Litsea cubeba

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Litsea cubeba is a special woody oil plant resource in China, which is mainly distributed in provinces and regions in the south of the Yangtze River. For the past several decades, *Litsea cubeba* essential oil has been the only product for trading, which can be extracted from its whole plant, including leaves, flowers, fruits, trunks, and even roots. Based on the zero-waste biorefinery concept, residues generated from *Litsea cubeba* essential oil processing have recently been found as potential sources for various plant-based products with added values, i.e., kernel oil with a high content of medium chain fatty acids might be used for the sustainable production of surfactants or even biofuels, pomace including protein might be used as a good source for feeding.

Keywords: woody oil plant resource ; valorization ; essential oil bioactivities ; citral-based value-added products ; kernel oil and protein ; zero-waste sustainable biorefinery

1. Introduction

Woody oil plant is the general term for the fruits or seeds of woody plants that can be used for oil extraction. It is a renewable resource for a diversity of oil products, such as edible oils, aromatic oils, industrial oils, and even oils for bioenergy [1]. The exploitation of woody oil plant based on the biorefinery concept is superior to conventional agriculture and more adaptable to climate change, which has advantages of using marginal land resources to develop value-added products and alleviating economic growth from dependence on fossil energy to some extent. Moreover, the industrial development of woody oil plants is also conducive to reduce the tight supply of edible oil, and meanwhile, maintain both economic stability and ecological security.

It is stated by the General Office of the State Council of China that 800 key counties would plant woody oilseeds like camellia, walnut, and peony by 2020, and a batch of standardized and industrialized demonstration bases would be established, resulting in the growing planting area from the existing 8 million hectares to around 13 million hectares, with an annual output of about 1.5 million tons of woody edible oils. Although woody oil plants in China are diverse and abundant, they are still unexploited or underdeveloped due to lack of scientific knowledge and systematic technical studies on intensive processing and integrated utilization.

The depletion of fossil energy and the unprecedented global climate change force people to eagerly seek raw materials for the available biomass, responding to the demand of sustainable energy supply [2][3]. In recent years, with the rapid development of society and the increasing demand for a better life, some local woody oil plants in China like *Camellia oleifera* have gradually attracted more and more attention in both domestic and foreign markets due to their unique chemical compositions and functional properties [4]. Besides this, *Litsea cubeba* was found as an emerging woody oil plant resource that can be implemented in a total valorization toward a zero-waste biorefinery, resulting in various plant-based products with added value for different application fields.

2. *Litsea cubeba* Essential Oil and Citral-Based Derivatives

Litsea cubeba is a deciduous shrub or a small tree belonging to the Lauraceae family, which is a characteristic potential industrial crop in China. This plant is photophilous and shade tolerant, the good germination of its seed can make it easy to survive in the undergrowth or at the roadside. The yield and composition of essential oils vary depending on plant organs extracted (i.e., fruits, leaves, flowers, roots, and trunk), environmental conditions (e.g., soil, altitude, climate and irrigation), and other parameters (e.g., harvest time, ripeness and postharvest technologies). The main chemical constituents of *Litsea cubeba* essential oil are terpenoids, especially for citral, whose biological activities including antioxidant, anti-inflammatory, antimicrobial, and anthelmintic capacities, have attracted more interests in recent years. Furthermore, high value-added synthetic products derived from citral have also been favored by enterprises in the field of perfume, flavor and fragrance, and nutraceuticals.

2.1. Extraction of *Litsea cubeba* Essential Oils

It is reported that the yield of essential oil from *Litsea cubeba* fruits is higher than that from other plant organs [5]. The traditional methods for extracting essential oil from *Litsea cubeba* is hydrodistillation, which is simple to operate but usually with a low extraction rate. Moreover, the long treatment time may cause unexpected changes in chemical composition and smell. Hence, innovative extraction technologies (e.g., microwave-assisted extraction, ultrasonic-assisted extraction, enzymatic-assisted extraction, supercritical fluid extraction, and other combined extractions) have been developed for essential oils in the past decade [6][7][8][9][10], which can not only enhance the extraction efficiency in a shorter time, but also maintain or improve the quality of essential oils.

As reported, emerging techniques like microwave and ultrasound could effectively enhance the extraction of *Litsea cubeba* essential oils with good quality, which paved the way for the total valorization of this plant. The strong penetrating power of electromagnetic microwaves could accelerate diffusion and transfer of essential oils from interior gland cells of fruit and peel tissues to exterior. Compared to traditional steam distillation, the optimized microwave-assisted extraction (MAE) could increase the yield by 36.5–37.5% but shorten the treatment time by four times [11]. Moreover, the citral content in essential oils was 5% higher and the amount of its loss in purification reduced by 33.3%. MAE was also optimized with an average essential oil yield of up to 10.29% (g/g), which could improve to 14.19% (g/g) with the combination of ultrasound [12].

Ultrasonic cavitation can speed up the breakage of plant cell walls, which is conducive to the release of essential oils and the penetration of solvents. Ultrasonic-assisted vacuum extraction was reported to have an extraction rate of 6.94% under optimal conditions, which had a citral content of as high as 87.65% [57]. Peng et al. [58] found that the yield of essential oils could be enhanced with the assistance of ultrasound, where the yield reached the highest when ultrasonic time was 25 min. Although crushing could increase the extraction ratio, the purity of essential oils was lower, which was not recommended to the extraction.

It is well known that enzymes are highly specific and have high catalytic efficiency with mild reaction conditions. Generally, the use of appropriate enzymes can improve the extraction rate without any damages to the raw materials, which is beneficial to the further exploitation of the residues after extraction. The yield of essential oil varies widely depending on relevant factors such as enzyme types and dosages. Xie et al. [13] used recombinant expansin and cellulase to extract the *Litsea cubeba* essential oil, which significantly increased the yield of essential oil compared to conventional extraction. The function of expansin is to break the hydrogen bond between cellulose microfilaments and hemicellulose. The interaction of expansin and hydrolase facilitates expansion and disruption of plant cell wall without any losses of nutrients. Therefore, expansin, as a product of genetic engineering, is worthy to be further explored regarding the high price and large amount of enzyme used in the large-scale extraction.

The type of solvent is of great importance to extraction. Supercritical CO_2 is one recognized green solvent for the extraction of bioactive components from natural plants, considering its excellent extractive properties, safety, eco-friendliness, and economica viability [14]. Supercritical CO_2 extraction could not only avoid oxidation, degradation, and organic solvent residues resulting from solvent extraction and distillation, but also overcome the disadvantages of pressing methods like the volatilization of essential oils and color darkening. Zhang et al. [15] studied the influencing factors in the supercritical CO_2 extraction of *Litsea cubeba* essential oil, where the particle size, CO_2 flow rate, extraction pressure, temperature, and time were optimized to obtain the highest extraction ratio. Although supercritical CO_2 extracts could maintain the naturalness of the raw materials, there are still some limitations for wide industrial applications of this technique like poor robustness of the early commercial equipment, lack of standard extraction procedures, difficulties in extracting polar compounds, and inefficiency in clean-up [16].

2.2. Bioactivities of *Litsea cubeba* Essential Oils

In China, *Litsea cubeba* is also called‘Bì Chéng Qié’, which has been commonly used as a traditional herbal medicine. From the perspective of traditional Chinese medicine, fruits, root bark, and leaves from *Litsea cubeba* can be used as medicines to treat stomach pain, vomit, fever, snake bites, swelling, cholera, and detoxification. Modern pharmacology studies have shown that *Litsea cubeba* extracts have a variety of biological activities such as antioxidant, antimicrobial, anti-inflammatory, anti-tumor, and insecticide, which gives it the potential to be widely used in the field of agriculture, medicine, food, flavor, and fragrance, etc.

2.3. Purification of *Litsea cubeba* Essential Oil and Its Derivatives

Citral is the main biologically active component in *Litsea cubeba* essential oils, which has two configurations including geranial (α-citral) and neral (β-citral). It is a light yellow volatile oily liquid with a lemon flavor, which is authorized as a food-grade flavor in China. Citral is mainly used in the preparation of lemon, citrus and assorted fruit-type food flavors, dishwashing detergents, soaps, toilet water, and flavoring agents. Also, it can be an intermediate for formulating and synthesizing high value-added functional ingredients, which are often used in perfumes and nutraceuticals.

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