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山苍子精油的化学成分和功能活性 及其应用研究进展

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摘要:【目的】山苍子是我国特色木本精油植物资源,也是药材和生物质能源树种。山苍子的根、茎和果实均可入药,具有祛风散寒、理气止痛等功效,在民间被广泛应用。山苍子的花、叶、枝条和果实等部位均含有精油,其中果实的含油率最高,是提取山苍子精油的主要原料。山苍子精油具有浓厚的柠檬香味,是一种淡黄色到橙黄色的油状液体,成分复杂,主要是由单萜和倍半萜类化合物组成,它的主要成分柠檬醛是食品添加剂、合成香料、香水及制药工业等的重要原料,具有较高的经济价值。因此,对山苍子精油的功效成分、提取方法、功能活性及其应用等方面进行总结,旨在为山苍子精油的综合利用和深度开发提供参考。【方法】对山苍子精油的提取方法、化学成分和功能活性方面的研究进展进行系统介绍,并阐述山苍子精油在医学、食品和化妆品等相关领域中的应用。【结果】不同的提取方法对山苍子精油中化学成分的组成及含量均有影响。目前,提取山苍子精油的主要方法有水蒸气蒸馏法、超临界二氧化碳萃取法、微波辅助萃取法和酶法等,其中水蒸气蒸馏法是山苍子精油最传统的提取方法。山苍子精油具有良好的抗氧化、抑菌、杀虫和抗炎等多种生物活性,可以用来生产抗微生物的药物、杀虫剂和抗氧化剂等,被广泛用于医疗、食品及化工行业,可作为食品防腐剂、保鲜剂的重要原料。【结论】山苍子精油因其抑菌、抗炎、驱避杀虫和抗氧化等功能活性在实际应用中取得了积极的效果,具有广阔的应用领域和良好的发展前景。文章最后简要分析了山苍子精油在实际应用中存在的问题,在山苍子精油建立质量判断标准、提高稳定性和开发新型饲料添加剂等研究方面需要深入探索和研究,以期在山苍子精油的研究提供思路,对山苍子精油开发利用起到促进作用。

关键词:山苍子;精油成分;功能活性;应用;研究进展

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Research Progress in Application and Functions of *Litsea cubeba* Essential Oil Components

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Abstract: [Objective] *Litsea cubeba* is a characteristic woody essential oil plant resource in China, as well as a medicinal material and biomass energy tree species. Its roots, stems and fruits can all be used as medicine, which have been traditionally widely used for removing pathogenic wind and dampness, smoothing circulation and stopping pains in the folk. The flowers, leaves, branches and fruits contain essential oils, while the essential oil content in the fruit is the highest and it is the main raw material for extraction of it. The essential oil from *Litsea cubeba* is a yellowish to orange-yellow oily liquid with a strong lemon flavor. The composition of the essential oil is complex, mainly being composed of monoterpenes and sesquiterpenes and the main component is citral, which is an important raw material for food additives, synthetic spices, perfumes and pharmaceutical industry with a high economic value. Therefore, through the summary of researches on the functional components, extraction methods, functional activities and applications of *Litsea cubeba* essential oil, the aim of this study is to provide a theoretical reference for the comprehensive utilization and in-depth development of *Litsea cubeba* essential oil. [Method] This study systematically reviews the researches on the extraction methods, chemical components and functional activities of *Litsea cubeba* essential oil, also elaborates the application of *Litsea cubeba* essential oil in related fields such as medicine, food and cosmetics. [Results] Different extraction methods have an impact on the composition and content of chemical components in *Litsea cubeba* essential oil. At present, the main extraction methods for extraction of the essential oil from *Litsea cubeba* are steam distillation, supercritical carbon dioxide extraction, microwave assisted extraction and enzymatic extraction, while steam distillation is the most traditional one. The essential oil has many biological activities, including anti-oxidation, bacteriostasis, insecticidal and anti-inflammation, and can be also used to produce anti-microbial drugs, insecticides and antioxidants. It is widely used in medical, food and chemical industries and as also an important raw material for food preservatives. [Conclusion] Judging from the review of this article, *Litsea cubeba* essential oil has achieved positive effects in practical applications, including antibacterial, anti-inflammation, repellent and insecticide, and antioxidant activities with broad application fields and good development prospects. At the end of the article, the problems in the practical application of *Litsea cubeba* essential oil are briefly analyzed. In-depth exploration and research of establishing quality judgment standards, improving stability and developing a new feed additive of *Litsea cubeba* essential oil are needed to provide ideas for the researches on it and also promote the development and utilization of the essential oil from *Litsea cubeba*.

Keywords: *Litsea cubeba*; essential oil composition; functional activity; utilization; research progress

【研究意义】山苍子(*Litsea cubeba*(Lour.)pers.)是樟科木姜子属多年生灌木或小乔木,又名山鸡椒、木姜子、山胡椒、山姜子,中医药上又名毕澄茄^[1],主要分布在亚洲东部、大洋洲及太平洋诸岛等地。我国山苍子以野生为主,广泛分布于长江以南诸省区^[2-4],近年来江西、湖南、重庆、四川、贵州和福建等地有大规模人工培育。山苍子是我国重要木本精油芳香植物资源,其果实、叶和枝条等部位均含精油,实际生产主要从果实中蒸馏得到精油^[5]。山苍子精油因其重要的经济价值被广泛用于医疗、食品及化工行业,可作为食品防腐剂、保鲜剂的重要原料,具有广阔的应用领域和良好的发展前景。【前人研究进展】山苍子精油具有浓厚柠檬香味,主要成分是柠檬醛,呈淡黄色到橙黄色,是我国GB 2760—2014规定允许使用的天然食用香料,也是合成香料、食品添加剂及制药工业等的重要原料^[6-9]。山苍子精油具良好抑菌、抗氧化等生物活性,可用作抗微生物药物^[10-14]、杀虫剂^[15-16]和抗氧化剂^[17-18]等,具有良好的经济价值。【本研究切入点】我国山苍子资源丰富,而山苍子精油作为植物天然提取物,绿色环保,因其广谱的抑菌性备受关注。随着各种分离和检测技术的不断进步,山苍子精油的组成、结构和功能越来越被人们所认知,但还有许多方面需要深入探索和研究。【拟解决的关键问题】本文主要对山苍子的化学成分、提取方法、功能活性及其应用等方面的研究现状进行总结,旨在为山苍子精油的深度加工利用和产品开发提供参考。

1 山苍子精油提取及其主要成分研究

1.1 山苍子精油提取工艺

目前山苍子精油的提取工艺主要有水蒸气蒸馏法、超临界二氧化碳萃取法、微波辅助萃取法和酶法提取等。水蒸气蒸馏法是山苍子精油最传统的提纯方法,其工艺流程、设备、操作等方面都比较成熟,但得率低,热敏成分会发生分解及原料焦化。Ho等^[19]从台湾的山苍子叶和果提取精油,得率分别为13.9%、4.0%;Si等^[20]通过水蒸气蒸馏法从我国8个地区山苍子果实中提取的精油得率为3.1%~4.6%。

超临界CO₂流体萃取技术是目前最流行的超临界流体萃取法,其工艺简单、操作方便,无污染,超临界流体可重复使用,但设备比较昂贵,而且操作费用高,不适宜在大规模的生产中运用^[21-22]。张德权等^[23]就超临界CO₂流体技术萃取山苍子精油的工艺条件进行了探讨,最适工艺条件为:山苍子粉粒度60~80目、萃取压力25 MPa、萃取温度45℃、CO₂流量1.5 mL/min、萃取时间60 min,此条件下,并萃取出了高质量的富含山苍子精油和核仁油的山苍子油,山苍子的萃取率为30.19%。

有研究发现利用微波的穿透性来提高山苍子精油的萃取率,Yang等^[24]在水汽蒸馏的基础上,将山苍子烘干后通过微波超声辅助萃取法提取精油,最佳提取工艺为:提取时间8 min,微波功率600 W,料液比1:7 g/mL,提取温度85℃,山苍子精油的平均产量可达14.188%。刘晓庚等^[25]采用微波辅助提取山苍子精油,所得精油得率为5.5%~8.6%,与水蒸气蒸馏法相比,精油得率提高了2.48%。李文爽等^[26]采用超声波辅助提取山苍子精油,液固比18:1(mL/g)、超声时间12 min,提取时间3.4 h、提取率为5.33%。超声波和微波能有效地提高山苍子精油的提取工艺。

谢永芳等^[27]通过重组膨胀素和纤维素酶法,对山苍子破壁处理后再常压提取,结果表明复合酶法提取山苍子精油比单纯的纤维素酶法提取、蒸馏法提取要效率高。酶法提取是选用合适的酶将植物中的杂质(如淀粉、果胶、蛋白质)除去,酶特异性强、高度催化效能、反应条件温和,有利于油料饼粕的后续利用,但酶法提取存在酶的价格高,用量大等问题^[28],利用酶法提取山苍子精油的工艺研究还不够深入和成熟。

1.2 山苍子精油化学成分组成

山苍子精油化学成分复杂,主要是萜类化合物及其衍生物(图1),柠檬醛是其主要成分,但其化学成分的具体种类及含量存在着较大差异,且与提取部位、品种和产地等密切相关。

Wang等^[29]对山苍子根、茎、叶、花蕾、花、果实等挥发油成分进行了分析,结果显示根油主要含橙花醛(21.53%)、香茅醛(8.57%)、芳樟醇(7.45%)、异胡薄荷醇(6.05%)和 β -水芹烯(5.24%);茎油主要含 β -水芹

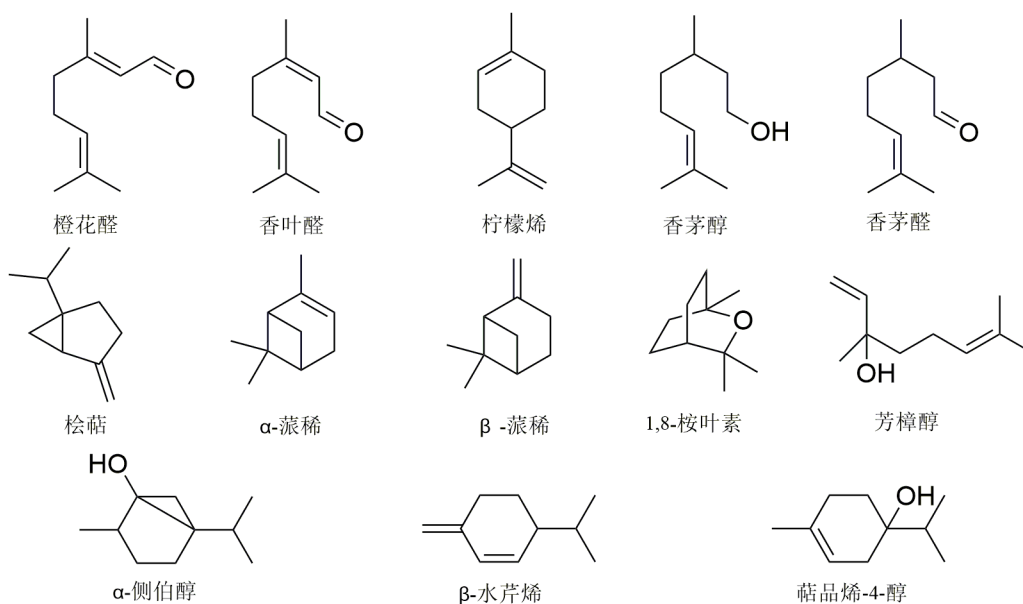


图1 山苍子精油中主要成分的化学结构

Fig.1 Chemical structures of the most predominant substances identified in *Litsea cubeba* essential oils

烯(18.74%)、萜品烯-4-醇(12.05%)、柠檬烯(9.82%)、 α -侧柏醇(9.43%)和 β -蒎烯(6.94%);叶油主要含桉叶素(13.97%)、 γ -榄香烯(8.27%)、石竹烯(8.04%)、芳樟醇(6.94%)、柠檬烯(6.78%);花蕾油主要含 β -水芹烯(33.74%)、桉叶素(11.24%)、 α -蒎烯(8.95%)和 β -蒎烯(8.33%);花油主要含 β -松油烯、桉叶素、 α -蒎烯和 β -蒎烯,含量分别为33.17%、13.68%、7.51%和7.25%;果油主要含橙花醛(63.75%)和柠檬烯(7.38%)。

山苍子精油的成分和含量随产地的影响而显著变化,Si等^[20]测定了我国8个地区采收的果实中山苍子精油化学成分,主要成分为单萜类化合物(94.4%~98.4%),橙花醛和香叶醛(78.7~87.4%),D-柠檬烯(0.7~5.3%)。Ho等^[19]从产于台湾的山苍子叶精油中共鉴定出53个化合物,主要成分为1,8-桉叶素(57.6%),桉烯(12.3%)、 α -乙酸松油酯(9.8%)、 α -蒎烯(4.1%)和 β -蒎烯(3.2%);此外,Hai等^[30]测定了越南6个省份的山苍子叶精油,4个产地山苍子叶油除了以1,8-桉叶素(23.4%~51.7%)为主要成分,还发现2个产地的山苍子叶油分别富含芳樟醇(91.1%)和桉烯(48.1%)。

此外,影响山苍子精油化学成分的因素还有很多,采收期、成熟度、采收后的处理方法和提取方法等均会影响山苍子精油的组成和含量^[5,31-33]。

2 山苍子精油功能活性研究

2.1 山苍子精油抑菌活性

山苍子精油具有较为广谱的抑菌活性,对细菌和真菌都具一定抑制作用,包括食品腐败菌和食源性致病菌。有报道山苍子精油对大肠杆菌、金黄色葡萄球菌、白曲霉、青霉4种菌的最低抑菌浓度分别为1.88,0.94,0.94,0.47 $\mu\text{L/mL}$ ^[34]。Hu等^[35]研究了山苍子精油对耐甲氧西林金黄色葡萄球菌的抑菌作用及其机制,最低抑菌浓度和最小杀菌浓度分别为0.5 mg/mL和1 mg/mL;山苍子精油对耐甲氧西林金黄色葡萄球菌的细胞膜有破坏作用,使细胞内生物大分子泄漏,另外主要成分柠檬醛还可与DNA发生嵌合。Li等^[36]研究发现,山苍子精油对甘草中黄曲霉菌的最低抑菌浓度为0.5 $\mu\text{L/mL}$,最低杀菌浓度为1.0 $\mu\text{L/mL}$,对3株产毒黄曲霉菌株具有较高的毒性,对黄曲霉毒素B1在甘草中的积累具有较强的抑制作用。Luo等^[37]从细胞膜、细胞器和细胞内大分子水平研究了柠檬醛对黄曲霉孢子的影响,结果表明,柠檬醛通过损伤黄曲霉孢子质膜,随后进入细胞,不仅影响线粒体的遗传表达和形态,而且改变类蛋白大分子的聚集;细胞、细胞器和大分子失去了它们正常的结构和功能,最终导致黄曲霉孢子的萌发能力丧失。

山苍子精油对医药领域的念珠菌有抑制效果,方芳等^[38]研究了山苍子精油对5种医学常见念珠菌的抑菌效果,其最小抑菌浓度分别为:白念株菌(14.14 $\pm$ 3.64) $\mu\text{g/mL}$ 、热带念珠菌(23.22 $\pm$ 2.85) $\mu\text{g/mL}$ 、光滑念珠菌(31.24 $\pm$ 22.88) $\mu\text{g/mL}$ 、近平滑念珠菌(76.194 $\pm$ 4.40) $\mu\text{g/mL}$ 、克柔念珠菌(28.30 $\pm$ 2.54) $\mu\text{g/mL}$;通过电镜观察发现,山苍子精油处理组克柔念珠菌细胞壁被溶解破坏,细胞膜被连续性破坏,细胞器肿胀溶解乃至坏死溶解。此外,曾跃斌等^[39]研究发现山苍子精油对白念珠菌全基因组表达谱的影响比较大,表达差异的基因数量约占总基因数量的11%。

此外,山苍子精油对植物病菌也有抑制效果。国颖等^[40]研究发现柠檬醛及其衍生物对油茶炭疽病菌有一定的抑制作用,800.00 mg/mL的天然柠檬醛的抑菌率为88.19%。Yang等^[41]研究发现,对西藏采集的山苍子果实提取物中鉴定出33个化合物,其中化合物1(2,6-二甲基-6-羟基-2E,4E-庚-2,4-二烯酸)和化合物2(6R-3,7-二甲基-7-羟基-2-辛烯-6-内酯)在588和272 $\mu\text{mol/L}$ 的浓度下对核盘菌、黄瓜枯萎病菌、假孢霉菌和炭疽病菌均有较好的杀菌活性,山苍子精油对黄瓜枯萎病菌和核盘菌有较好的杀菌活性, IC_{50} 分别为115.58 $\mu\text{g/mL}$ 和151.25 $\mu\text{g/mL}$ 。山苍子精油对辣椒疫霉的4个发育阶段均有抑制作用,对孢子囊的形成抑制作用最强,能破坏细胞膜的通透性^[42]。

2.2 山苍子精油的驱避和杀虫活性

山苍子精油对害虫具有不同程度的引诱、驱避、拒食、毒杀和抑制生长发育活性,山苍子精油中含有 β -水芹烯、柠檬烯、桉油精、 β -蒎烯、 α -蒎烯、松油烯、(-)-4-萜品醇、松油醇、乙酸松油酯,这些物质多为萜烯类物质及其氧化物均具有一定的驱虫活性^[43]。Amer等^[15]研究了41种植物精油和11种精油混合物对埃及伊蚊、斯氏按蚊和库蚊的保护时间和驱避活性,发现山苍子精油是5种最有效的精油之一。Jiang等^[44]研究发现山苍子精油及对粉纹夜蛾有接触毒性, LD_{50} 为112.5 $\mu\text{g/larva}$,其中 γ -萜品烯是起主要作用的功效成分。此外,Jeon等^[45]研究发现山苍子精油对粉尘螨、蕨菜夜蛾和腐霉病菌的抗性比苯甲酸苄酯

更有效,LD₅₀值分别为1.54,1.83,3.90 μg/cm²,分别是苯甲酸苄酯LD₅₀(9.84,9.16,12.01 μg/cm²)的6.39,5.01和3.08倍,具有开发成熏蒸剂的潜力。

2.3 山苍子精油的抗炎活性

山苍子精油具有一定的抗炎抗癌活性,钟艳梅等^[46]以山苍子精油为材料,采用小鼠巨噬细胞Raw264.7和小鼠肝癌细胞Hepa 1c1c7为模型,以一氧化氮抑制率为抗炎指标、醌还原酶活性为抗癌指标研究山苍子精油的抗炎抗癌活性,结果表明山苍子精油对一氧化氮抑制率≥50%的浓度为31.25~62.50 μg/mL,诱导醌还原酶活性倍增的浓度为15.63~31.25 μg/mL。山苍子精油可降低脂多糖(LPS)刺激的树突细胞中TNF-α和细胞因子IL-12的产生^[47]。Liao等^[48]从山苍子果实精油中分离出橙花醛和香叶醛,发现橙花醛具有较强的抗炎活性,可显著抑制LPS刺激的巨噬细胞分泌细胞因子(TNF-α、IL-6和IL-1β)和炎症分子的表达(Pro-IL-1β、iNOS、GOX-2和NLRP-3)。

2.4 山苍子精油的抗氧化作用

有研究表明山苍子精油具有较好的抗氧化活性,可作为一种易获得的天然抗氧化剂来源。李欣欣等^[43]测定了山苍子雌花和雄花精油的体外抗氧化活性,测定还原力的IC₅₀值雄花为2.330 mg/mL,雌花为1.473 mg/mL;清除DPPH自由基IC₅₀值雄花为41.62 mg/mL,雌花为9.663 mg/mL;清除羟自由基IC₅₀值雄花为56.95 mg/mL,雌花为77.98 mg/mL。Wang等^[17]发现山苍子精油的主要成分柠檬醛在抗氧化活性和清除自由基方面起着重要作用;苍子精油抗氧化活性高于柠檬醛,猜测这可能是因为精油是多种萜类和烯烃的混合物,成分间存在相互作用。

3 山苍子精油应用的研究

3.1 山苍子精油在医学中的应用

山苍子全株均可入药,具有温中散寒、理气止痛的功效,在传统医药上可用于治疗因胃寒所致的呃逆呕吐、脘腹疼痛等,以及风寒感冒、咳嗽气喘、消化不良等症^[49-50]。常在丽等^[51]研究发现,山苍子精油对絮状表皮癣菌、红色毛癣菌、石膏样毛癣菌、犬小孢子菌4种皮肤癣菌均有抑制作用,制成含山苍子精油3%霜剂应用于临床浅部真菌病的治疗,在26例患者中,有22例痊愈,3例好转,1例无效,所有病例均无明显不良反应,总有效率达96%,尤其对水疱型、浸渍糜烂型足癣疗效显著。殷志勇等^[52]发现柠檬醛气雾剂具有一定的平喘、镇咳和祛痰作用,具有良好的支气管解痉作用;柠檬醛气雾剂对乙酰胆碱-组胺所致的豚鼠哮喘有明显的平喘作用,能极显著性地延长豚鼠哮喘潜伏期,同时可以抑制ACh引起的豚鼠气管平滑肌收缩,又能使ACh收缩的豚鼠气管平滑肌累积量效曲线右移,表明其平喘机理与其竞争性拮抗气管平滑肌上的M受体有关,从而抑制了胆碱引起的平滑肌收缩。此外,有研究发现山苍子精油对人肺癌、肝癌和口腔癌细胞有细胞毒性^[19,53]。

3.2 山苍子精油在食品中的应用

山苍子鲜果被产区居民直接用作食品,将鲜果捣烂作为调味品,用于汤、面食、豆花、凉粉、米线等烹调食品中,可作为风味独特的天然调味香料。山苍子精油是我国允许使用的食用香料,目前常用于合成紫罗兰酮类、柠檬腈、香叶醇、香茅醇、橙花醇等香精香料、制药以及作为食品添加剂等^[54]。任彩萍等^[55]制备了一种用于调味的山茶油山苍子食用香油组合物,用山苍子精油作为特色调味油料形成独特的口味,不仅有利于增强食欲、促进消化,而且还有一定的保健价值。

天然抑菌剂的应用可有效防止果蔬因病菌污染而造成的腐烂损失,旷春桃等^[56]将山苍子油、姜黄油和桉叶油的组合物用于防腐保鲜,能明显延长肉类和果蔬的货架期。彭湘莲等^[57]以山苍子精油为主涂膜材料,添加壳聚糖、丙三醇等成膜助剂,配成复合涂膜液,发现山苍子精油壳聚糖复合涂膜抑制了金柑的失重,处理组的可溶性固形物、可滴定酸、VC、总糖、感官均优于对照组,可见山苍子精油壳聚糖复合涂膜降低了营养物质的消耗,有效地延缓了金柑的衰老。将山苍子果实精油制成无菌涂膜剂,山苍子油对虾肉有较好的保鲜作用,其最佳添加浓度为0.3%,添加0.1%植酸和0.1%VE对山苍子油(0~4℃贮藏温度)保鲜效果有增效作用,使货架期明显延长,能够有效抑制虾肉黑变,减缓挥发性盐基氮、三甲胺和细菌总数等的产生速率,抑制pH值的升高^[58]。

山苍子精油对人畜无毒无副作用、无环境污染,又具有愉悦的香气,它是大米、花生、玉米、小麦等粮

食作物储藏的优良防治霉菌和虫害的天然药剂。赵阔等^[59]通过模拟花生仁仓储条件,熏蒸山苍子精油,熏蒸处理后的花生仁色泽气味、含油量、蛋白质含量等理化指标与熏蒸处理前相差不大,脂肪酸值略有上升,但各项指标均明显优于对照组。

3.3 山苍子精油在化妆品中的应用

近年来,消费者越来越追求天然和无添加的化妆品,由天然植物萃取而得的植物精油在抗菌、消炎、抗氧化等方面具有突出的生物活性。山苍子精油广泛应用于化妆品中和中国南方的民间护肤品。Huang等^[60]研究发现山苍子精油对酪氨酸酶有很强的抑制作用,具有良好的抗氧化活性以及对紫外线-二氧化钛-二氧化氮的-诱导蛋白和酪氨酸硝化有明显保护作用,并建议山苍子精油可以作为一种新的天然皮肤增白剂。周晓浩^[61]制备了一种含山苍子精油的雪花膏,使用后明显感到舒适、柔软,无油腻感,具有消炎抗菌、温和调节肌肤,对皮肤具有良好的持续补水保湿、嫩肤养颜的效果。此外,还可将山苍子油加入洗手液中,能够有效清除手上的油污、细菌、同时还具有抗菌作用^[62]。

3.4 山苍子精油在其他方面的应用

山苍子精油具有较好的杀虫、抗菌、防腐等功能活性,可研制杀虫和抗病菌的药剂。陈善千等^[63]制备了含有山苍子精油和40%氧化乐果乳剂,该组合物用作防治危害柑桔的矢尖蚧的药剂,高效低毒,可达到全年一次性防治的良好效果。山苍子精油有防霉和脱霉的作用,将其添加到饲料中,有柠檬醛的特殊气味,可作为饲料风味剂,刺激动物食欲,提高采食量^[64]。此外,祝盆鑫等^[65]将山苍子精油与混合植物浆料配合,制得一种内墙乳胶漆,增强了抗菌效果,而且气味清新,可改善室内空气。

4 展 望

综上所述,山苍子精油作为植物天然提取物,绿色环保,具有较好的驱避、杀虫、抗菌、和抗氧化等多项生物活性,被广泛应用于食品、医药、化妆品等方面。随着各种分离和检测技术的不断进步,山苍子精油的组成、结构和功能越来越被人们所认知,但还有许多方面需要深入探索和研究。(1)山苍子精油的成分和含量还受到产地、提取部位和采收期等影响,需建立一套山苍子精油组分的标准指纹图谱,为其识别和质量判断提供一定依据。(2)山苍子精油在应用于食品、医药和化妆品方面,完善其作用机制研究,建立体外和体内毒理实验,进行安全评价。(3)山苍子精油水溶性较差、热稳定性不佳、易挥发、易氧化,贮存过程中容易变质等方面的缺陷限制了其应用,可通过包埋技术包埋山苍子精油,提高山苍子精油的水溶性、稳定性以及有效控制香气的释放和延长抑菌功效,提高在医学方面的功能活性,避免山苍子精油与食品直接接触,降低对食品风味和品质的影响。(4)山苍子精油具有开发为新型饲料防霉、脱毒剂的潜力,增强动物的免疫力,减少抗生素的使用,有潜力成为一种新型饲料添加剂,但此方面的研究较少,可进一步深入研究其机理。

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