

Proceeding Paper



Extraction Methods Applied to Natural *Lamiaceae*-Derived Compounds: An Overview Based on Patents ⁺

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Abstract: Secondary metabolites of plants are of major interest for pharmaceutical, cosmetic, and food applications. The extraction methods for these compounds must be optimized to have the best possible yield without altering the effectiveness of the targeted compounds. In this paper, we examine the methods of extraction of plant compounds, especially those applied to plants of the *Lamiaceae* family, renowned for their aromatic and medicinal roles. To do so, we consulted databases specialized in patent documentation, using appropriate keywords with the help of International Patent Classification (IPC) codes. Our results present the analysis of the 140 relevant documents selected. The first patent relating to our field of study was granted in the United States in 1998. It concerned a process for obtaining antioxidants from plant materials. While the year 2020 saw the registration of the largest number of these documents (15). Most of the documents identified (66) were filed in China. The French company Naturex, which specializes in plant-based products, is the number one depositor in the field of plant extraction. The relevant patents selected describe processes using various extraction methods and agents, most of which are valid for many plants, while some focus on genera of *Lamiaceae*, such as *Salvia, Ziziphoria*, and *Clinopodium*.

Keywords: extraction; Lamiaceae; innovation; patent; plant compound

1. Introduction

Extraction methods for natural plant-derived compounds are essential processes used to isolate bioactive molecules, flavors, fragrances, and other valuable substances from plants [1]. However, it is essential to note that these methods vary depending on the type of compounds, the plant source, and the equipment used [2].

There are a large number of medicinal and aromatic plants (MAPs), and various varieties are cultivated extensively for medicinal, ornamental, and culinary purposes, among them the *Lamiaceae* [3]. The *Lamiaceae* family comprises approximately 200 genera and around 3300 species, consisting of aromatic plants that are either annual or perennial herbs or low shrubs. This plant family finds significant representation in both the Mediterranean region and the British Isles. The flowers within this family are typically bisexual and exhibit zygomorphic characteristics. These flowers are often organized in clusters known as verticillasters [4].

The Lamiaceae genera include, among others, Rosmarinus, Lavandula, Marrubium, Lamium, Salvia, Origanum, Thymus, Mentha, and Ocimum [5]. Many members of the family are used as culinary or medicinal herbs, as sources of volatile oils, and, in some cases, for the preparation of constituents of volatile oils such as menthol and thymol [6]. Besides volatile oils, the Lamiaceae family encompasses a diverse array of compounds. These con-

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Copyright: © 2023 by the authors. Submitted for possible open access publication under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/license s/by/4.0/). stituents include, but are not limited to, diterpenoids and triterpenoids, flavonoids, saponins, tannins, alkaloids, and iridoids, along with their glycosides, quinones, furanoids, cyclitols, and coumarin, as well as the sugars raffinose and stachyose [5,7].

The diversity of natural *Lamiaceae*-derived compounds implies the need to adapt extraction methods to achieve the best possible selectivity. Moreover, the extraction methods for these compounds must be optimized to have the best possible yield without altering the effectiveness of the targeted compounds [5,8].

In this work, we examine the extraction methods of plant compounds based on patents, which can serve as a reference for researchers interested in MAPs as well as phytotherapy and cosmetology. Our overview focuses especially on extraction methods applied to plants of the *Lamiaceae* family.

2. Resources and Methods

To identify the most commonly used extraction method related to natural *Lamiaceae*derived compounds and the innovation underlying it, we examined four patent documentation databases:

- 1. Patentscope [9];
- 2. PatFT-AppFT database [10];
- 3. Espacenet Patent Search [11];
- 4. Lens Patent Data Set [12].

Different terms and keywords related to "extraction methods applied to natural *La-miaceae*-derived compounds" have been used. Further, the International Patent Classification (IPC) codes have also been used to refine the results. The IPC is defined as "*a code-based hierarchical system that separates all technological domains into sections, classes, subclasses, groups, and subgroups.*" [13,14]. In our case, the code A61k36/53 was employed. It concerns medicinal preparations of undetermined constitution containing material from plants or derivatives such as *Lamiaceae* [15,16]. Finally, the found patent documents have been filtered to include only patent applications and granted patents.

3. Results and Discussion

Following our research methodology and using the four cited databases, we found 140 patent documents between 1978 and 2023. There are concerning 112 patent applications and 28 granted patents.

Figure 1 presents the evolution of patent documents (i.e., patent applications and granted patents) related to innovation in *Lamiaceae* component extraction as a function of publication date (year).



Figure 1. Evolution of patent documents (i.e., patent applications and granted patents) related to innovation in *Lamiaceae*-component extraction.

Based on our previous study, we define the publication date as "the date on which a patent document is published with an assigned publication number by a patent authority, thereby making it part of the state of the art." [1]. The first patent application in the field of plant extraction, in relation to Lamiaceae, was filed in 1977 (and published in 1978) in Japan [17]. On the other hand, the first patent was granted in 1998 in the United States for a process for obtaining antioxidants from plant materials [18]. Moreover, the number of patent applications filed has steadily increased in recent years, with a peak of 15 patent documents in 2020.

We previously defined patent applicant and patent jurisdiction, respectively, as "*ei*ther a natural person or a legal entity that submits a patent application. In some cases, the applicant can also be the inventor, and there may be more than one applicant for a single patent application." and "the country or region where applicants can file a patent application through the appropriate patent office." [1].

For innovation in *Lamiaceae*-component extraction, Figure 2 presents the top 10 applicants and the top 10 jurisdictions. As a result, the French botanical company Naturex (Avignon, France) is the leading patent depository in this field of study. It specializes in the production and distribution of plant extracts and plant-based natural ingredients tailored for the food industry, serving various purposes such as flavoring, coloring, and preservation. The company boasts a global presence, with manufacturing facilities spanning, among others, Europe, the United States, Canada, Brazil, Morocco, and India. On the other hand, "Avignon Université" (Avignon, France) is leading the way in patenting the extraction methods applied to natural *Lamiaceae*-derived compounds.

China is the jurisdiction with the most patents in this area (i.e., 66 patent documents), followed by the United States, Europe's jurisdiction, and the Patent Cooperation Treaty (PCT) international system. This system, overseen by the World Intellectual Property Organization (WIPO), enables inventors to obtain protection for their inventions in many countries worldwide by filing a single patent application [19].



Figure 2. Patent information related to innovation in *Lamiaceae*-component extraction: (**a**) Top 10 applicants; (**b**) Top 10 jurisdictions.

Through the analysis of the selected granted patents (Supplementary Materials: Table S1), the extraction methods applied to natural *Lamiaceae*-derived compounds are summarized in Figure 3. These extraction methods concern nine techniques classified by the number of granted patents appearing as fellows: decoction, maceration, supercritical fluid extraction, soxhlet extraction, ultrasound-assisted extraction, pressurized liquid extraction, microwave-assisted extraction, extraction by eutectic solvents, and mechanochemical-assisted extraction. Based on prior investigations, Table 1 outlines each extraction method according to its description, strengths, and weaknesses [20–22].



Figure 3. The extraction methods cited and used in the studied granted patents (Supplementary Materials: Table S1).

Table 1. A summary of plant extraction methods cited in selected granted patents * (The description of the methods is adapted from Jha and Sit, 2022 [20], Fan et al., 2022 [21], and Lefebvre et al., 2021 [22]).

Extraction Method	Description	Strengths	Weaknesses
Decoction	The plant is boiled in a solvent.	More effective than macera- tion for extracting water-solu- ble compounds.	Degradation of heat sensitive compounds.
Maceration	The plant is left to soak in a solvent for a certain period of time.	It is inexpensive and easy to implement.	Ineffective for extract- ing the most water-sol- uble compounds.
Supercritical fluid ex- traction	The solvent is heated to a supercritical state for selective extraction.	Selective and efficient extrac- tion.	Expensive.
Soxhlet extraction	The solvent is heated and circulated around the plant.	Complete and efficient extrac- tion.	Long and expensive.
Ultrasound-assisted ex- traction	Sound waves break down plant cells and facilitate the extraction of com- pounds.	Rapid and efficient extrac- tion.	Expensive.
Pressurized liquid ex- traction	The solvent is pressurized and heated to a higher temperature.	Rapid and efficient extrac- tion.	Expensive.
Microwave-assisted ex- traction	Microwaves heat the solvent for faster extraction.	Rapid and efficient extrac- tion.	Degradation of heat sensitive compounds.
Extraction by eutectic solvents	The solvent is a mixture of two or more compounds that has a very low melting point.	More efficient than tradi- tional extraction methods.	Safer because tempera- tures are lower.
Mechanochemical-as- sisted extraction	The solvent is used with mechanical en- ergy to break down plant cells and facil- itate the extraction of compounds.	More efficient and selective than traditional methods.	Less damaging to the plant material, which may limit the extrac- tion of certain com- pounds.

* The detailed list of the selected granted patents are presented in Table S1 (Supplementary Materials). Finally, analysis of the selected granted patents ensures that the relevant inventions selected describe processes for obtaining different natural compounds using various extraction methods and agents, most of which are valid for many plants, while some focus on genera of *Lamiaceae*. In this regard, *Lamiaceae* genera concerned by the extraction innovation in this study focus on genera such as *Thymus*, *Mentha*, *Origanum*, *Salvia*, *Rabdosia*, *Clerodendranthus*, etc. (Figure 4).



Figure 4. Lamiaceae genera that are concerned by the extraction innovation.

4. Conclusions and Outlook

In this patent study, we examined the methods of extraction of *Lamiaceae*-derived compounds renowned for their aromatic and medicinal roles. According to our research methodology, 140 patent documents were found between 1978 and 2023. They encompass 112 patent applications and 28 granted patents. The relevant patents selected describe processes using various extraction methods and agents, most of which are valid for many plants, while some focus on genera of *Lamiaceae*. These extraction methods concerned nine techniques, such as decoction, maceration, and supercritical fluid extraction. However, each method has its benefits and drawbacks, and scientists frequently choose the most appropriate technique based on their objectives and the resources at their disposal. Moreover, the selection of the extraction method depends especially on the specific compounds, the characteristics of the plant material, and the suitable final product.

Supplementary Materials: The following supporting information can be downloaded at: www.mdpi.com/xxx/s1, Table S1: The detailed list of the selected granted patents related to extraction methods applied to natural *Lamiaceae*-derived compounds.

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