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Phenomenological determination of mass transfer parameters of oil extraction from grape biomass waste

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ABSTRACT

Grape seeds are one of the by-products of the grape processing industry and, if not appropriately treated, can cause serious environmental problems. However, this biomass residue is a source of oil with nutritional, cosmetic and pharmaceutical applications. This work aimed at evaluating the effect of the drying temperature of grape seeds on the oil yield. The oil was obtained using different solvents in the Soxhlet and ultrasound extraction techniques. The yield was high at elevated temperatures, when using dichloromethane, ethyl acetate and hexane. The highest yield obtained using the Soxhlet extraction was 17.99%, and the highest yield obtained using the ultrasound extraction was 12.90%. In order to describe the extraction process, a phenomenological mathematical model was developed. The model was validated in the conditions of the highest and lowest drying temperatures. The values of the mean relative errors between the experimental values and the values predicted by the model were 4.27% and 8.68% for the Soxhlet and the ultrasound extraction, respectively. Thus, as the model satisfactorily described the extraction process, a routine to simulate the commercial scale extraction was also developed.

1. Introduction

Grape is listed in the ranking of the Food and Agriculture Organization of the United Nations (FAO) as a fruit with extensive areas dedicated exclusively to its cultivation (FAO, 2016). According to estimates, 71% of the world's production of grapes is aimed at winemaking, 27% is consumed as fresh fruit, and 2% is consumed as dried fruit (Demiral and Ayan, 2011). Concerning the wine production, the pressing of grapes produces approximately 12 million tons of bagasse every year (Minjares-Fuentes et al., 2014). This product consists of seeds (50%), stalks (25%), and skins (25%) (Farías-Campomanes et al., 2013).

The bagasse from winemaking, as well as other agricultural residues, is considered an environmental problem (Sun et al., 2016). Some wineries utilize the bagasse as animal feed (Demiral and Güngör, 2016). However, this utilization is not advised due to the high fibrousness and low digestibility of the residue. An alternative use is the extraction of edible oils from the seeds due to their oleaginous content (Beveridge et al., 2005) and phenolic and phytochemicals compounds (Porto et al., 2013). Based on these factors and the benefits for the human health, the grape seed oil is a product of commercial interest (Maier et al., 2009).

Solvent extraction is considered a traditional extraction method and an essential part of the manufacturing process of pharmaceuticals, cosmetics, perfumes, food, biofuels, and fine chemicals (Chemat et al., 2012; Rabrenović et al., 2014). Nonetheless, this process can be costly, pose a risk to human health, and contaminate the environment (Todd and Baroutian, 2017). Moreover, traditional extraction methods (maceration, hydrodistillation, infusion, pressing, percolating, and Soxhlet) still have some limitations, such as high solvent consumption, extended extraction period, and low yield and quality of extracts (Rombaut et al., 2014).

Regarding the search for green and cost-effective technologies, ultrasonic wave-assisted extraction has been used to obtain

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Nomenclature

C_E	Concentration of oil in the fluid phase (kg m^{-3})
C_I	Concentration of oil in the macropores (kg m^{-3})
C_{I0}	Initial concentration of oil in the macropores (kg m^{-3})
C_S	Concentration of oil in the solid phase (kg m^{-3})
C_{S0}	Initial concentration of oil in the solid phase (kg m^{-3})
C_{T0}	Total oil concentration (kg m^{-3})
DCM	Dichloromethane
D_{eff}	Effective mass diffusivity (m^2 min^{-1})
$\overline{D}_n$	Mean particle diameter (m)
D_{Sauter}	Sauter mean diameter (m)
EA	Ethyl acetate
HE	Hexane
K	Equilibrium constant
k_F	External mass transfer coefficient (m min^{-1})
k_P	Internal mass transfer coefficient (min^{-1})
m	Initial ratio of oil concentration in the porous phase in relation to the total concentration
m_{oil}	Mass of extracted oil (kg)
m_{seed}	Mass of grape seed (kg)
nexp	Number of experimental data
r	Radial coordinate (m)
R	Radius of equivalent sphere (m)
R^2	Coefficient of determination
RME	Relative mean error
t	Time (min)
ε	Particle porosity
φ_n	Mass fraction

different compounds from plant matrices. This extraction technique is known as a green technique since it reduces the energy consumption and allows the use of alternative solvents and renewable materials while maintaining the safety of the process and quality of the extract (Chemat et al., 2012). Ultrasonic extraction is an easily automated technique that improves the process efficiency through local changes in temperature and pressure, requiring less time, and allowing the simultaneous extraction of several components. This process also allows a more efficient penetration of the solvent in the solid matrix, intensifying the mass transfer due to the cell rupture caused by the collapse of cavitation bubbles near the cell walls (Yang et al., 2016).

Phenomenological mathematical modeling is an essential tool for industrial operations since it enables the prediction and scale-up of the process (Gaspar et al., 2003). Regarding extraction processes, the mathematical model and its kinetic parameters can be applied to optimize the operation and estimate the energy and time consumption, and the extraction yield (Krishnan et al., 2013). In this context, some studies have evaluated mathematical models when describing batch extraction processes. However, the majority of the published studies focus on pseudo-kinetic models, requiring the experimental determination of the pseudo-chemical order of extraction and the kinetic constant. Among these studies, one can mention Meziane et al. (2006) who studied the kinetics of oil extraction from olive foot cake, Dutta et al. (2016) who studied the batch extraction of *Crotalaria juncea* seed oil using 2-propanol, Rezazi et al. (2016) who evaluated the essential oil extraction process from *Marrubium vulgare* L., and Dias et al. (2017) who studied the ultrasound-assisted extraction of bioactive compounds from dedo de moça pepper. Meanwhile, the development and assessment of phenomenological models applied to oil extraction, although widely employed in studies to optimize and control industrial processes, are scarce. Among these studies, it is possible to cite Franco et al. (2007) who studied the ethanolic extraction of *Rosa rubiginosa* soluble compounds, Carrín and Crapiste (2008) who studied the oil–solvent extraction in a multistage horizontal extractor, and Baumler et al. (2011) who studied the solvent extraction of vegetable oils.

Some works in the literature evaluate the extraction of grape seed oil using different techniques. One can cite, for example, the study of supercritical CO_2 extraction with constant temperature and different flows and pressures from seeds with and without an enzyme pretreatment (Passos et al., 2009), and with different pressures and temperatures (Passos et al., 2010). Other studies include the effect of the operating parameters on the pressurized extraction with ethyl acetate and dichloromethane (Freitas et al., 2013), the application of an ultrasound-assisted extraction method (Porto et al., 2013), the study of cold press extraction (Rombaut et al., 2015), and the assessment of conventional extraction with n-hexane and supercritical CO_2 extraction (Mohamed et al., 2016). Nevertheless, few studies numerically evaluate the grape seed oil extraction (Beveridge et al., 2005; Duba et al., 2015; Song et al., 2017).

The drying operation is usually considered a preprocessing step since it results in a more significant contact between the solvent and the solute to be extracted. Moreover, this operation is also related to the obtention of high extraction rates. Some authors have evaluated the influence of the drying conditions on the oil yield extracted from raw materials such as lemon verbena (Shahhoseini et al., 2013), lemon balm (Argyropoulos and Muller, 2014), sea buckthorn berries and leaves (Kyriakopoulou et al., 2013), peppermint leaves (Torki-Harchegani et al., 2016), and papaya seeds (Chielle et al., 2016).

In a previous study (Johann et al., 2016), the drying kinetics of grape seeds was evaluated. First and third type boundary conditions were tested to describe the drying process. As a conclusion, it was observed that the convective condition adequately described the process, in addition to the fact that the drying air velocity did not have a significant influence on the drying rate. Thus, the present paper is a continuation of the previous one, and it is motivated by the importance of conducting studies to assess alternative uses of the winemaking bagasse other than its disposal.

The aim of this paper is to evaluate the effect of the drying temperature on the pretreatment (30, 40, 50, 60, 70, and 80 °C) of the Bordo grape regarding the oil yield using the Soxhlet and ultrasound-assisted extraction techniques, create a mathematical model that represents the extraction process, and obtain the kinetic parameters.

2. Materials and methods

The seeds of the Burgundy grape variety were acquired at Vinícola Vinhos Randon Ltda, in the city of Pinheiro Preto, SC, Brazil. To remove the skins, stems, and sugar, the seeds were placed in sieves and washed in running water. Then the seeds were washed again with deionized water and deionized water heated at 100 °C. After cleaning, they were packed in plastic bags and stored at −15 °C.

The initial humidity of the whole seeds was determined by placing them in an oven at 105 °C for 24 h, and determining the Sauter mean diameter, D_{Sauter}: