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Potentialities of using liquefied gases as alternative solvents to substitute hexane for the extraction of aromas from fresh and dry natural products

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ABSTRACT

The potential of liquefied gases, *n*-butane, dimethyl ether, and HFO-1234ze as effective and green alternative solvents to substitute hexane has been evaluated for the extraction of aromatic compounds from dry lavender flowers (*Lavandula angustifolia* Mill.) and fresh orange peels (*Citrus sinensis* (L.) Osbeck). The performance of these liquefied gases as solvents has been evaluated in terms of yield, olfactory perception, and composition of the extract and also in terms of energy used, green and economic impacts, and regulatory issues. First, a predictive evaluation of the solvation performance of each solvent was carried out using simulations with the conductor-like screening model for real solvent. Then solid–liquid extractions were performed using liquefied gases at a laboratory scale to determine the extraction yield, the chemical composition, and the olfactory perception of each extract. Finally, the applicability of liquefied gas extractions in an industrial process was assessed, taking into account the potential impact on process, quality, safety, regulation, and environment.

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1. Introduction

Nowadays aromas are essential ingredients that find widespread use in the food and perfume industries. The use of petroleum-based aromas in the modern food and perfume industries is a common practice, but there is a growing concern about their actual or potential effect on human health, resulting in an increasing demand of consumers for natural aromas. This concern has led to an increasing interest and use of natural products as alternative food aromas. It is therefore not surprising that the

global market value for aromas was estimated to be US \$26 billion in 2015 and is projected to reach nearly US \$30 billion in 2020, with a compound annual growth rate of 5% [1,2].

Extraction of aromas from natural products could be considered as a “clean process” in comparison with extraction processes currently used in other industries; however, recent studies have shown that their environmental impact is greater than what appeared at first [3]. As the raw extraction yields are often very low (<1%), the production of aromas requires huge amounts of plant materials and solvents, mainly hexane as the most consumed petroleum-based solvent for the production of concrete. Other solvents could be used such as toluene, pentane, or

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dichloromethane. After this first extraction, ethanol (bio-based solvent) is needed for a second step in this process, for production of the absolute from the concrete. Aromas could also be obtained in the form of essential oils by steam or hydrodistillation (HD) of fresh aromatic herbs or dry spices. Evaporation of such amounts of solvents is known to be very energy intensive, such that the extraction and evaporation step is often responsible for more than 50% of the overall process energy consumption. Moreover, the traditional extraction techniques (steam distillation and solvent extraction) generally involve high process temperatures that may greatly affect the product quality, in particular in the case of sensitive flowers that contain thermos-sensitive compounds (rose and jasmine).

To promote eco-friendly extraction processes in the industry, Chemat et al. [4] established the “six principles of green extraction of natural products”, inspired from the principles of green chemistry. By definition, green extraction “is based on the discovery and design of extraction processes which will reduce energy consumption, allows use of alternative solvents and renewable natural products, and ensure a safe and high quality extract/product”. The application of these principles already led to several success stories, with the emergence of new innovative green extraction processes based on innovative techniques and alternative solvents [5,6].

Still, more work needs to be done to find viable alternatives to conventional toxic solvents, in particular *n*-hexane, which is still commonly used for the extraction of lipophilic compounds. This constant search for alternative solvents revived the interest of scientists for liquefied gases, in particular propane, *n*-butane, dimethyl ether (DME), 1,1,1,2-tetrafluoroethane (R134a), 1,3,3,3-tetrafluoropropane (HFO-1234ze), and 2,3,3,3-tetrafluoropropane (HFO-1234yf). Indeed, these gases require only gentle pressure (<1 MPa) to remain in a liquid state and they can be very easily evaporated at low temperature. Therefore, liquefied gas extraction processes can be performed at room temperature, with low energy consumption and very few traces of residual solvent. Therefore, liquefied gas extraction preserves the quality of both the raw material and extract and reduces the number of post-treatment steps. Moreover, their chemical structures make them suitable for the extraction of lipophilic compounds in replacement of existing toxic solvents. In that sense, liquefied gas extractions offer a potential alternative technology in accordance with the principles of green extraction of natural products.

Recently, many experimental studies have been carried out to evaluate their potential. In particular, propane, *n*-butane, and DME have been extensively investigated for the extraction of fats and oil from seeds [7–12] or microorganisms [13–18]. Surprisingly, despite the potential of liquefied gases for the extraction of volatile and sensitive molecules, only a few studies on aromatic compounds can be found in the literature [19–22].

To fill this gap, we tried to compare in this article the performance of three liquefied gases (*n*-butane, DME, and HFO-1234ze) as potential alternative solvents for the extraction of aromatic compounds from two plant materials: dry lavender flowers (*Lavandula angustifolia* Mill.) and

fresh orange peels (*Citrus sinensis* (L.) Osbeck). The solvent evaluation was performed using distinct approaches (Fig. 1). First, a predictive approach was conducted using the conductor-like screening model for real solvents (COSMO-RS) to predict the relative solubility of some selected target components in the liquefied gases. This theoretical approach was coupled with laboratory-scale experiments to assess the quantitative (extraction yields) and qualitative (chemical composition and sensorial analysis) aspects of the extracts to compare the efficiency of liquefied gases with *n*-hexane, chosen as a reference conventional solvent. Finally, their industrial applicability was evaluated in terms of safety, regulation, and environmental impacts.

2. Materials and methods

2.1. Chemicals

The liquefied gases used for pilot-scale extractions, that is, *n*-butane 95% purity, DME 99.9% purity, and *trans*-1,3,3,3-tetrafluoropropane-1-ene (HFO-1234ze) 99.5% purity, were purchased from Inventec Performance Chemical (St. Priest, France), whereas *n*-hexane of analytical grade was purchased from VWR International (Radnor, USA).

2.2. Plant materials

Dried lavender flowers (*L. angustifolia* Mill.; water content, 7.5%) and frozen orange peels (*C. sinensis* (L.) Osbeck; water content, 72%) were purchased from local suppliers.

2.3. Liquefied gases apparatus and extraction procedure

Extractions were performed using a 1-L pilot plant (NECTACEL, Celsius sarl, Villettel-de-Vienne, France) shown in Fig. 2 and described in previous works [8,9].

Liquefied *n*-butane extractions were performed using the following general procedure: first, 100 g of a plant material is introduced in a cellulose sock (porosity 50 μm) placed inside the extractor (5) and the entire unit is put under vacuum to remove air. Next, 0.9 L of liquefied gas is transferred from the gas bottle (0) to the storage tank (4). The solvent is then flowed to the extractor (5) containing the plant material. The mixture is maintained at 25 °C via the double jacket. After 1 h of maceration, the solvent containing the extract is transferred to the evaporator (7) where the solvent is evaporated. The solvent vapors naturally rise to the condenser (8) for recycling whereas the extract remains at the bottom of the evaporator. The plant material is then re-extracted three times, using the recycled solvent, for a maximum extraction yield. In the end, the total duration is four times 1 h. The remaining extract is finally collected in a flask and inner walls are washed with 100 mL of *n*-hexane for maximum recovery. Each extraction was made in duplicate.

2.4. Hexane macerations of lavender flowers and orange peels

As the liquefied gases chosen in this study are mostly lipophilic, *n*-hexane was chosen as a reference

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