C. R. Chimie 17 (2014) 284–292

Contents lists available at ScienceDirect

Comptes Rendus Chimie

www.sciencedirect.com

Full paper/Mémoire

Grape seed oil extraction: Interest of supercritical fluid extraction and gas-assisted mechanical extraction for enhancing polyphenol co-extraction in oil

Natacha Rombaut [a,b], Raphaëlle Savoire [a,*], Brigitte Thomasset [b], Typhanie Bélliard [a], Jérémie Castello [a], Élisabeth Van Hecke [a], Jean-Louis Lanoisellé [c]

[a] UTC/ESCOM, EA4297 TIMR, rue Personne-de-Roberval, BP 20529, 60205 Compiègne, France
[b] UTC, CNRS–FRE 3580 GEC, rue Personne-de-Roberval, BP 20529, 60205 Compiègne, France
[c] Université Bretagne-Sud, EA 4250, LIMATB, 56300 Pontivy, France

ARTICLE INFO

Article history:
Received 10 July 2013
Accepted after revision 28 November 2013
Available online 13 February 2014

Keywords:
Lipids
Natural products
Green chemistry
Supercritical fluids
Gas-Assisted Mechanical Expression (GAME)
Grape seed

ABSTRACT

The aim of this study is to compare three oil extraction methods and to evaluate their efficiency for producing an oil rich in polyphenols. The three extraction methods are screw pressing, extraction by supercritical CO_2 percolation and the combination of these two processes (Gas-Assisted Mechanical Expression: GAME). Screw pressing is the most efficient process for producing grape seed oil with a high yield, but supercritical CO_2 process permits an increase of polyphenol co-extraction with oil. The GAME process allows extraction of more polyphenols than screw pressing and constitutes an interesting process considering oil yield.

Mots clés :
Lipides
Produits naturels
Chimie verte
Fluides supercritiques
Pressage mécanique assisté par fluides supercritiques
Pépins de raisin

RÉSUMÉ

L'objectif de cette étude est de comparer trois procédés d'extraction et d'évaluer leur efficacité pour produire une huile riche en polyphénols. Les trois procédés étudiés sont le pressage à vis, l'extraction par percolation au CO_2 supercritique et la combinaison du pressage et de cette extraction : le pressage mécanique assisté par fluide supercritique (PAFSC). Le pressage à vis est le procédé le plus efficace en termes de rendement en huile, alors que l'extraction par CO_2 supercritique permet une augmentation de la co-extraction de polyphénols dans l'huile. Le PAFSC conduit à une augmentation de la co-extraction des polyphénols en comparaison du pressage à vis, et constitue ainsi un procédé alternatif intéressant pour la production d'huile.

1. Introduction

Grape (*Vitis vinifera* L.) is one of the major crop produced worldwide (66 million tons in 2009 [1]). Its utilizations include fruit consumption, pharmaceutics and wine making (from 70 to 80%). Residue of wine making is named grape pomace and accounts for 20% of grape (w/w). It is composed of seeds, 38 to 52% on a dry matter basis, but also of stems, pulps and skins [2]. The presence of oil and the high phenolic content of grape seeds offers alternative valorization pathways for these by-products [3].

* Corresponding author.
E-mail address: raphaelle.savoire@enscbp.fr (R. Savoire).

1631-0748/$ – see front matter
http://dx.doi.org/10.1016/j.crci.2013.11.014

Grape seed contains from 8 to 20% of oil (dry basis), which is mainly composed of unsaturated fatty acids (linoleic and oleic fatty acids: 58 to 78% and 10 to 20%, respectively [4,5]). Grape seed oil has an unusual high smoking point (190–230 °C, according to Morin [6]) due to the presence of saturated fatty acids (10%). Additionally, this oil is reported to contain minor bioactive components such as phenolic compounds (between 59 to 360 mg Gallic Acid Equivalent [GAE]/kg) [7,2]. Polyphenols identified in grape seed are catechin, epicatechin, trans-resveratrol and procyanidin B1 [2,8]. These phenolic compounds are reported to be involved in a wide range of biological activities [9], but are mostly known for their antioxidant properties. Given the unsaturation level of grape seed oils, those compounds are beneficial for oil conservation [9]. The grape seed oil extraction method does not affect the fatty acid profile considering solvent extraction (hexane or petroleum ether) and Supercritical Fluid Extraction (SFE) [10]. The quality of grape seed oil (free fatty acid, iodine index, saponification index, unsaponifiable fraction, peroxide index, and fatty acid composition) extracted by SFE is similar to that of oil extracted by organic solvent and then refined according to Molero Gómez et al. [11]. On the other hand, total phytosterol extraction was higher with SFE than with petroleum ether [12].

At an industrial scale, oil contained in oilseeds is commonly extracted by screw pressing, which is often followed by organic solvent extraction steps to enhance the global oil yield. However, in the case of seeds with a low oil content such as grape seeds, solvent extraction is preferably used to maximize oil extraction yield [13]. Although high oil yields are achieved by this process, use of organic solvents has several limitations, among which:

- the environmental toxicity;
- the fluctuating price of solvent accordingly to petroleum;
- the non-selective solubility towards lipophilic compounds [14].

Apart from mechanical (screw pressing or hydraulic pressing) and solvent processes, alternative oil extraction technologies focus mainly on water processes (enzymatic) and supercritical fluid extraction. The latter has been extensively studied using supercritical CO_2 on different raw materials (e.g., linseed [15], rapeseed [16], grape seeds [11,17,18]). The oil yield can be maximized according to processing parameters, among which CO_2 pressure and temperature. However, to reach high oil yields, intensive pressure has to be applied due to increase of the solvent power of CO_2, inducing an increase of operating costs [19]. Within an objective of reduction of cost and energy, an alternative expression process was developed: Gas-Assisted Mechanical Expression (GAME) [20,21]. The principle of GAME relies notably on a partial displacement of oil by CO_2 during the pressing, resulting in an increase of oil yield [22]. Studies of this batch process include at first a step of seed conditioning in supercritical CO_2 followed by oil expression under uniaxial compression. The use of a continuous flux of CO_2 during expression would then be another step towards an industrialization of this process.

In this work, the GAME process under a continuous flux of supercritical CO_2 is evaluated. The objective of this study is to compare oil yield and total polyphenol content of oils extracted by GAME, supercritical fluid extraction without pressing (SFE) and screw pressing as a reference process.

2. Experimental procedures

2.1. Grape seed processing

2.1.1. Raw material

Grape seeds were provided by the Distillerie Jean-Goyard (Aÿ, France). The duration between grape pressing and air drying of grape seeds was 15 days. Their moisture content was lowered to about 7% (db) by air drying at the Distillerie. Oil and water content of the seeds were determined according to French standard procedures [23,24], respectively. The studied grape seed are composed of 12.2 ± 0.5% of oil (db: dry basis) and 6.78 ± 0.03% of water (db).

The material was kept in a closed bag, at room temperature, until processing. For SFE and GAME experiments, grape seeds were grounded using a knife mill (Urshel, USA). The particle size (70%) was comprised between 600 and 1180 μm.

2.1.2. SFE and GAME experiments

2.1.2.1. Set up. Oil extraction by supercritical CO_2 was performed using a device designed by Separex (France). The extractor vessel has a capacity of 2 L, with maximal working pressure, temperature and flow rate of 70 MP, 150 °C and 25 kg CO_2/h, respectively. The experimental setup is presented in Fig. 1. For GAME experiments, a water circuit ensures the mechanical compression of the seed bed, water being used as hydraulic fluid. CO_2 (purity 99,5%) was purchased from Air Liquide, France.

2.1.2.2. SFE experimental procedure. The extractor vessel was first pre-heated at the desired temperature (for 45 to 60 min). Grounded grape seeds (200.0 ± 0.1 g) were introduced into the extractor vessel, above a PET filter (0,45 μm, Sefar-Fyltis, France) to prevent the seed particles to clog the CO_2 circulation lines. The recirculation valve and all exit valves were then closed and the extractor was pressurized at the desired pressure using the CO_2 pump. The CO_2 pressure was manually maintained in the extractor by a back-pressure valve (BRP 1, Fig. 1) located between the extractor and the cyclonic separators. When the desired pressure was reached, the recirculation valve was opened and extraction was maintained for 120 min, under a continuous flux of CO_2. Pressure in the separators was not controlled during the experiments and was comprised between 4.5 and 5.6 MPa. Temperatures were maintained at 60 ± 1 °C and 35 ± 1 °C, respectively for separators 1 and 2. The extracts were collected during the experiments from the two separators.

2.1.2.3. GAME experimental procedure. The pressing chamber was pre-heated between 45 and 60 min before the experiment. Grounded seeds (200.0 ± 0.1 g) were inserted