Comparison of Manufacturing Cost of Thyme Extract Obtained by Supercritical Fluid Extraction and Steam Distillation

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Thyme is an aromatic herb that has many species and is used extensively to add a distinctive aroma and flavor to food. Its essential oil has many biological properties: antiseptic, carminative, antimicrobial and antioxidant, among others. The main active compound of thyme volatile oil is thymol. The objective of this study was to estimate the cost of manufacturing (COM) of two green processes (SFE: supercritical fluid extraction, and SD: steam distillation) to obtain thyme extract. The cost estimation was done using a class 5 methodology that defines COM as a weighed sum of five factors: fixed cost of investment, cost of operational labor, cost of raw material, cost of waste treatment and cost of utilities. The operational data used was obtained from literature. The scale-up procedure assumed that both the yield and the extraction time of the industrial process are equal to the laboratorial scale if the ratio between the solvent mass and the mass of particles inside the extractor is kept constant. Three operational conditions for SFE extraction were evaluated: 8 MPa/300 K, 15 MPa/313 K and 20 MPa/313 K and three conditions for SD were studied: 373 K, 448 K and 523 K. The COMs were estimated for US$ 8,016/ton as raw material cost, after several sources of raw material were contacted for pricing. For SD and SFE extracts, the COMs varied from US$ 210 to 1030/kg for SD volatile oil and from U$S 190 to 2,200/kg for SFE extracts. The price of the commercial essential oil sold in market depends on the specie of the thyme, and can range from US$ 150 to 280/kg. Thus, depending on the thyme specie and extraction conditions, the COMs estimated for SD and SFE extracts can compete with commercial product and between them.

Key-words: Cost of manufacturing, Steam distillation, Supercritical fluid extraction, Thyme

INTRODUCTION

It is well known that South American countries have a huge biodiversity. Besides the richness of its flora, Brazil has a high potential for the production of great amounts of raw material at low cost due to its dimension and tradition in agricultural production [1]. Thus, the production of plant extracts, which is a way of adding value to the raw material, should be enhanced.

Adding value to the raw material using an ecologically correct technology would be ideal to increase income without degrading the environment. Two technologies that fulfill those requirements are steam distillation (SD) and supercritical fluid extraction (SFE). Both have the green label, since there is no use of toxic solvents. The highest advantages presented for SD are the low cost of investment and low development necessity, since it is a widespread technology. On the other hand, it is a high energy consuming process, for it uses high processing temperatures, which can induce the degradation of thermo sensitive compounds of plant extracts.

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As for SFE, it solves the temperature problem presented by SD and presents low operational cost, but the cost of investment has been pointed out as the main drawback for its spread. In spite of the technical advantages of using SFE, there is still no industrial unit operating with it in South America. The common sense says it is due to high investment costs related to a SFE plant [2]. However, the cost of SFE units has been decreasing in the last years in spite of significant technical improvements because of the competition between suppliers [3]. Moreover, recent studies have shown that supercritical extraction may be economically viable to obtain vegetable extracts [2, 4-7]. Even though each technology presents advantages and drawbacks, the choice of a suitable extraction process depends on the intended used of the plant extract, since chemical compositions can vary a lot with extraction technique [8].

Thyme is an aromatic herb extensively used to add a distinctive aroma and flavor to food. It is rich in volatile oil, which is used in perfumes, soaps and toothpastes formulations. Several biological properties are attributed to the volatile oil; it presents fungicidal, antiseptic, and antioxidant activities, and is an excellent tonic, besides carminative, antispasmodic and expectorant properties [9]. In folk medicine the main applications of thyme have been in the treatment of digestive complaints and respiratory problems, and in the prevention and treatment of infection [10]. The major phenolic components in thyme extracts, especially thymol and carvacrol, present higher antioxidant activity than the well-known BHT and α-tocopherol antioxidants [11]. Still, the volatile oils from *Thymus* genus, especially *Thymus hyemalis*, *T. zygis* and *T. vulgaris*, are potent bactericide agents that can be used in food industry, increasing shelf life and improving food products preservation [12].

The genus *Thymus* includes several species, so the yield and the chemical composition of its essential oil are variable [13]. In each specie there are different chemotypes, which are classified by their main volatile components, such as 1,8-cineole, linalool, α-terpineol, geraniol, *trans*-thujane/terpinen-4-ol, thymol, linalool/thymol, carvacrol, carvacrol/thymol, etc. [8, 12, 14].

The traditional extraction method for obtaining thyme oil is SD of the aerial parts of the plant [15]. One of the quality parameters of thyme extract is the content of thymol, which can suffer thermal degradation during SD [13]. Because of this, SFE is proposed as an alternative extraction method for obtaining thyme extracts [8, 10, 15, 16]. By this process, besides the volatile oil, heavier compounds are co-extracted, leading to a product having different chemical composition and sensorial attributes from SD volatile oil [8, 15, 16]. This is far from being a disadvantage, since SFE extracts faithfully represent the aroma of the natural plant [8], so they are preferred in sensorial evaluation [16], which is particularly important when the extracts are destined for food and perfume industries.

The objective of the present work was to carry out an economical evaluation of both extraction methods: steam distillation and supercritical extraction. Experimental literature data were used to estimate the manufacturing costs of thyme extracts.

MATERIALS AND METHODS

Operational data for cost of manufacturing (COM) estimations were collected from literature. Three operational conditions for SFE extraction were evaluated: 8 MPa/300 K, 15 MPa/313 K and 20 MPa/313 K [15]. As for thyme SD, three temperatures were studied (373, 448 and 523 K), at two different steam flow rates (1.6 kg/h and 2.5 kg/h) [13]. Although Rouatbi et al. [13] do not state the thyme specie used for SD, by the chemical composition presented, it can be concluded it belongs to thymol chemotype. The plant used in SFE by Moldão-Martins et al. [15] (*Thymus zygis*) also belong to thymol chemotype [14].

The COM of thyme extract (obtained by SFE) and volatile oil (obtained by SD) was estimated using the methodology described by Rosa and Meireles [1], which is based in the expression proposed by Turton et al. [17], that defines COM as a weighed sum of five main costs: fixed cost of investment (FCI), cost of operational labor (COL), cost of utilities (CUT), cost of waste treatment (CWT) and cost of raw material (CRM). The expression proposed by Turton et al. [17] is given by Equation (1).

$$COM = 0.304\,FCI + 2.73\,COL + 1.23\,(CRM + CUT + CWT) \quad (1)$$

For SFE fixed cost of investment (FCI), it was considered an industrial scale unit equipped with a CO_2 recycling system. It is composed by two 0.4 m^3 columns, one flash tank separator, one CO_2 reservoir, one CO_2 condenser, one pump and one heat exchanger, and costs around US$ 2,000,000.00. For SD FCI, it was considered an industrial unit composed by two 0.5 m^3 distillation columns, one shell and tube condenser and one separator, which costs US$ 50,000.00. Annual depreciation was considered to be 10% for both units.

The operational labor cost (COL) was calculated using information of man-hour per operation-hour according to the tables presented by Ulrich (1984), cited by Turton et al. [17]. Two and three operators per shift are needed to work in the SFE and SD units, respectively, at a cost of US$ 3.00/hour. The total operational time of both extraction units was considered as 7920 h per year, which corresponds to 330 days per year of continuous 24 h per day shift.

The raw material cost (CRM) includes the cost of the solid substrate and the cost of the solvent lost during the process. The CO_2 (US$ 100.00/ton) loss was assumed to be 2%, and it is mainly due to extractor depressurization at the end of each extraction cycle [3]. The pre-processing cost (drying and milling) was estimated by Rosa and Meireles [1] using the SuperPro Designs Software v4.7 as US$ 30.00/ton. After several sources of raw material were contacted for pricing, the cost of thyme was determined as US$ 8,016.00/ton.

The utilities cost (CUT) for SFE was estimated considering the energy involved in the CO_2 cycle, using the temperature-entropy diagram. The utilities used were steam (US$ 0.01333/Mcal), cold water (US$ 0.0837/Mcal) and electricity (US$ 0.0703/Mcal). For SD, the steam (US$ 16.22/ton) and cold water (US$ 14.80/ton) costs were based on the values proposed by Turton et al. [17].

The cost of waste treatment (CWT) was neglected, since the only accumulated waste in SFE and SD are the exhausted solid and the wet solid, respectively, which may both be sold to make fertilizer or incorporated to the soil, considering they are harmless residues. The CO_2 that is lost during the process in SFE does not need any treatment, since in small quantities it is not harmful.

The mass of feed in the industrial extraction vessel was determined according to apparent bed density. For SD and SFE the scale-up procedure assumed that both yield and extraction time of the industrial process will be the same as the ones obtained in laboratorial scale if the ratio between the solvent mass and the mass of particles inside the extractor is kept constant [1].

RESULTS

Figure 1 presents the COM estimated to produce thyme extracts by SFE and SD. The COM ranged from U$S 190.00/kg to US$ 2,200.00/kg for SFE extracts and from US$ 210.00/kg to US$ 1,030.00/kg for SD volatile oil. For SFE, the extraction condition that presented the lowest COM (US$ 190.00/kg) was 20 MPa/313 K for an extraction time of 200 min. For SD, the operation temperature of 523 K presented the lowest COM (US$ 210.00/kg) for 20 min of processing using steam at flow rate of 2.5 kg/h. Although the COMs estimated for both processes were in the same order of magnitude, the composition of the cost (FCI, CUT, COL, CRM and CWT) was different.

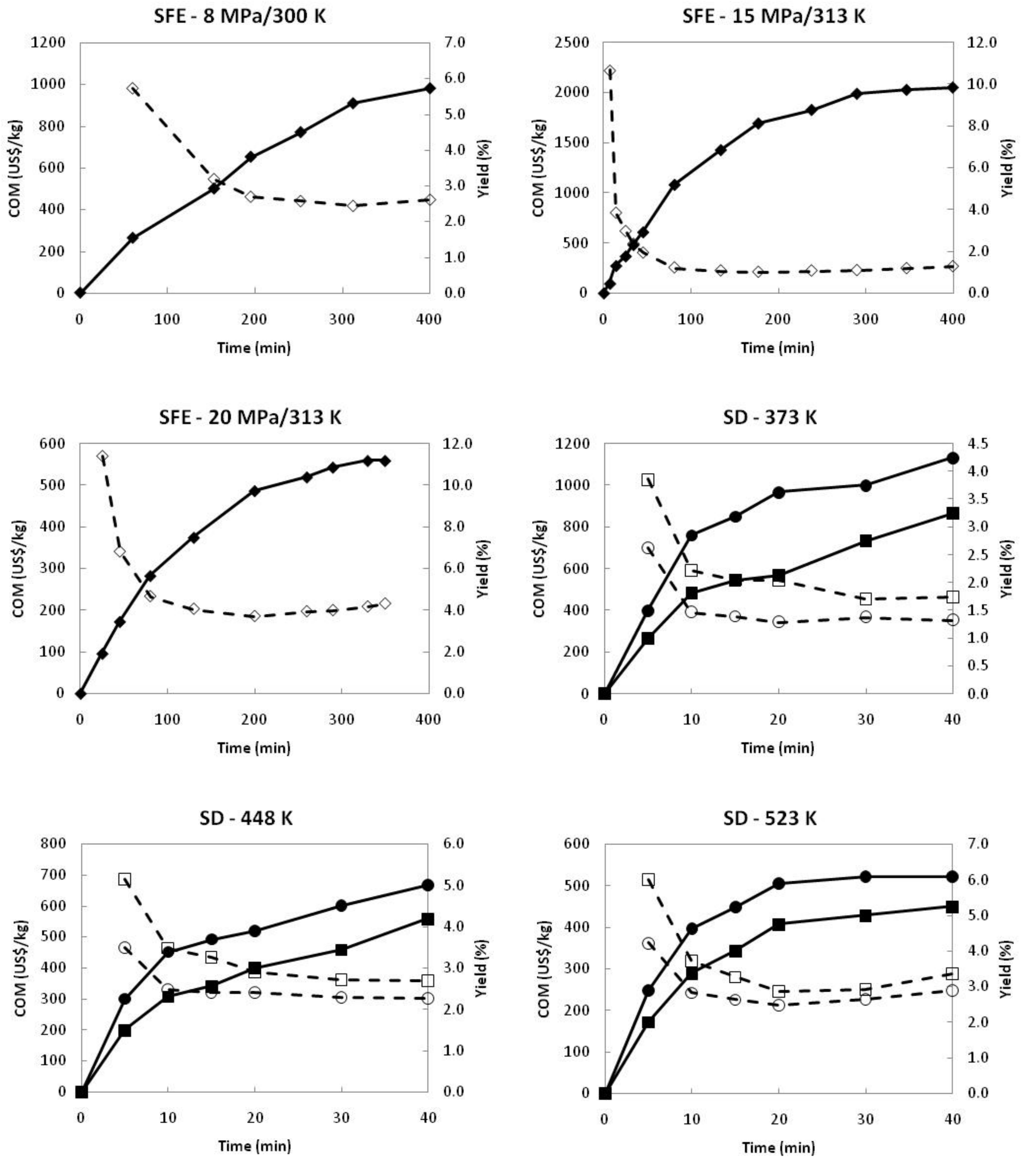

Figure 1: COM estimations for thyme SFE (♦ = yield [15], ◊ = COM) and SD (■ = yield at steam flow rate of 1.6 kg/h [13], □ = COM at 1.6 kg/h, ● = yield at steam flow rate of 2.5 kg/h [13], ○ = COM at 2.5 kg/h).

In SFE process, it can be seen COM strongly decreases with processing time up to 150 min. After that point, COM is approximately constant, and can present a slight increase for longer cycles (over 250 min). This behavior is due to number of cycles that can be conducted in a year for different processing times. For short cycles, although there is a high number of them over the year, too much raw material is used with a low yield being reached, since the extraction bed is not exhausted, which leads to more expensive products. As for long cycles, although the raw material consumption is lower because of low number of cycles over the year, there is no much increase in yield after 250 min, leading to a decrease in total extract that could be recovered throughout the year with shorter cycles operating, which increases the final COM. The economical evaluation allows finding out where is the equilibrium point between processing cost and yield.

Thyme volatile oil yield increases, and, therefore, COM decreases, with both temperature and steam flow rate. However, it was observed quality loss in the product obtained at 523 K due to thermal degradation of thymol, leading Rouatbi et al. [13] to conclude that SD at 448 K is a better choice for processing thyme. At his temperature, the lowest COM (US$ 300.00/kg) was reached at 40 min of distillation with steam flowing at 2.5 kg/h. The same behavior observed in SFE of COM *vs.* time is observed for SD: high COMs for short processing times, stabilization of COM and possible slight increase in COM for long processing times.

This kind of behavior presented both by SFE and SD processes, suggests that the raw material cost is an important factor in COM. Plants from which essential oils containing biological properties can be extracted, usually have high prices, leading to expensive products regardless the extractive technique, because the investment cost ends up being diluted by the cost of raw material [6, 7]. For the data studied, CRM was responsible for 39% (8 MPa/300 K and 15 MPa/313 K, 400 min) to 97% (15 MPa/313 K, 7 min) and 66% (40 min) to 96% (5 min) of COM in SFE and SD processes, respectively.

As for FCI and CUT, they have opposite importance in SFE and SD, as expected. In SFE, while FCI ranges between 2% (15 MPa/313 K, 7 min) and 50% (8 MPa/300 K, 400 min), CUT represents no over 2.5% of COM. As for SD, CUT is the most important cost after CRM, with 4% (5 min) to 32% (40 min) of COM, while FCI represents at maximum 0.1% of COM.

CWT was neglected for both technologies, since there is no toxic residue generation in either one of them. As for COL, it stayed below 10% of COM for SFE and below 1% of COM for SD.

The price of the commercial essential oil sold in market depends on the specie and chemotype of the thyme, and can range between US$ 150.00/kg and US$ 280.00/kg for SD products [18]. The COMs estimated in the present work for both SFE and SD are in the range of the commercial prices. In a scale ranging from 1 to 5, where 1 is the most detailed study, the COM studied in the present work is labeled as 5 to 4 [19], which means it is a preliminary study on feasibility. Thus, the COMs here estimated are the highest possible values. With further development of the industrial project, using more precise technical information, the COM tends to decrease. Thus, depending on the extraction conditions selected, the COMs estimated for SD and SFE extracts can compete with commercial product and between them.

CONCLUSION

The COM ranged from U$S 190.00/kg to US$ 2,200.00/kg for SFE extracts and from US$ 210.00/kg to US$ 1,030.00/kg for SD volatile oil. For SFE, the extraction condition that presented the lowest COM was 20 MPa/313 K, 200 min. For SD, the operation temperature of 523 K presented the lowest COM for 20 min of processing at steam flow rate of 2.5 kg/h, but thymol degradation at this temperature suggests that 448 K would be a better choice, with a minimum COM of US$ 300.00/kg (40 min). CRM presented the highest share in COM composition (39-97% for SFE and 66-96% for SD). The second highest share was represented by FCI in SFE (2-50%) and CUT in SD (4-32%). The COMs estimated for both extractive techniques are in the same order of magnitude, and in the range of commercial SD price. Therefore, thyme extracts obtained by SFE and SD can compete with commercial product and between them. The choice of the process depends on the desired chemical composition of the extract.

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