Effect of the Process Parameters on the Oil Extraction Yield During Supercritical Fluid Extraction from Grape Seed

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Abstract

The supercritical fluid extraction is an effective technology in the recovery of biologically valuable compounds from grape seed. In this work, the effect of the main operational parameters involved in the supercritical CO₂ extraction of oil from grape seeds was investigated. The oil extraction yield oil was determined as a function of pressure (20 MPa and 30 MPa), temperature (313 K and 333K), solvent flow (1 g/min and 5 g/min) and particle size (0.4 mm and 0.80 mm). The oil extraction yield increased with increasing pressure and decreasing temperature, due to the influence of pressure and temperature on oil solubility. On the other hand, the smaller was the particles, the higher the final yield. Although the oil extraction yield increased with the increased solvent flow, it should be noted that specific CO₂ consumption also increased.

Keywords: Operational parameters, Vitis labrusca, supercritical CO₂ extraction, seed oil, optimization
1. Introduction

Making use of waste products is a general research trend in the scientific community. In many countries, grape seeds are considered a disposable waste material by most wineries. Therefore, reusing the waste from this industry is of great interest [1]. In addition, grape seeds oil is rich in unsaturated fatty acid and vitamin E (tocopherols and tocotrienols) and exhibits high antioxidant activities which make it increasingly attractive in culinary, pharmaceutical, cosmetic and medical applications [2].

Traditionally, seed oils are extracted either by an organic solvent or mechanical techniques. Organic solvent extraction gives better extraction yield, but the technique requires solvent recovery through distillation which may degrade thermally labile compounds [2]. The conventional industrial extraction of vegetable oil from vegetable seeds involves several stages, and extraction with n-hexane is the most important [3]. However, this organic solvent is non-selective and achieves the simultaneous removal of non-volatile pigments and waxes, resulting in viscous, dark-coloured extracts contaminated with solvent residues. This makes them difficult to handle without further refinement, and may even dictate the future commercial viability of oil [4]. Furthermore, the presence of traces of residual solvent in the final product makes the process less attractive from health and environmental point of views [2]. In this regard, the use of supercritical carbon dioxide (sc-CO$_2$) has become an alternative solvent for seed oil extraction, because it can achieve comparable oil yield with respect to the conventional organic solvent extraction with better product quality similar to that of mechanical pressing [2]. In addition, sc-CO$_2$ is essentially non-toxic, non-flammable, inexpensive at the industrial level, can be recycled, has easily accessible supercritical conditions, and is totally dissipated from extracts at atmospheric pressure avoiding the necessity of further expensive and harmful refining treatments [5]. In fact, the supercritical fluid extraction (SFE) of oil from grape seeds has already been researched previously, showing quite satisfactory results [1,2,4,6].

On the other hand, the oil extraction yield depends on the operating conditions employed and several works have demonstrated the importance to study parameters like pressure, temperature, flow rate and the particle size in order to optimize the process. Therefore, in this work, the effect of the main process variables affecting the sc-CO$_2$ extraction of oil from grape was corroborated.

2. Methodology

2.1 Raw material

Grape (*Vitis labrusca*) was obtained from winemakers in southwest Colombia. Following the procedure of Duba and Fiori [2], at the winery, stalks were separated
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from the seeds and skins. The mixture of seeds and skins was stored at 253 K before drying. The samples were dried at 333 K for 48 h, and then the skins and seeds were separated by means of sieves and further cleaned manually. Then, the seeds were stored in dark under vacuum at ambient temperature. Finally, dried grape seeds were milled for two cycles of 10 s and then sieved.

2.2 Installation and operation procedure

All the extractions were done in a homemade semi-continuous flow apparatus that consisted of a pure CO₂ feed line and an SFE equipment with 0.05 L. A heating jacket was placed around it to control the temperature inside. The CO₂ flow rate was fixed with a pump, while pressure was controlled by a backpressure regulator. The quantity of CO₂ was measured using a mass-flow meter that was placed at the end of the line. An approximate amount of 5 g of sample was introduced in the vessel forming a fixed. A wire mesh was placed on top of the bed to prevent particles entrainment. The extractor was then closed and pre-heated. After that, the CO₂ was pumped in, and, once the desired pressure was reached, the BPR was opened to provide a continuous flow of the CO₂ through the bed. The CO₂ was distributed through the bed and after the BPR, this was depressurized; the solvent power of the supercritical fluid dropped, and the oil was precipitated in a previously weighed glass flask. The amount of CO₂ circulated was read by the mass flow meter. When the operation was over, the apparatus was depressurized and the exhausted sample unloaded. After each treatment, lines were cleaned before to start the next experiment. The experimental error was deduced from selected tests that were repeated six times. The maximum standard deviation in extraction yield was 3.0 %.

For a better comparison of the oil yield extractions, the operation was over at the same consumption of CO₂ (3 kg) for all the treatments. This, to recollect the sample at the end of the line stage of extraction. For this, a previous extraction curve was done at the highest pressure (30 MPa), the lowest temperature (313 K), a mass flow of 5 g/min and a particle diameter of 0.40 mm.

Additionally, conventional extraction was carried out for comparison, using 10 g of milled seed, and 0.2 L of n-hexane in a Soxhlet apparatus during 4 h.

2.3 Experimental design

The experimental design proposed to visualize the influence of the main operating variables was general factorial design ($2^4$). The design factors considered were pressure (20 MPa and 30 MPa), temperature (313 K and 333K), solvent flow (1 g/min and 5 g/min) and particle diameter (0.4 and 0.80 mm). Experimental conditions were selected in order to test the best conditions of solubilities predicted and taking in count the limitations of the experimental installation used.
A total of 16 experiments resulted from the experimental design, however, just selected experiments were carried out in randomized run order to test the influence of the main parameter. Finally, five experiments and its duplicate were carried out. This was done for a better visualization of the effects. The only response variable selected was oil extraction yield expressed as percent of dry weight/initial sample weight. Table 1 shows the factor levels corresponding to the experimental matrix design and their physical values, along with the results obtained for the selected experiments.

### 3. Results

The results of experimental tests are reported in the present section. Table 1 shows the results obtained for the different extracts in terms of extraction yield (percent dry weight/initial weight) under the different experimental conditions tested. A column including the CO₂ density has also been given for information. The effect of each single process variable was analysed separately. It is worth noting the grape seed oil extracted by conventional Soxhlet produces similar results than the obtained by SFE.

#### Table 1. Factor levels of the experimental matrix design and results obtained for yield (% dry weight/initial weight, along with values of density)

<table>
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<tr>
<th>Run</th>
<th>P (MPa)</th>
<th>T (K)</th>
<th>CO₂ density (kg/m³)</th>
<th>Mass flow (g/min)</th>
<th>Particle diameter (mm)</th>
<th>Yield (%)</th>
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</table>

Conventional Soxhlet extraction with n-hexane 11.4
3.1 Effect of pressure

The effect of pressure on the extraction of oil from grape seeds was investigated with sc-CO$_2$ at pressures of 20 MPa and 30 MPa. The temperature, mass flow and particle size were kept constant at 313 K, 5 g/min and 0.40 mm respectively. The results are presented in Table 1. As expected, it was observed that extraction yield strongly increased with pressure (6.6 % and 10.5 % at 20 MPa and 30 MPa, respectively), according to the basic principles of supercritical fluid extraction [5] and similar to the other studies on oil extraction from grape seeds [1,2,4,6]. These studies showed that, in general, at a constant temperature, the solubility of a substance in an SCF increases with pressure for increasing density (See data in Table 1).

3.2 Effect of temperature

Another set of experiments were conducted in order to study the effect of temperature on the SFE of grape seed oil. All other parameters were fixed (30 MPa, 5 g/min and 0.40 mm) while the temperature was increased from 313 K to 333 K. The influence of temperature in the cumulative content of oil extracted is shown in Table 1. The highest extract yield extraction was observed at the lowest temperature (10.5 % and 8.7 % at 313 K and 333 K, respectively), which was due to the high solvent power of sc-CO$_2$ due to its higher density at the lower temperature [7].

3.3 Effect of flow rate

The solvent ratio is the most important parameter for gas extraction, once approximate values of pressure and temperature are selected [5]. Table 1 shows the oil extractions yields resulted from the use of three CO$_2$ flow rates that ranged from 1 g/min to 5 g/min, operating at 30 MPa, 313 K and a particle size of 0.40 mm. The higher was the flow rate, the higher the oil extraction yield (9.8 % and 10.5 % at 1 g/min and 5 g/min, respectively). According to Duba and Fiori [2], this behaviour is in line with an increase in both the external and internal mass transfer parameters. These same authors claimed that conversely, the specific consumption of the solvent increased at increasing sc-CO$_2$ flow rate and ultimately, for commercial applications the solvent flow rate should be optimized in terms of extraction time and solvent volume used per operation [2].

3.4 Effect of particle size

Extractions were performed to check out the effect of particle size on the oil recoveries as well. Oil yield increased with decreasing particle size working at 30 MPa, 313 K and 5 g/min (See Table 1). Mass transfer in sc-CO$_2$ extraction from solid substrates in most cases depends heavily on the transport rate in the solid phase. In general, the oil extraction yield increased with decreasing particle size
(10.5 % and 9.4 % at 0.40 mm and 0.80 mm, respectively). The length of the transport path determines mass transport in the solid phase. Large particles hinder the penetration of the supercritical solvent and the solubilisation of the solute [5,7].

4. Conclusions

The effect of the main process parameters during supercritical fluid extraction of grape seed using carbon dioxide was corroborated. The highest oil extraction yield was 10 % in mass fraction and the oil extraction yield increased with increasing pressure and decreasing temperature, due to their effect upon the oil solubility essentially. On the other hand, the smaller was the particles, the higher the final yield. Although the oil extraction yield increased with the increased solvent flow, it should be noted that specific CO$_2$ consumption also increased. This technique can be used as an alternative to conventional organic solvent extraction methods.

References


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