



Proceeding Paper

# Extract of Rosemary as Food Additive: The Landmark Patents †

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Abstract: Food additives are substances of natural origin or obtained by chemical synthesis. Among these additives, food antioxidants play an essential role in preserving food quality by neutralizing free radicals and inhibiting oxidation reactions, which helps prevent food spoilage and maintain its freshness, taste, and nutritional value. In addition, dietary antioxidants may also be beneficial to health. This is particularly the perception that consumers have in relation to antioxidants of natural origin, such as plant extracts. In this paper, we focused on the case of rosemary extract as a food additive which is authorized in many countries, including the United States and Europe, under the code E392. Through the examination of patent documents available in specialized databases and using appropriate keywords, our research made it possible to go back in time to first show the uses of rosemary extract and then specify the exploitation of rosemary extract in the field of food. We selected 411 relevant documents (granted patents and patent applications), almost 40% of which were filed in China. The first patent found was published in 1984, and it concerned a method to obtain a highly effective antioxidant from rosemary suitable for preventing the oxidation of food. This patent was followed by others which claimed, among others, more effective methods of making rosemary extract. More recently, innovations have focused on the synergistic effect of rosemary extract with other compounds, particularly for the antimicrobial action of food preservation.

Keywords: rosemary; antioxidant; food additive; patent



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#### 1. Introduction

Rosemary is a small, evergreen perennial shrub of the Lamiaceae family, known for its medicinal and aromatic properties [1]. Its extract is used in food [2]. It acts on the shelf life of foodstuffs, reducing spoilage by oxidation [3,4]. Rosemary extract is usually produced from dried rosemary leaves ground using food-grade ethanol or acetone as solvents. After the separation of the dried leaves via concentration and/or precipitation, rosemary extract powder is obtained via the filtration of the leaf residues, the evaporation of the solvent under vacuum, drying, and sieving. The final marketed product may undergo deodorization and discoloration steps with food-grade excipients [5]. Rosemary extract (Salvia rosmarinus) is now widely used in food. It has been recognized worldwide as a safe natural antioxidant [6]. This antioxidant effect is mainly attributed to phenolic diterpenes such as carnosol and carnosic acid and phenolic acids such as rosmarinic and caffeic acids [7,8]. In the European Union, it is authorized as an additive under code E392 in several types of food, such as dehydrated milk, practically anhydrous fats and oils, fruit and vegetable preparations, nut butters and nut spreads, and meat products [9]. Regarding the importance of natural additives, they are perceived by consumers as healthier [10]. In this paper, we present an analysis of patents that directly relate to rosemary extract and then focus on the exploitation of the plant extract studied in the field of food, presenting chronologically classified data from relevant patent documents.

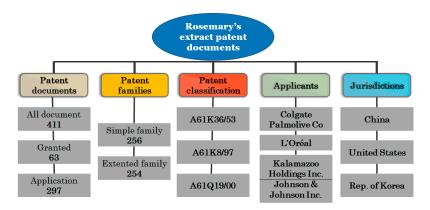
# 2. Resource and Methodology

The patent documents selected for our analysis were of the application or granted patent type. In order to consider the most relevant documents, the search was performed on patent titles, using appropriate keywords. We searched different patent databases, namely Lens [11], PatentGuru [12], and Patentscope [13].

## 3. Results and Discussions

### 3.1. Analysis of Patents Related to Rosemary Extract

Figure 1 presents data on patent documents whose innovation relates to the use of rosemary extract. For greater relevance, only documents whose titles evoked rosemary or *rosmarinus* extract were studied. Thus, 411 documents were identified, about 72% of which are patent applications and about 15% are granted patents. In total, 30% of all these documents are still active.



**Figure 1.** Summary of extracted data from patent documents related to the use of rosemary extract (top 3).

These documents constitute 256 simple families which represent a group of patent documents relating to the same innovation, known as priority. They form 254 extended families which represent a grouping of patent documents relating to a common technology [14].

International patent classification (IPC) codes are applied to patents [15]. According to this classification, the documents studied mainly refer to preparations for medical, dental, or toilet use (A61K), containing plant material either from *Lamiaceae* (A61K36/53), or for herbal cosmetic use (A61K8/97). At the level of the top ten IPCs used in the documents studied, we observed two codes directly related to food technologies. In seventh place is the code relating to the preservation of food or food products (A23L3/3472), and in tenth place is the code relating to the modification of the nutritional quality of foods or dietary products by the addition of a plant extract, their artificial duplicates, or their derivatives (A23L33/105) [16].

According to the data collected, the top three patent applicants in our field of study are primarily companies' goods in the cosmetic and pharmaceutical fields. However, the company Kalamazoo Holdings Inc. (Kalamazoo, MI, USA), which specializes in the manufacture and processing of food products, also represents the food industry in this ranking.

According to the regulation of patent rights, the legal protection of this form of intellectual property protection has a specific geographical scope [17]. In our case, China, with 162 patent documents, tops the list of jurisdictions where the selected patent documents were filed, followed by the United States (45 patent documents) and the Republic of Korea (43 patent documents).

#### 3.2. Relevant Patents Filed for Using Rosemary Extract in Food

In order to show the evolution of the patentability of processes and products related to rosemary extract, we selected nine patent documents according to the chronological Biol. Life Sci. Forum **2023**, 26, 37

evolution criteria, considering the oldest and most recent documents as well as the most relevant patents (Table 1).

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Identifier	Title	Published	Family *	Reference
US4450097A	Antioxidative compound, method of extracting same from rosemary, and use of same	22 May 1984	1 s/5 ex	[18]
CN1144835A	Method for extracting antioxidant from rosemary plant	12 March 1997	1 s/1 ex	[19]
AU703958B2	High purity carnosic acid from rosemary and sage extracts by pH-controlled precipitation	1 April 1999	8 s/8 ex	[20]
WO2000053206A1	Rosemary having highly efficacious antioxidant extracts	14 September 2000	2 s/2 ex	[21]
ES2159246B1	Carne precocinada para almacenamiento refrigerado a largo plazo.	1 March 2003	3 s/3 ex	[22]
CN1281709C	Method of extracting rosemary as natural antioxidant	25 October 2006	2 s/2 ex	[23]
RU2680901C1	Method of manufacture of coffee ice cream with liquor and nanostructured rosemary extract	28 February 2019	1 s/1 ex	[24]
US11045514B2	Method of using a rosemary extract composition for weight management	29 June 2021	3 s/3 ex	[25]
US11559058B2	Synergistic antimicrobial effects among rosemary extract, cultured dextrose and buffered vinegar	24 January 2023	9 s/9 ex	[26]

<sup>\*</sup> s: simple family; ex: extended family.

The oldest patent that exploits the antioxidant property of rosemary for food use was published in 1984 [18]. This patent document describes the process by which rosemary is steam-distilled. The extracted residue with a non-polar solvent is treated with an alkaline solution to collect the desired extract. The organic solvent used is methylene chloride or benzene, while the distillation residue is treated with ethyl ether or n-hexane. The purpose of steam distillation is to remove any unwanted volatile matter when the antioxidant is added to food, making this extraction method suitable for food preservation (Figure 2) [18].

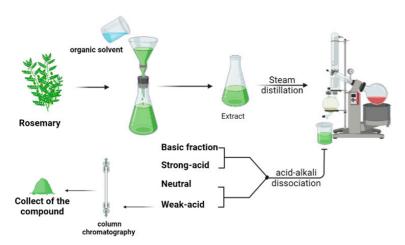


Figure 2. Illustration of the process described in the patent of Nakatani et al., 1984 [18].

In 1997, another process for extracting compounds from rosemary was published [19]. In addition to steam distillation, this process uses ultra-critical  $CO_2$  extraction to produce refined rosemary oil. The antioxidant is then extracted with solvents. The refined oil

obtained can be used in the perfume industry, while the antioxidant obtained is suitable for use in the food industry, especially since its extraction is inexpensive [19].

Two years later, a patent claimed a process for extracting and isolating concentrated carnosic acid from plants of the *Lamiaceae* family, such as rosemary [20]. The plant material is placed in contact with a water-miscible solvent to form a plant extract. Adjusting the pH of the plant extract reduces it to a value between 7 and 10. The addition of water causes impurities to precipitate, which will be removed, while carnosic acid is preserved in the solution [20].

Given the interest in rosemary compounds and the importance of their industrial applications, extraction processes tried to achieve the maximum possible yield. However, to have an extract richer in desired compounds, selection, crossing, and plant modification are proving to be potential solutions. As an example, in the year 2000, a published patent claimed that rosemary plants had extracts with antioxidant activity superior to that found in natural plants [21]. The process described therein is based on the production of heterozygous seeds by pollination. The plants produced by these seeds were crossed to obtain the best possible varieties [21].

In 2001, rosemary extract was cited, along with phosphate and citrus concentrate, as the main components of a combination of antioxidants used to preserve the flavors of a meat preparation [22]. Thus, this meat can be frozen for a long time, at least 60 days. Indeed, this patent claims a process using this antioxidant mixture for the preservation of meat precooked at about 76 °C, vacuum-packed, and frozen. The method described also provides for antimicrobial and surface treatment by caramel coloring (i.e., caramelizing) the prepared meat to ensure optimal organoleptic qualities [22].

In 2006, a granted patent claimed to significantly improve the extraction method to be used for rosemary [23]. Indeed, this invention directly uses fresh grass, which is steam-distilled to obtain refined rosemary oil and an antioxidant material. Thus, following a rapid grinding of the plant matter, it is mixed with an edible-grade organic solvent in a leaching device at a ratio from 1:5 to 1:3. The supercritical extraction and high-pressure separation of the leaching liquid make it possible to recover, after drying, a yellowish antioxidant. This antioxidant, being separated from the refined oil, will not have a particular flavor and can be directly used for food preservation. This method has the advantages of high efficiency, simple technology, and time savings, which reduce the investment in equipment and lower operating costs on a large industrial scale [23].

Among the inventions that benefit from the bioactive substances of rosemary in the food industry, a patent published in 2019 claims a process for producing coffee ice cream with rosemary extract and liquor [24]. The description of the patent provides for the addition, in the process of manufacturing a frozen dessert, of freeze-dried coffee and nanostructured rosemary extract in sodium alginate or carrageenan, a liquor that can be added when cooling the mixture to a temperature of about  $-21\,^{\circ}\text{C}$ . In an example of the execution of the invention, it is intended to use 1 g of rosemary extract and 180 mL of liquor for each 1 kg of ice cream produced [24].

Rosemary extract is mainly used as a food-preserving antioxidant; however, we can find other uses for it in food. In 2021, a patent was granted for a process for reducing the body weight of an obese individual [25]. The claimed composition comprises an extract of rosemary with about  $50\% \ w/w$  ursolic acid. This rosemary extract inhibits the activity of pancreatic lipase. Although this innovation appears to be for medical purposes, it has a dietary execution of said composition in the form of a food, snack, nutritional supplement, food supplement, or beverage [25].

Among recent patents, an innovation published this year presents an antimicrobial composition including rosemary essential oil (minimum concentration of 0.5%), cultivated dextrose (minimum concentration of 0.112%), and buffered vinegar (minimum concentration of 0.044%) [26]. The proposed composition has synergistic antimicrobial activity among the components that constitute it, allowing for high efficacy of the product. In addition, the

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invention meets the expectations of consumers for natural, healthy, and eco-friendly food products thanks to the natural origin of these constituents [26].

#### 4. Conclusions

The analysis of the patent documents collected shows that rosemary is widely exploited for its medicinal virtues, especially for its antioxidant effect. It was shown that China is the jurisdiction in which the largest number of patents relating to our field of study have been filed. These results come from both the exploitation of the rich traditional Chinese pharmacopoeia and from the importance of the Chinese market for this kind of application. Our research also presents the most significant patents that reflect the exploitation of rosemary extract as a food additive. It was observed that extraction methods for the antioxidant constituents of rosemary have evolved toward simpler and more effective techniques that are also less expensive. Considered an effective and safe antioxidant, it was also observed that rosemary extract is introduced into very different food preparations.

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#### References

- 1. El Boukhari, R.; Fatimi, A. A review of the patentability of rosemary-derived drugs and bioactive compounds. *Drugs Drug Candidates* **2023**, *2*, 172–188. [CrossRef]
- 2. Rocío Teruel, M.; Garrido, M.D.; Espinosa, M.C.; Linares, M.B. Effect of different format-solvent rosemary extracts (*Rosmarinus officinalis*) on frozen chicken nuggets quality. *Food Chem.* **2015**, *172*, 40–46. [CrossRef] [PubMed]
- 3. Yao, Y.; Liu, Y.; Li, C.; Huang, X.; Zhang, X.; Deng, P.; Jiang, G.; Dai, Q. Effects of rosemary extract supplementation in feed on growth performance, meat quality, serum biochemistry, antioxidant capacity, and immune function of meat ducks. *Poult. Sci.* 2023, 102, 102357. [CrossRef] [PubMed]
- 4. Yang, J.; Goksen, G.; Zhang, W. Rosemary essential oil: Chemical and biological properties, with emphasis on its delivery systems for food preservation. *Food Control* **2023**, *154*, 110003. [CrossRef]
- 5. Jannavi, R.S. Rosemary extract. In Proceedings of the 82nd JECFA—Chemical and Technical Assessment (CTA), Geneva, Switzerland, 7–16 June 2016.
- 6. Martínez, M.L.; Penci, M.C.; Ixtaina, V.; Ribotta, P.D.; Maestri, D. Effect of natural and synthetic antioxidants on the oxidative stability of walnut oil under different storage conditions. *LWT-Food Sci. Technol.* **2013**, *51*, 44–50. [CrossRef]
- 7. Cuvelier, M.-E.; Richard, H.; Berset, C. Antioxidative activity and phenolic composition of pilot-plant and commercial extracts of sage and rosemary. *J. Am. Oil Chem. Soc.* **1996**, *73*, 645–652. [CrossRef]
- 8. Frankel, E.N.; Huang, S.-W.; Aeschbach, R.; Prior, E. Antioxidant Activity of a Rosemary Extract and Its Constituents, Carnosic Acid, Carnosol, and Rosmarinic Acid, in Bulk Oil and Oil-in-Water Emulsion. *J. Agric. Food Chem.* **1996**, *44*, 131–135. [CrossRef]
- 9. European Union. Regulation (EC) No 1333/2008 of the European Parliament and of the Council of 16 December 2008 on food additives. *Off. J. Eur. Union* **2008**, *L*354/16, 16–33.
- 10. Bearth, A.; Cousin, M.-E.; Siegrist, M. The consumer's perception of artificial food additives: Influences on acceptance, risk and benefit perceptions. *Food Qual. Prefer.* **2014**, *38*, 14–23. [CrossRef]
- 11. Cambia Institute. The Lens Patent Data Set. Available online: www.lens.org (accessed on 25 July 2023).
- 12. Patent Guru Inc. PatentGuru Patent Database. Available online: www.patentguru.com (accessed on 25 July 2023).
- 13. World Intellectual Property Organization. The Patentscope. Available online: https://patentscope.wipo.int (accessed on 25 January 2023).
- 14. Fatimi, A. Cellulose-based hydrogels: Patent analysis. J. Res. Updates Polym. Sci. 2022, 11, 16–24. [CrossRef]

- 15. Fatimi, A. A patent data analysis of the innovation trends in biological control agent formulations. *Recent Adv. Food Nutr. Agric.* **2022**, *13*, 59–69. [CrossRef] [PubMed]
- 16. World Intellectual Property Organization. IPC Publication. Available online: www.wipo.int/classifications/ipc/ipcpub (accessed on 25 July 2023).
- 17. Fatimi, A. Exploring the patent landscape and innovation of hydrogel-based bioinks used for 3D bioprinting. *Recent Adv. Drug Deliv. Formul.* **2022**, *16*, 145–163. [CrossRef] [PubMed]
- 18. Nakatani, N.; Inatani, R.; Konishi, T. Antioxidative Compound, Method of Extracting Same from Rosemary, and Use of Same. U.S. Patent 44,500,97A, 22 May 1984.
- 19. Wenzhong, W.; Guangyu, W.; Lijian, S. Method for Extracting Antioxidant from Rosemary Plant. CN1144835A, 12 March 1997.
- 20. King, B.; Bank, V.R.; Bailey, D.T.; Richheimer, S.L. High Purity Carnosic Acid from Rosemary and Sage Extracts by pH-controlled Precipitation. AU703958B2, 1 April 1999.
- 21. Greaves, J.A.; Haworth, J.E.; Brinkhaus, F. Rosemary Having Highly Efficacious Antioxidant Extracts. WO2000053206A1, 14 September 2000.
- 22. Lloyd, S.D.; Ellen, S.B.; Jeffrey, G.S. Carne Precocinada Para Almacenamiento Refrigerado a Largo Plazo. ES2159246B1, 10 August 1999.
- 23. Ma, W.P. Method of Extracting Rosemary as Matural Antioxidant. CN1281709C, 25 October 2006.
- 24. Krolevets, A.A. Method of Manufacture of Coffee Ice Cream with Liquor and Nanostructured Rosemary Extract. GRU2680901C1, 28 February 2019.
- 25. Prasad, K.S. Method of Using a Rosemary Extract Composition for Weight Management. U.S. Patent 11045514B2, 29 June 2021.
- 26. Lee, A.; Beckett, A.; Joseph, P.; Stoufer, S.; Pham-Mondala, A. Synergistic Antimicrobial Effects among Rosemary Extract, Cultured Dextrose and Buffered Vinegar. U.S. Patent 11559058B2, 24 January 2023.

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