste which is then diluted for formulation of ketchup, sauces and other value-added products (Anthon & Barrett, 2012). The quality of tomato paste is dependent on raw materials as well as the processes used to convert the tomato pulp into paste. In the common production technique of tomato paste, the juice obtained from the fruit moves through a series of evaporators to remove the water at elevated temperatures. Although this conventional thermal evaporation is performed under a vacuum, the technique always results in the deterioration of product quality since this process causes damage to heat sensitive tomato juice components as well as inducing undesirable color changes. Moreover, this heat treatment may also be responsible for the formation of some mutagenic and carcinogenic compounds such as furan and hydroxymethylfurfural (HMF) (Anese, Manzocco, Calligaris, & Nicoli, 2013).

In-depth description of precursors and mechanisms of furan and HMF accumulation in food have been published by various researchers (Anese & Suman, 2013; Crews & Castle, 2007; Moro et al., 2012). Briefly, HMF can be formed as an intermediate in the Maillard reaction, which occurs when carbohydrates are heated in the presence of amino acids or proteins, or alternatively, by thermal degradation of sugars under acidic conditions (Anese & Suman, 2013). Although HMF has been used for years as a quality indicator of thermally processed food, several studies showed that HMF may also induce genotoxic and mutagenic effects in bacterial and human cells and promote colon and liver cancers in rats and mice (Anese & Suman, 2013). Furan has also been identified as a contaminant of heat-treated food resulting from the thermal degradation and re-arrangement of various precursors, including sugars, amino acids, ascorbic acid (AA) and polyunsaturated fatty acids (Crews, Roberts, Lauryssen, & Kramer, 2009). The presence of furan in processed food is a concern because furan is listed as "reasonably anticipated to be human carcinogen" in the Department of Health and Human Services Report on Carcinogens and is considered "possibly

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carcinogenic to humans" by the International Agency for Research on Cancer (Fan & Geveke, 2007).

Since those of deterioration in quality seems to be unavoidable in conventional thermal evaporation, alternative processes that avoid high temperatures of operation need to be used to overcome these unwanted changes. In this respect, attempts to replace the conventional evaporative concentration method with membrane technology have led to proposals of reverse osmosis as an alternative methodology. However, this method is proved to be effective only for preconcentrating the tomato juice up to a level of 8.0–9.0 °Bx. At higher concentrations, the fouling problem and the high osmotic pressure of the juice sharply reduce the water flux through the membrane (Petrotos et al., 2010). Technological advances related to the development of new membranes and improvements in process engineering have been proved to overcome these limitations. New membrane processes including membrane distillation (MD), osmotic distillation (OD) and integrated membrane processes may contribute to the improvement of concentrated juice processing (Jiao, Cassano, & Drioli, 2004).

MD as a separation process involves transport of water vapor through the pores of hydrophobic membranes, where the driving force is the vapor pressure difference created by the temperature difference across the membrane (Bahamanyar, Asghari, & Khoobi, 2012). Because MD can be carried out at the atmospheric pressure and at a temperature, which can be much lower than the boiling point of the solution, it can be used to concentrate solutes sensitive to high temperature (Jiao et al., 2004). OD is another process in which the driving force is also vapor pressure difference between the two sides of a non-wetted porous hydrophobic membrane. The same membrane modules used in MD can also be employed in OD. The difference between MD and OD is the way to generate the driving force. In OD the vapor pressure gradient results from a concentration gradient from the feed to the permeate side of the membrane, generated using an extracting solution on the permeate side of the module (Cojocaru & Khayet, 2011). OD can be performed at ambient temperature, which is particularly attractive for concentration of heat sensitive products such as juices.

Since these membrane technologies could be actually considered as a potential alternative to conventional thermal evaporation, the aim of this work was to evaluate the effects of membrane processes on some quality characteristics of concentrated tomato juice. There are also some publications describing the effect of heat processing on the nutritional properties of concentrated tomato products, whereas the formation of thermal process contaminants, HMF and especially furan, have not been considered. Therefore, another aim of this study was to investigate how the production processes affect the formation of HMF and furan.

2. Materials and methods

2.1. Materials

The hot-break tomato juice to be concentrated was supplied from a large tomato paste processing plant in Adana, Turkey. The other batch of tomato juice (cold-break) was prepared from fresh tomatoes, which were purchased from a local market. Tomatoes were washed with tap water to remove residues and extraneous matter, adhering to the fruit, were cut in quarters and tomato pulp was produced using a laboratory type of pulper (Imperia Solemio, Italy), which could separate the seeds and peel from the tomato pulp. Then, tomato pulp was subjected to heat treatment up to 65 °C and immediately cooled to room temperature.

2.2. Concentration procedures

After serum–pulp separation by centrifugation (9000 *g*, 10 min), concentration of the serum part was performed using a laboratory-size membrane module (MD 020 CP 2N, Mycrodyn, Germany) having 40 polypropylene capillaries with a 2.8 mm outer and a 1.8 mm inner diameter. The effective internal area of the membrane was 0.1 m^2 and the average pore size was 0.2 μm. In the MD process, tomato juice (1000 g) was pumped in the tube side and the deionized water was recirculated in the shell side of the membrane in a countercurrent mode by using peristaltic pumps (Heidolph PD 5206, Germany). The recycle flow rate was 30 L/h on both sides. The temperature difference between the feed (30 °C at the inlet) and the permeate side (10 °C at the inlet) of the membrane was maintained constant at 20 ± 1 °C using two heat exchangers (Lauda RE 2025, Germany). The same experimental procedure was used in the OD process; however, in this case, calcium chloride dehydrate (65%, *w*/*w*) was used as stripping solution and circulated in the shell side of the membrane, and the experiments were carried out at room temperature. The initial weight of the stripping solution was five times higher (5000 g) compared to that of the tomato juice in order to prevent a significant dilution with consequent decreasing of the driving force during the process. In the coupled operation of OD and MD, tomato juice was pumped in the tube side and the osmotic solution was circulated in the shell side of the membrane. A temperature difference was imposed between the shell (10 °C) and the tube side (30 °C), in order to provide an additional driving force. After each trial, the membrane module was cleaned by a five step cleaning process according to the method used by Onsekizoglu, Bahçeci, and Acar (2010).

The average initial evaporation flux obtained by OD (1.07 kg m^{-2} h^{-1}) was higher than that observed in MD (0.94 kg m^{-2} h^{-1}). However, it was decreased more rapidly in OD due to the dilution effects, and reached a similar value of 0.88 kg m^{-2} h^{-1} after 4 h of operation in both processes. In the coupled operation of OD and MD, the flux was enhanced (1.97 kg m^{-2} h^{-1} initially) by adding two driving forces.

Tomato juice (serum part) with an initial concentration of 5.0–5.5 °Bx was concentrated up to 35–40 °Bx by membrane systems, and the concentrated serum was recombined with the tomato pulp that was separated before. Final soluble solid contents of the samples were between 23–25 °Bx. The tomato juice was also concentrated by conventional thermal evaporation (CTE) at a temperature of 70 °C by using a rotary evaporator (Ika RV 10). Concentrated samples were stored at −20 °C for further analysis.

2.3. Analytical measurements

2.3.1. Soluble solids, pH and total acidity

The total soluble solids expressed as °Brix and measured with a refractometer (Atago Pal-3, Japan). The pH was measured using a pH meter (Adwa AD1020), and total titratable acidity (expressed as a percentage of citric acid) was analyzed by titrating the product with 0.1 N NaOH to pH 8.1.

2.3.2. Total phenolic compounds

Total polyphenol content was quantified using a colorimetric assay and Folin–Ciocalteau's phenol reagent. Before the analysis, the samples were subjected to a procedure of extraction/hydrolysis. For that, 1 mL of 1 N HCl was added to 1 g of the sample, vortexed for 1 min and incubated at 37 °C for 30 min. Later, 1 mL of 2 N NaOH in a 75% methanol solution was added, and the resulting mixture was vortexed for 2 min and incubated at 37 °C for 30 min. Then, 1 mL of 0.75 M of metaphosphoric acid was added after being vortexed for 2 min and the sample was centrifuged at 9000 g for 10 min. The supernatant was removed and transferred into a 10 mL flask and the pellet was resuspended in 1 mL of acetone:water (1:1, *v*/*v*), vortexed for 1 min and centrifuged at 9000 *g* for 10 min. Finally, both supernatants were combined and the 10 mL volumetric flask was filled with acetone:water (1:1, *v*/*v*) (Perez-Conesa et al., 2009). Afterward, 2.2 mL of 0.2 N Folin–Ciocalteu's phenol reagent and 1.6 mL of a 2 M Na_2CO_3 solution were added to 200 μL of sample solution. The samples were allowed to stand in the dark for 2 h at room temperature before the absorbance at 750 nm was