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Short communication

Rectification extraction of Chinese herbs' volatile oils and comparison with conventional steam distillation

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ABSTRACT

Rectification is a very efficient technique for gas–liquid separation and has being widely applied in the fields of chemical, petrochemical engineering, etc. In order to explore adaptability of using rectification extraction (RE) to isolate volatile oils from Chinese herbs, RE and steam distillation (SD) techniques were applied to isolate volatile oils from three typical Chinese herbs *Bupleurum*, *Pogostemon cablin (Blanco) Benth* (PCB), *Pericarpium Citri Reticulatae* (PCR) in this paper. The experiment shows that SD technique cannot get volatile oils and only aromatic water from *Bupleurum* but RE can prepare both. RE technique increases 10% oil yield of PCB and 20% of PCR compared with SD, respectively. Therefore, RE would be more effective and suitable for enriching volatile oils from plants or herbs in which volatile oils are water-soluble and with low-content. GC–MS analysis of the products indicates that most compounds in volatile oils of PCB and PCR prepared by SD and RE methods (techniques) are the same, and the percentage contents of the most common compounds change little. Therefore, the quality features of volatile oils extracted by RE are similar to those produced by SD. So RE is not only a way to prepare samples for the aim of volatile oils' quality analysis but also a good alternative technology in industry for isolating volatile oils from Chinese herbs or other plants.

1. Introduction

Volatile oils, the bioactive components of many plant materials, are natural organic compounds with strong odors and have been widely used in daily life as food addictives, flavors, fragrances and pharmaceuticals, even in medicine directly for many centuries [1,2]. As reported in Chinese Pharmacopoeia [3], volatile oils of some Chinese herbs are added into 80 traditional Chinese medicines as active groups. Therefore, the preparation of volatile oils is very important both in laboratory for the aim of quality analysis and the production of traditional Chinese medicines. Many methods, such as traditional steam distillation (SD) and solvent extraction (SE) and some new techniques such as ultrasound [4,5] or microwave assisted extraction [6–8], supercritical fluid CO_2 extraction (SFE-CO_2) [9,10] and column partition fractionation [11] were applied to enrich volatile oils from plants. Ultrasonic and microwave assisted extraction of volatile oils from plants could accelerate volatile oils' dissolving rate and show advantage in laboratory analysis. SFE can increase yield of volatile oils, but the constituents of the volatile oils extracted by SFE usually differ from that of SD not only in the number of compounds, but also in the compound categories [12].

In Chinese Pharmacopoeia [13], SD was stipulated as the method of preparing volatile oils of the traditional Chinese medicines. However, volatile oils in some kinds of plants, such as traditional Chinese herbs *Liguisticum chuanxiong* Hort (Chuanxiong) and *Bupleurum* Chinese D.C. (Chaihu), are water-soluble and with low-content. These volatile compounds cannot be efficiently isolated from the plant materials by SD. Organic solvent extraction is often applied to further isolate the volatile oils from water after SD. For example, Tian et al. extracted and analyzed volatile oils from leaves of *Eucalyptus dunnii* and *Eucalyptus citriodora*. They not only collected oils by SD but also obtained volatile oils from residual distillation liquid, using ether as extraction solvent. Some volatile constituents, such as aromadendrin, existed both in the oil and water phase [14]. However, the toxic residue would increase safety risk of the oils extracted by solvent extraction.

Rectification is a very efficient technique for gas–liquid separation and has being widely applied in the fields of chemical, petrochemical engineering, etc. [15] In our previous work [16], we applied RE technology to isolate volatile oils from Chinese herb *Ligusticum chuanxiong* Hort. The oil yield of the volatile oils extracted by RE (0.6%) is triple of that by SD (0.2%) under the same extraction conditions. The common constituents occupied 98.94%

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and 98.80% of the volatile oils by SD and RE, respectively. The result indicates that RE can not only increase oil yield of Chuanxiong but also keep similar quality feature to that of Chuanxiong's volatile oils by SD. The work exhibits that RE maybe a good alternative technique to prepare volatile oils from herbs.

Bupleurum, *Pogostemon cablin (Blanco) Benth* (PCB) and *Pericarpium Citri Reticulatae* (PCR) are three kinds of Chinese herbs widely applied in Chinese medicine. There are volatile oils, saponins, etc. in *Bupleurum* [17–19]. Ni et al.'s investigation also indicates that volatile constituents of *Bupleurum* can strengthen antipyretic efficacy of its extract [20]. However, the volatile oils of *Bupleurum* are very few and easily dissolve in water, which makes the extraction of its volatile oils very difficult.

PCB has antiemetic effect and can improve appetite, alleviate discomfort feelings caused by heat stroke such as chest distress, dizziness, and nausea [21] A traditional Chinese medicine, Ageratum-liquid, in which patchouli oil is a main active group, is widely applied in China. Besides, patchouli oil is also a widely applied flavor in fragrance and flavor industry and usually extracted by SD technology.

The volatile oils of PCR can promote secretion of digestive juice and eliminate intestinal pneumatosis, and have effect of resolving phlegm and stopping cough, dredging cardio-cerebrovascular [22]. Thus volatile oils of PCR are widely applied in many traditional Chinese medicines.

Among these three herbs, *Bupleurum* are the dried radix of *Bupleurum* Chinese D.C., whose volatile oil content is very low (<1%). PCB is the leaf of *P. cablin* and PCR the dry peel of oranges or *Citrus reticulata*. The volatile oil contents both in PCB and PCR are relatively high (>2%). Therefore, the herbs are representative whether in appearance morphology and volatile oil contents. In order to investigate the adaptability of RE method for isolating volatile oils from Chinese herbs, both RE and SD were applied to isolate volatile oils from the three typical herbs and GC–MS was applied to analyze the products.

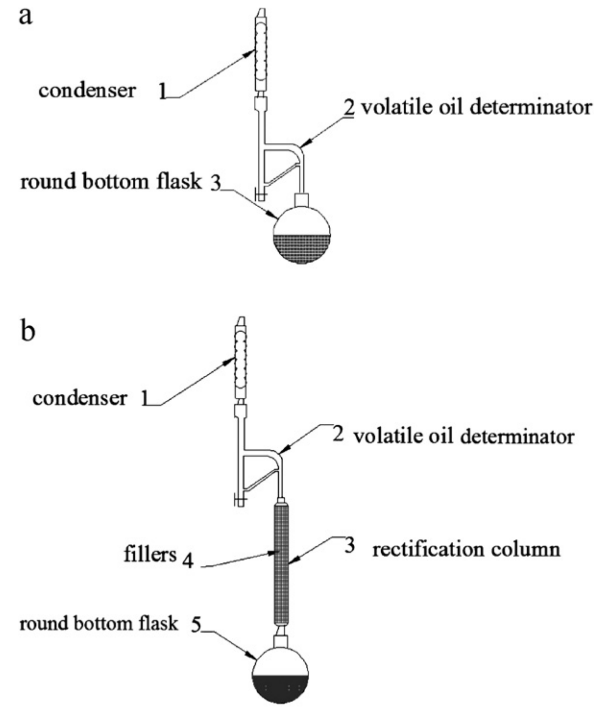

Fig. 1. Schematic diagrams of conventional steam distillation device (a) and rectification extraction device (b).

2. Experiment and principle

2.1. Materials and apparatus

Chengteh Jingfukang Pharmaceutical Co. Ltd. of China provided *Radix Bupleurum* and *Pericarpium Citri Reticulatae* (PCR), which were identified as dry rhizomes of *Bupleurum* Chinese D.C. and dry peels of oranges, respectively, by pharmacist Chunmin Wang, Chengteh Jingfukang Pharmaceutical Co. Ltd. of China. Firmenich Aromatics (China) Co., Ltd. provided patchouli, and Dr. Yong-Ming Yuan (botanist) of Firmenich Company identified that the PCB was leaf of *P. cablin (Blanco) Benth*. Methanol was HPLC grade (E. Merck, Darmstadt, Germany).

Clarus 500 GC-MS (PerkinElmer Company, USA), a HP-5 MS capillary column (30 mm × 0.25 mm i.d.) coated with 0.25 μm film 5% phenyl methyl siloxane was used for separation, and MS search V.2.0 (2) software was used to analyze volatile oil components.

Volatile oil determinator (i.e., Clevenger apparatus) was made by Shanghai glass instrument factory according to the Chinese Pharmacopoeia appendix XD.

2.2. Principle and apparatus of steam distillation (SD) method

Conventional steam distillation apparatus (Clevenger apparatus) for extracting volatile oils consists of an extractor with heating facility, a condenser and a volatile oil determinator, as shown in Fig. 1a, which was stipulated in the Chinese Pharmacopoeia. Extracting volatile oils from plants by SD goes through solid-to-liquid, liquid-to-gas, gas-to-liquid and liquid-to-liquid mass transfer processes. Herb materials and water are heated in the extractor to boiling point, and volatile constituents then begin dissolving into the water, i.e., they transfer from herbs (solid phase) into water (liquid phase). Then volatile constituents gradually evaporate from the extraction liquid to steam during heating period, this is the second process of volatile constituents transfer from liquid phase to gas phase. Furthermore, the steam containing volatile oils would condensate in the condenser, this is the third mass transfer step that volatile constituents transfer from gas phase to liquid phase. If the content of volatile constituents in the herbs (or plants) is high enough, the concentration of volatile oils in the condensate is higher than their saturation solubility in water, then volatile oils would transfer from water phase into oil phase, which is called liquid–liquid mass transfer process in chemical unit operation. The water in the volatile oil determinator will flow back to the flask to supplement the water consumed in the evaporating process.

Liquid in the extractor continuously evaporated into the condenser during the process of SD, the amount of volatile oils extracted from herbs depends on heating time to a large extent. In practice, heating time is limited on account of energy consumption. 5–6 h are often thought enough to completely isolate the volatile oils in the herbs [13].

In fact, if the concentration of volatile oils in the condensate is lower than their saturation solubility in water, it is difficult to transfer volatile constituents from water phase into oil phase by conventional SD method. According to above analysis and description of chemical separation principle of SD, the key step of isolating volatile constituents is to increase their concentrations in gas phase as much as possible, thus when they are condensed into liquid in the condenser, their concentration would be higher than their solubility in the water. Then the liquid in the volatile oil determinator would form two layers consisting of oil phase and water phase, thus the volatile oil could be easily separated from water.