

# Extraction of Lovage (*Levisticum officinale* Koch.) Roots by Carbon Dioxide. 1. Effect of CO<sub>2</sub> Parameters on the Yield of the Extract

Egidijus Daukšas and Petras Rimantas Venskutonis\*

Department of Food Technology, Kaunas University of Technology,  
Radvilenu pl. 19, Kaunas 3028, Lithuania

Björn Sivik

Food Technology, Chemical Center, University of Lund, P.O. Box 124, S-221 00 Lund, Sweden

This paper describes the experiments on sub- and supercritical CO<sub>2</sub> extraction of lovage (*Levisticum officinale*) root to determine the effect of CO<sub>2</sub> parameters on the yield of the extract. Two series of extractions were carried out: first, to establish the effect of CO<sub>2</sub> pressure and temperature; second, to determine the dynamics of the extraction at constant temperature. With pressure varied from 80 to 300 bar and temperature varied from +10 to +55 °C, it was found that the best result was at 50 °C and 200 bar. During the second series the temperature was kept at 50 °C and the extracts were collected at various stages of extraction process at the pressures from 80 to 350 bar. It was found that the yield of the extract increases when the pressure is raised from 80 to 200 bar; however, further increase of the pressure to 350 bar was not efficient.

**Keywords:** *Lovage root; Levisticum officinale Koch.; supercritical CO<sub>2</sub> extraction; extract yield*

## INTRODUCTION

Lovage (*Levisticum officinale* Koch.) is a tall perennial Umbelliferae family plant cultivated in numerous European countries. All botanical parts of the plant (leaves, stems, roots, and seeds) exhibit a strong flavor, which generally characterizes celery. In particular, the root extract possesses a warm-spicy note, whereas the seed and leaf oil impart a more diffusive and penetrating tendency (Toulemonde and Noleau, 1988).

Flavor constituents are isolated from the aromatic plants according to different methods, mainly by distillation and extraction. The essential oil recoveries from different botanical parts of lovage may vary from 0.8 to 1.1% in the seeds, from 0.6 to 1.0% in the roots, and from 0.05 to 0.15% in the leaf (Toulemonde and Noleau, 1988). In the other source, particularly dealing with lovage grown in Lithuania, the yield of the essential oil was 1.14% in the root stocks, 1.01% in the roots, 0.68% in the leaves, 1.31% in the raceme, and 1.8% in the seeds (Jaskonis, 1989). However, in the case of lovage root, the yield of the steam distillation process is very low and does not reproduce the characteristic flavor.

The extraction of flavors and fragrances with compressed CO<sub>2</sub> was comprehensively reviewed and described by Moyler in 1993. The information about lovage extraction by CO<sub>2</sub> in sub- and/or supercritical state was not found in the surveyed literature. However, this method has been used with some other Umbelliferae family plants, for example, coriander and angelica (Kerrola and Kallio, 1993, 1994), caraway (Sovová et al., 1994; Kallio et al., 1994), anise (Ondarza and Sanchez, 1990), and celery and coriander (Catchpole

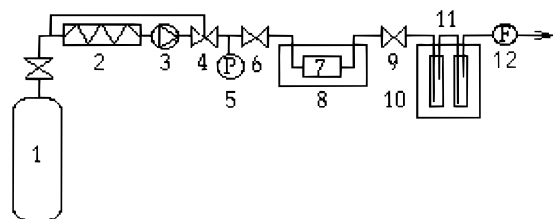
et al., 1996). The yields of steam-distilled oil and extract by liquid CO<sub>2</sub> from celery (*Apium graveolens*) seeds were almost equal and constituted 2.5–3.5 and 3%, respectively, whereas yields from parsley (*Petroselinum crispum*) seeds were 2.0–3.5% oil during 5 h of steam distillation and 9.8% extract obtained by CO<sub>2</sub> during 2 h of extraction at 58 bar and 20 °C (Moyler, 1994).

Supercritical fluid extraction (SFE) is an effective process in some food industries as it readily produces solvent-free extracts. Also, CO<sub>2</sub> is a nontoxic solvent. On the other hand, because of the high price of the equipment, it can be applied only when high added value products are expected (Caragay and Little, 1981).

The basic process, that is, single-stage high-pressure extraction followed by separation at subcritical pressure, is often not sufficient to obtain the desired products. Most of the natural materials contain many lipophilic compounds that are likely to be extracted along with the target products, even when optimal extraction is used. These coextracts reduce the quality of the end products and in some cases limit their industrial use (Pellerin, 1988). Moreover, to obtain the essential oils by supercritical CO<sub>2</sub> extraction, it is necessary to operate at maximum selectivity to avoid coextraction of undesired compounds. In addition, cuticular waxes have to be eliminated from the extraction (Boelens and Boelens, 1997).

The power of solubilization increases with the density of the fluid; high densities of a supercritical fluid are possible at high pressures and allow it to dissolve large quantities of organic compounds (Poiana et al., 1998). In 1997 Manninen et al. extracted the dried press residue of cloudberry by varying CO<sub>2</sub> pressures from 90 to 300 bar and temperatures from 40 to 60 °C. The extracts were either solids or viscous oils depending on the amount of neutral lipids, which increased with

\* Author to whom correspondence should be addressed (telephone +370-7-756426; fax +370-7-756647; e-mail rimas.venskutonis@ctf.ktu.lt).



**Figure 1.** Schematic drawing of the supercritical extraction equipment: 1, gas tube; 2, ethanol bath,  $-8\text{ }^{\circ}\text{C}$ ; 3, pump; 4, relief valve; 5, pressure meter; 6, shut-off valve; 7, extractor; 8, water bath; 9, micrometering valve; 10, dry ice-ethanol bath; 11, glass tubes; 12, flow meter.

increasing pressure. Subcritical liquid  $\text{CO}_2$  has been used as a solvent to selectively extract essential oils from ground botanicals at temperatures between 0 and  $10\text{ }^{\circ}\text{C}$  and pressures from 50 to 80 bar. However, the yields usually are higher when supercritical  $\text{CO}_2$  is used, as was recently shown in the case of basil (Lachowich et al., 1997). Supercritical fluid  $\text{CO}_2$  is used commercially mostly to extract hops (Gardner, 1993) and coffee or tea (Lack and Seidlitz, 1993) at temperatures from 40 to  $80\text{ }^{\circ}\text{C}$  and pressures from 200 to 350 bar. Supercritical fluid  $\text{CO}_2$  has also been used to extract and fractionate gluten lipids and glycerides (King and Bott, 1993) and to postprocess a solid tuberoses extract produced by liquid solvent extraction (Reverchon and Della Porta, 1997).

$\text{CO}_2$  extracts only part of the water from relatively dry plant material. Any water extracted can be physically separated from the product as it leaves the system (Moyler and Heath, 1988).

In the present study lovage roots were extracted by  $\text{CO}_2$  under varying extraction parameters in a wide range of pressures and temperatures to determine the yield of the extract.

## MATERIALS AND METHODS

**Materials.** The roots of lovage (*Levisticum officinale* Koch.) were collected manually from the experimental garden of the Lithuanian Institute of Horticulture in 1995. The roots were washed, dried at  $30\text{ }^{\circ}\text{C}$  in a ventilated drying oven, and stored in paper bags at ambient temperature protected from direct light until further analysis. The moisture content in the dried roots was 4–8%, and the yield of hydrodistilled essential oil was only 0.054 mL/100 g. The roots were ground to pass a mesh (60 in. $^{-1}$ ) sieve before extraction.

Carbon dioxide, 99.99% purity, was obtained from Alfax, Sweden. Cyclohexane, extra purity (>99%), and anhydrous sodium sulfate (99%) were obtained from Merck (Darmstadt, Germany).

**Extraction Parameters.** Two series of experiments were carried out:

(1) *Effect of Temperature and Pressure at Constant Amount of  $\text{CO}_2$  (50 L).* Temperatures were 10, 20, 30, 35, 40, 45, 50, and  $55\text{ }^{\circ}\text{C}$ ; pressures were 80, 90, 120, 150, 200, 250, 300, and 350 bar.

(2) *Effect of  $\text{CO}_2$  Pressure and Solvent Amount at Constant Temperature ( $50\text{ }^{\circ}\text{C}$ ).* Pressures used were the same as in the first series of experiments; the yield of the extract was measured in the first test tube (Figure 1, 11) after 5, 10, 20, 30, 40, 50, 60, 70, and 80 L of  $\text{CO}_2$  had been passed. Finally, after 80 L of solvent had been passed, the amount of the extract was also measured in the second test tube (Figure 1, 11), and this amount was added to the yield obtained in the first test tube.

**Apparatus.** Two sets of extraction apparatus from different companies were used for the experiments. A schematic diagram of the experimental apparatus used in this study is

shown in Figure 1. A LEWA pump (LEWA Herbert Ott GmbH and Co.) was used in the first series of extractions. SFE was conducted under the following experimental conditions: capacity of extraction vessel, 7 mL; amount of extracted sample, 2.5 g;  $\text{CO}_2$  flow, 0.25 L/min at atmospheric pressure.

A Dosapro Milton Roy pump (Milroyal B-C) was used in the second series of extractions. The capacity of extraction vessel was 47 mL, the amount of extracted root material was 10 g, and the  $\text{CO}_2$  flow was 0.25 L/min at atmospheric pressure. Two grams of  $\text{Na}_2\text{SO}_4$  was added into the vessel on the top of the sample for the adsorption of water. The samples were taken after 5, 10, 20, 30, 40, 50, 60, and 80 L of  $\text{CO}_2$  had passed through the sample.

The extracts were collected in the two test tubes (5 mL) connected in series, which were immersed in a dry ice-ethanol bath at  $-82\text{ }^{\circ}\text{C}$ . The yield of the total extract was weighed by the precision balances (Mettler AE 163, readability 0.01 mg; Mettler Instruments AG, Switzerland).

All samples were stored in a freezer before testing. So far as the yields of the extracts were  $<0.2\text{ g}$ , it was difficult to separate the extract from the water. Therefore, the amount of the dry extract was measured chromatographically. For that purpose, 100  $\mu\text{L}$  (0.078 g) of cyclohexane was added to the test tubes, which were shaken for  $\sim 0.5$  min. The separated top layer of the solvent was used for the GC. The amount of the dry extract was calculated from the ratio of the GC peak area of solvent and extracted constituents.

Essential oil was hydrodistilled in a Clevenger type apparatus during 3 h.

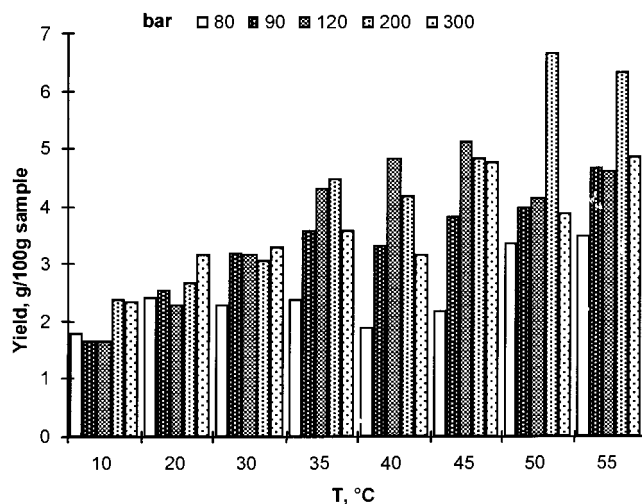
**GC Conditions.** GC analyses were carried out on a Varian 3400 capillary gas chromatograph equipped with a flame ionization detector connected to a Vista 420 integrator (Varian Associates, Walnut Creek, CA) and a fused silica capillary column, Supelcowax 10 (60 m, 0.32 mm i.d., 0.50  $\mu\text{m}$  film thickness). The oven temperature was programmed from 60 to  $230\text{ }^{\circ}\text{C}$  at  $2\text{ }^{\circ}\text{C}/\text{min}$ . The temperature of the on-column injector was raised from 40 to  $140\text{ }^{\circ}\text{C}$  at  $50\text{ }^{\circ}\text{C}/\text{min}$  and kept at  $140\text{ }^{\circ}\text{C}$  for 5 min. Helium was used as carrier gas with the flow rate of 4 mL/min.

## RESULTS AND DISCUSSION

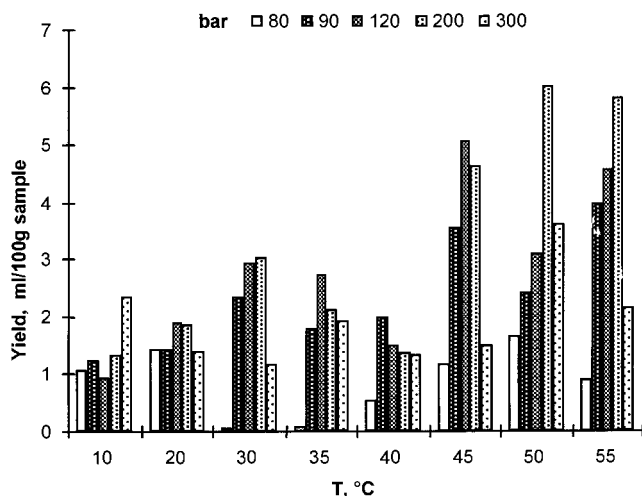
**Effect of Temperature and Pressure.** During the first series of experiments, both the pressure (from 80 to 300 bar) and the temperature (from 10 to  $55\text{ }^{\circ}\text{C}$ ) were changed, and the yield of the extract was measured after 50 L of the solvent had been passed. Total yield (dry matter plus water) was weighed and the amount of dry substances estimated by dissolving the extract in cyclohexane and analyzing by GC.

The total yields of  $\text{CO}_2$  extracts from lovage roots at different temperatures and pressures are shown in Figure 2. In general, it was found that the yield of the extract increases with increasing pressure and temperature; however, this dependence was not so simple: for instance, at  $35\text{--}50\text{ }^{\circ}\text{C}$  the increase of the pressure to 300 bar was not effective in terms of the total extract yield. The peaks of the yield at 50 and  $55\text{ }^{\circ}\text{C}$  were observed when an extraction pressure of 200 bar was used. A slight decrease in the yield was measured at  $40\text{ }^{\circ}\text{C}$ , and this is in agreement with the minimal solubility temperature for water in dense  $\text{CO}_2$ , which is lower at  $40\text{ }^{\circ}\text{C}$  (Coan and King, 1971; Francis, 1954).

Figure 3 shows the yields of the water-free extracts at different temperatures and pressures. The effect of varied parameters on the yield of dry extracts was somewhat similar to the effect on the total extract amount. Some decrease in the yield was observed at the temperature of  $35\text{--}40\text{ }^{\circ}\text{C}$ . Results on the yield of the extracts without water show that a critical temperature for the solubility of the extract in dense  $\text{CO}_2$  is  $30\text{--}35\text{ }^{\circ}\text{C}$  at low pressure (80 bar).



**Figure 2.** Total yield of the extract (including extracted water) from roots of lovage at different CO<sub>2</sub> temperatures and pressures.



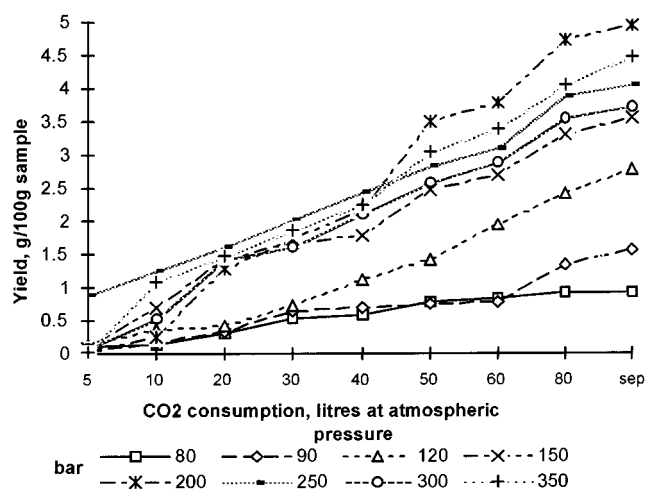
**Figure 3.** Total yield of the dried extracts from roots of lovage at different CO<sub>2</sub> temperatures and pressures.

The biggest amounts of extract were obtained at 200 bar and 50 °C (6.03 mL/100 g), 200 bar and 55 °C (5.84 mL/100 g), and 120 bar and 45 °C (5.08 mL/100 g). Low pressure was not effective: the smallest amount of extract was obtained at a pressure of 80 bar and 30–35 °C (0.08 mL/100 g).

In general, the results obtained show that the yields of the supercritical CO<sub>2</sub> extracts from lovage roots increase when the pressure is increased to 200 bar and the temperature is increased to 55 °C due to the increase of the solvent density. However, after further increase of the pressure to 300 bar, the yield of the extract was lower as compared with that at 200 bar.

It should be noted that the appearance of the extracts slightly changed with the increase of temperature and pressure. The color, which is amber yellow when lower pressures are used, becomes darker with increasing pressure. The amount of some pale impurities in the oil also increased with pressure. When the temperature was increased from 10 to 35 °C, the color of the extract became lighter, while during further increase it darkened again.

**Effect of CO<sub>2</sub> Pressure and Solvent Amount.** The second series of experiments was carried out to examine the influence of the pressure and CO<sub>2</sub> amount on the



**Figure 4.** Dynamic of lovage root extraction at 50 °C and different CO<sub>2</sub> pressures. The yield is before water separation.

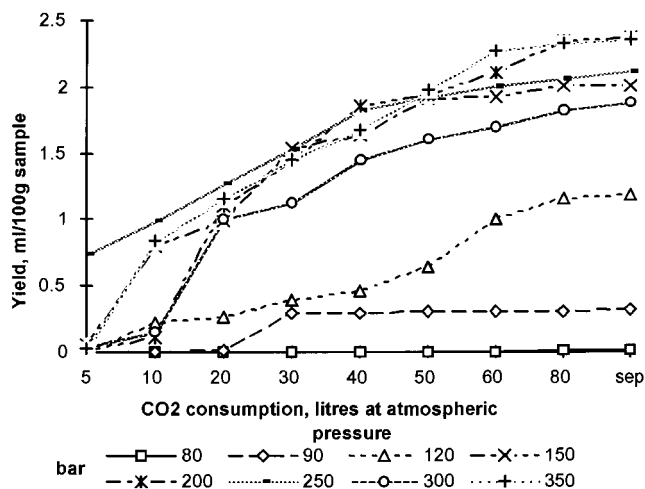
yield of the extract. A temperature of 50 °C was used for these experiments because the results with this temperature were the best in the first series of extractions. Pressure was varied in as wide a range as possible: from 80 bar (minimum for the supercritical CO<sub>2</sub>) to 350 bar (maximum for the pump).

So far as water has been extracted together with other substances, Na<sub>2</sub>SO<sub>4</sub> was used to dry the extract. The required amount of Na<sub>2</sub>SO<sub>4</sub> was calculated by taking into account the moisture content in the samples, which was <0.8 g. To bind this amount of water, <0.7 g of Na<sub>2</sub>SO<sub>4</sub> is needed. However, to ensure water absence in the extract, 2 g of absorbent was added on the top of the sample.

Figure 4 shows the dynamics of the extraction of lovage roots at 50 °C and different CO<sub>2</sub> pressures. “Sep” in the figures means the sum of the extracts in the first and second test tubes after 80 L of solvent had passed. The yield is from before water separation. It is obvious that the yield of the total extract was bigger when higher pressure was applied (80 and 90 versus 150–350 bar); however, at 150 bar, the extraction efficiency was not proportional to the further pressure increase. For example, the highest yield was obtained at 200 bar. A similar result was obtained during the first series of experiments (Figure 2). The total amount of the extracts after 80 L of CO<sub>2</sub> had passed was about 0.5–1.5 g/100 g at 80–90 bar, 2.5 g/100 g at 120 bar, and 3.5–5.0 g/100 g at 150–350 bar.

The effect of the pressure on the extraction dynamics of the dry extracts is shown in Figure 5. The amount of dry extracts after 80 L of solvent had passed was approximately 1.8–2.5 mL/100 g at pressure 150–350 bar, 1.2 mL/100 g at 120 bar, and only 0.3 mL/100 g at 90 bar. It was found that at 80 bar only a negligible amount of the extract could be obtained. It seems that the substances present in lovage roots become more soluble at higher pressure of the solvent. At the near subcritical stage CO<sub>2</sub>, the maximum of the extracted dry material was reached after only 30 L (90 bar) had passed, whereas at the increased pressure approximately 60 L was needed to take out extractable substances from lovage roots. When the curves in Figures 4 and 5 are compared, it is noted that the ratio of dry matter and water in the extracts is higher in the first portions of CO<sub>2</sub> (20–30 L); when the amount of solvent is increased, the extracts are diluted with water.





**Figure 5.** Dynamic of lovage root extraction (yield of dry extract) at 50 °C and different CO<sub>2</sub> pressures.

In supercritical condition and when water is taken out, it is observed that at 80 and 90 bar there is no change of oil yield, and from 120 to 350 bar the yield of extract increases until 50–80 L of CO<sub>2</sub> is used and then it remains constant for upper values. In contrast, when water is included, a continuous increase of yield is observed for all values of CO<sub>2</sub> for all pressures. Amounts of 40–50 and 60–80 L of CO<sub>2</sub> were the limit for all pressures because the amount of water was increasing very rapidly in the total amount of the extract.

SFE of lovage showed that the maximum extracts without water (2.37 and 2.36 mL/100 g) were obtained from 200 and 350 bar at 50 °C, although other extracts were quite similar (1.9–2.1 mL/100 g). These extracts (2.37 and 2.36 mL/100 g) are considerably greater than those obtained by hydrodistillation (0.054 mL/100 g) or other conditions of SFE at low pressure (0.01 mL/100 g at 80 bar, 0.3 mL/100 g at 90 bar, and 1.2 mL/100 g at 120 bar). SFE together with volatile compounds results in the isolation of nonvolatile substances, such as waxes, carotenoids, and chlorophyll. The behavior of extract at 80 and 90 bar at 50 °C can be explained by the low solubility of the oil at these pressures.

The solubility of water is ~0.1% in liquid CO<sub>2</sub> (Coan and King, 1971; Francis, 1954). The amounts of gaseous CO<sub>2</sub> for the extraction were 80 L or ~160 g. Consequently, this amount can contain ~0.16 g of water. The results after extraction present 0.07–0.25 g of water. It shows higher solubility of water in supercritical than in subcritical CO<sub>2</sub> and strange properties of Na<sub>2</sub>SO<sub>4</sub>. Perhaps some water was dissolved in CO<sub>2</sub> and was carried through the Na<sub>2</sub>SO<sub>4</sub> without absorption. The reason may be due to the properties of Na<sub>2</sub>SO<sub>4</sub>, which could be different in dense CO<sub>2</sub> compared with normal conditions.

A significant difference in the yield of the extracts can be noted by comparing the first and second series of experiments. For instance, during the first series the best result was 6.03 mL/100 g (200 bar and 50 °C), whereas during the second series the highest yield was only 2.37 mL/100 g at the same conditions. Some explanations for these results can be given. Two different sets of equipment were used in this work (see Materials and Methods). The extractions were made in different capacity vessels, 7 and 47 mL, respectively. Changing the vessel so that the diameter is constant but the length is increased can influence equilibrium

between the CO<sub>2</sub> and the sample. Also, different amounts of CO<sub>2</sub> were used: 50 L (measured at atmospheric pressure) for the first series of experiments (20 L of CO<sub>2</sub>/1 g of roots) and 80 L for the first second series of experiments (8 L of CO<sub>2</sub>/g of roots). The use of Na<sub>2</sub>SO<sub>4</sub> also could have some effect on the extraction process.

## CONCLUSIONS

Lovage roots were extracted by varying CO<sub>2</sub> pressure from 80 to 300 bar and its temperature from +10 to +55 °C. It was found that the highest yield of extract was obtained at 50 °C and 200 bar. The dynamics of the extraction process at 50 °C was measured at different pressures, and it was found that the yield of the extract increases when the pressure is raised from 80 to 200 bar; further increase of the pressure to 350 bar was not efficient.

It was found that by varying extraction parameters up to 6 mL of extract could be obtained from 100 g of lovage roots.

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