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Optimization of the sugar hydrothermal extraction process from olive cake using neuro-fuzzy models

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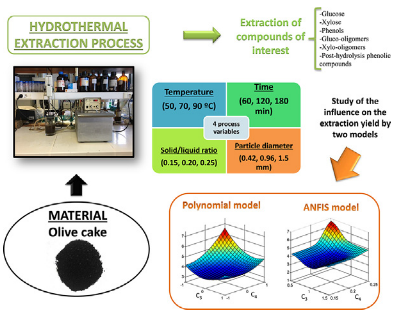
GRAPHICAL ABSTRACT



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ABSTRACT

The optimization of the hydrothermal extraction step in the biorefinery scheme in order to obtain sugars and antioxidants from olive cake was carried out. This process using water and low temperatures for the olive cake has not been previously studied by other authors. Central Composite Design of experiments was carried out. Four variables and three levels for each variable were tested: temperature (50, 70 and 90 °C) time (60, 120, 180 min), average particle diameter (0.42, 0.96 and 1.5 mm) and solid/liquid ratio (0.15, 0.20, 0.25). The main aim was to understand the relationship between the process variables and the yield of extraction of glucose, xylose, polyphenols and oligomers. The variables which most influenced the process were particle diameter and solid/liquid ratio. The experimental values were adjusted to a classical polynomial model and to a neuro-fuzzy system. The neuro-fuzzy demonstrated to be much more accurate when predicting the experimental values.

1. Introduction

Olive oil production is one of the biggest agri-food industries in Andalusia (Spain) and throughout the Mediterranean area. As a result, a large amount of by-products and wastes are generated by this industry. For many of them there is no an alternative use. In other cases, this kind of wastes causes problems in their management. One of these by-products is the "alpeorujo", which is the aqueous phase generated in the extraction of olive oil. For that reason, it presents high moisture content. Nowadays, the "alpeorujo" is dried and then, the remaining olive oil in the residue is obtained by extraction with hexane. The solid residue generated in the extraction process is currently used as a fuel (AAE, 2018; Buratti, et al., 2016). In this sense, Quesada et al. (2018) proposed that a previous step of washing would be desirable to reduce the water soluble compounds content, to enhance the calorific value and therefore, improve the properties of the material as a fuel.

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A biorefinery scheme is a clean and efficient option in order to obtain energy and products with commercial interest from olive cake (López et al., 2010; Dávila et al., 2017). The first step in the biorefinery should be the hydrothermal treatment, which is the simplest extraction process. In that process, the extracting agent is water and high temperature should be applied in order to increase the extraction achieved. The products of interest generated in this step are sugars, polyols, antioxidants, furfural and hydroxymethylfurfural among others. After that, more severe treatments such as chemical treatments and high pressure processes are conducted. The aim is to obtain a solid with high calorific value. Fermentable media for subsequent production of bioalcohol could be obtained (Alfaro et al., 2010; Feria et al., 2011; Lemes et al., 2016).

Mathematical adjustment models are very powerful tools when it comes to studying complex systems. The variables of operation in complex systems influence the process in a combined way, causing synergies between them. When a design of experiments is carried out, the operating variables are set in a range and the response variable values are obtained experimentally. In a Central Composite Design of experiments (CCD) the classical polynomial fit has been widely used. However, the neuro-fuzzy model has been reported to give better results when it is applied to multivariable systems. The process optimization is crucial in order to operate in a viable way from the environmental and economical point of view. The neuro-fuzzy model is a more accurate alternative for the optimization of the processes (Jiménez et al., 2008; Zamudio et al., 2011; Rodríguez et al., 2008; Ronda et al., 2015; Iáñez-Rodríguez et al., 2017; Calero et al., 2018).

In short, the main objective of this work is the optimization of the hydrothermal extraction step in the biorefinery scheme in order to obtain sugars and antioxidants from olive cake. For this purpose, a Central Composite Design of experiments was carried out. Temperature, time, average particle diameter and solid/liquid ratio were the selected variables. The main aim was to understand the relationship between the process variables and the yield of extraction of glucose, xylose, polyphenols and oligomers. The experimental values were adjusted to a classical polynomial model and to a neuro-fuzzy system. The results obtained by both methods were compared.

2. Materials and methods

2.1. Raw material

The olive cake (OS) was provided by a company located in Linares-Baeza (Jaén, Spain). The sample was prepared for subsequent fractionation (three particle sizes were selected using average diameters of 0.42, 0.96 y 1.5 mm).

2.2. Chemical characterization of olive cake

2.2.1. Chemical analysis

Firstly, removal of soluble hot water extractives was performed according to the TAPPI T 257. Then, ethanol–benzene extractable compounds were determined according to TAPPI T 204. Finally, lignin and holocellulose were determined by quantitative acid hydrolysis with 5 ml of 72% sulfuric acid for an hour (TAPPI T-248-em-85), and quantitative posthydrolysis with 4% sulfuric acid (adding water until 148.67 g) at 121 °C and 2 atm for 60 min in order to ensure quantitative conversion of oligomers into monomers. Before HPLC analysis, the solid residue from posthydrolysis process was recovered by filtration and considered as Klason lignin. The monosaccharide content was determined by HPLC in order to estimate the contents of cellulose (as glucose) and hemicelluloses (as xylose) in the samples. The moisture content in the raw material was considered as water in the material balances. Chromatographic determination was performed using a Metrohm 940 professional IC Vario equipped with Metrosep Carb 2 250/4.0 column under the following conditions: mobile phase, 100 mM NaOH and 10 mM NaAc; flow rate, 0.500 ml/s; and column temperature, 303 K.

2.2.2. Elemental analysis

Elemental analysis of dried olive cake sample was carried out by combustion analysis using an Elemental Fison's Instruments EA1108 CHNS.

2.3. Hydrothermal process and saccharides and oligosaccharides determination

Olive cake particles and water were mixed in the desired proportions. The mixture reacted in a 1 L jacketed glass reactor connected to a temperature-controlled bath. The reactor was fitted with double two-blade turbine impellers. The vessels were heated up to maximum temperatures in the range 50–90 °C with external hot water. The temperature of the process was automatically controlled with a thermostatic bath equipped with a stirring system. Several isothermal hydrothemal treatments were carried out in order to study the effect of the process on glucose, xylose, polyphenols and oligomers production. The yield of each compound was studied. Based on previous experiences, different temperatures, residence times, average particle sizes and liquid/solid ratios were selected to carry out the experiments. The values selected for each variable are indicated in the experimental design. Two preliminary tests were carried out under the central operating conditions (a temperature of 70 °C, a processing time of 120 min, 0.96 mm of particle diameter and a solid/liquid ratio of 20%).Then, several tests were performed using temperatures from 50 to 90 °C and times over the range of 60–180 min, particle diameter between 0.42 and 1.5 mm and solid/liquid ratio between 15 and 25%. A constant volume of 600 ml was used in all tests in order to ensure efficient mixing in the reactor. The model determined the need to carry out 24 experiments around the central one.

At the end of the hydrothermal treatment, the solid residue was recovered by filtration and then it was washed with distilled water for gravimetric yield determination. An aliquot of the liquor was filtered through 0.20 mm membranes and it was used for direct HPLC determination of monosaccharides and polyphenols concentration by 4 amineantipirine method (norm UNE-ISO 6439:2013). A second aliquot of the liquor (25 ml) was subjected to quantitative posthydrolysis (with 4% sulphuric acid at 121 °C and 2 atm for 60 min) before HPLC analysis. Operation conditions in HPLC were described in the Section 2.2.1. The increase in the concentration of monosaccharides caused by posthydrolysis could be considered as the concentration of oligomers bound to oligosaccharides. Otherwise, the sum of glucose and xylose represented the holocellulose content.

The content of sugars was presented as the percentage of dissolved sugars with respect to the total dissolved solids in the liquor.

$$ES(\%) = \frac{[ES]\cdot V\cdot Fh}{(1-R)\cdot Mt}\cdot 100 \quad (1)$$

where ES (%) represents the percentage of dissolved sugars with respect to the total dissolved solids in the liquor, [ES] is the concentration of sugars dissolved in the solution in mg/L, V is the total volume used in each experiment and Fh is the hydration content which is 162/180 for glucose and 132/150 for xylose. R represents the solid yield (so much per one expressed) and Mt is the total mass of olive cake used in each experiment.

For the case of the oligomers:

$$EO(\%) = \frac{(([EO]\cdot Fd)-[ES])\cdot V\cdot Fh}{(1-R)\cdot Mt}\cdot 100 \quad (2)$$

where EO (%) represents the percentage of dissolved oligomers with respect to the total dissolved solids in the liquor, [EO] is the sugars concentration after carrying out the hydrolysis to the liquid, Fd is the dilution factor (3.6 for all cases). For the case of polyphenols, the