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静电纺丝技术包埋姜黄素研究进展

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摘要　静电纺丝作为一种制备纳米纤维的技术越来越受到关注。近年，大量研究利用静电纺丝技术制备含姜黄素的纳米纤维和纳米颗粒。本文介绍静电纺丝的原理和影响因素，以及利用静电喷雾制备包埋姜黄素纳米颗粒的情况。总结姜黄素纳米纤维的制备方法并根据制备纳米纤维的原料（合成高分子和天然高分子）进行分类讨论。论述姜黄素包封的纳米纤维在缓释体系、组织工程、活性包装和微纳米传感器方面的应用。

关键词　静电纺丝；姜黄素；纳米纤维
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静电纺丝技术是一种简单易操作的用于制备纳米级和微米级纳米纤维的技术，最早于 1934 年被 Anton[1]提出。越来越多的研究者将静电纺丝应用到不同的领域，比如组织工程[2]、能源储存转换[3]、食品包装[4]、药物载运释放[5]、催化[6]、传感器[7]和过滤[8]等。食品领域主要将静电纺丝（电纺）纳米纤维应用于生物活性物质（如姜黄素）包埋载运，进而应用于食品活性包装[9]。姜黄素是一种提取于姜科、天南星科植物根茎中的二酮类化合物，可作为一种食品天然色素（图 1）。其最主要的姜黄素类化合物（curcuminoid），约占姜黄色素的 70%，约为姜黄的 3%~6%。其中姜黄素（curcumin）是主要的活性物质，已有研究表明姜黄素具有良好的抗氧化性、抑菌性、抗炎性、抗肿瘤、抗病毒等效果。但是由于姜黄素水溶性差，因此其生物利用度不高，因此许多研究通过包埋的方式以提升其在食品体系的溶解性[10]。静电纺丝技术作为一种微纳米载运体系的制备技术也被许多研究者应用于姜黄素载运缓释，并且可以提升姜黄素在体内和体外的生物利用度。本文综述利用静电纺丝技术包埋姜黄素的相关研究。

1　静电纺丝概述

静电纺丝（Electrospinning）技术，是指利用静电作用力将高分子聚合物转变成微纳米级超细纤维的一种技术。图 2 所示为实验室最普遍的溶液静电纺丝设备，主要包括高压电源、流量泵、注射器及针头、接收端。在直流高压静电场下，当高聚物溶液以一定流速被挤出注射器针头，针尖液滴会向最近的低电势点方向伸展，从而形成泰勒锥（Taylor Cone）结构。当电场产生的静电作用力克服泰勒锥尖端液滴的表面张力时，就会喷射出一股带电高聚物的细流。受静电作用力、库伦斥力、表面张力、流体黏弹力等影响，带电射流进一步加速拉伸并呈螺旋摆动，使得溶剂快速挥发，高分子聚合物从而形成连续超细纤维形态，被收集于接收端上。

影响静电纺丝纳米纤维形成的参数主要有以下几个方面：1）溶液参数；2）电纺过程参数；3）环境参数。溶液参数主要包括高分子相对分子质量、溶液浓度和黏度、溶剂种类和溶液导电能力。通常高分子的分子质量越大或者黏度越高越有利于纳米纤维的形成，但是分子质量过大和黏度过高易导致纤维直径增大，甚至挤出困难而电纺不成功。不同溶剂的组合能够改变聚合物分子链溶胀和缠结状态，从而对静电纺丝纤维形貌产生影响。改变溶液的电导率对电纺的影响是两方面的，一方面是改变了库仑力，另一方面改变了静电作用力。电纺工艺参数方面包括电纺电压、流速和针尖接收

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图 1　姜黄(左),姜黄素商品(中),姜黄素化学结构(右)

Fig.1　Turmeric (left), curcumin powder (medium), chemical structure of curcumin (right)

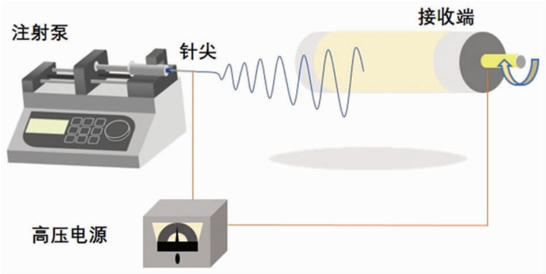

图 2　静电纺丝设备示意图

Fig.2　Illustration of the electrospinning set up

距离等参数。通常针对某一特定溶液超过某个临界电压即能够形成泰勒锥，随着电压增加纤维直径通常会降低。合适的流速对于纳米纤维的形成至关重要,流速过小不足以形成泰勒锥,流速过大又容易形成液滴堆积。环境参数方面,温度的升高对电纺过程最直接的影响是降低了高分子溶液的黏度,加速分子的运动,有利于降低纳米纤维的直径，这一点对于实现天然高分子水溶液的静电纺丝十分有利。越来越多的研究发现湿度对于控制纳米纤维形成过程中的固化和牵伸也十分重要。

通过静电纺丝能够得到各种形态的纳米纤维,最常见的为无序的纤维状,通过改变参数和设备的组件(图 3)也可以形成多孔状、定向型、蛛网状、树状等形态的纳米纤维。通过核壳结构的针头能够获得核壳纳米纤维，将姜黄素包埋于纳米纤维中,实现长时间的释放。通过气流辅助的针头可以实现超细纳米纤维的制备并提高产率。常规的纳米纤维的针尖接收距离在 5~25 cm，当针尖接收距离减小到 500 μm~5 cm 之间为近场静电纺丝。近场静电纺丝的过程中避免了溶液喷射后的鞭动运动,从而能够获得直写式的结构效果,做到静电纺丝和 3D 打印的结合。静电溶吹技术是利用较大针尖接收距离的一种方式,可以达到 80~100 cm[11-12],通过气流辅助电纺溶液的挥发及提高电纺的产率。姜黄素纳米纤维制备的相关研究大部分采用的是溶液静电纺丝的方式，接收端为平板或者接地转鼓。

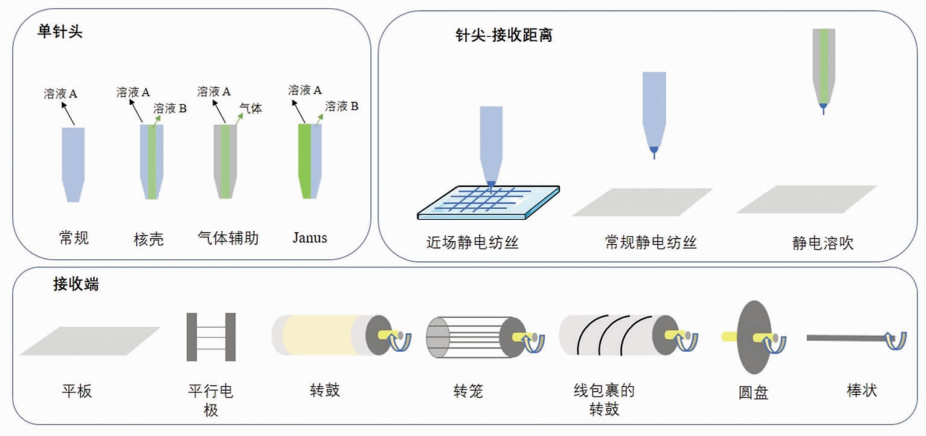

图 3　静电纺丝设备分类

Fig.3　The classifications of electrospinning set ups

2　静电喷雾包埋姜黄素

静电喷雾技术与静电纺丝过程类似，通过改变静电纺丝参数,如降低高分子浓度,形成纳米颗粒的过程。通过静电喷雾得到的纳米颗粒可以避

免如喷雾干燥过程中的高温条件，保留生物活性物质的功能，并且达到较高的包封率。表1列举了近年利用静电喷雾技术制备纳米颗粒包埋姜黄素的研究，其包封率都在85%以上，甚至有的研究报道接近100%。姜黄素的添加量一般为相比于干物质的5%~15%（质量分数）之间。静电喷雾得到的姜黄素纳米颗粒直径在微米级。

表1 通过静电喷雾技术制备姜黄素纳米颗粒相关研究

Table 1 Relevant researches about fabrication of curcumin nano-particles by electro-spraying

高分子	溶剂	高分子质量分数	姜黄素质量分数**/%	电纺参数	平均直径	包封率/%	参考文献
玉米醇溶蛋白	80%乙醇	1%~20%	10	14 kV,0.15 mL/h,7 cm		85~90	[13]
明胶	水/乙醇/乙酸	5%	10	14 kV,0.15 mL/h,10 cm	0.15~0.5 μm	100	[14],[15]
聚羟基乙酸	壳层:乙酸乙酯 核层:丙酮	壳层：10% 核层：1%	核层:4		2.07~5.92 μm	89.9~95.4	[16]
聚乳酸	丙酮,DCM,THF,TCM*	1%,3%,5%,7%	0,5,10,15	20 kV,15 cm,0.1~0.7 mL/h	3.8~4.4 μm	95%	[17]

注：* DCM：二氯甲烷；THF：四氢呋喃；TCM：三氯甲烷；** 相比于高分子质量分数。

3 姜黄素纳米纤维

3.1 电纺方式

制备姜黄素纳米纤维的方式可主要分为混合电纺、核壳电纺和乳液电纺的方式（图4）。混合电纺是直接将姜黄素溶解于电纺溶液中，进行静电纺丝得到含姜黄素的纳米纤维。核壳电纺是将姜黄素溶解于电纺芯层溶液中，通过核壳电纺即可得到姜黄素分布于芯层的纳米纤维。Aytac等[18]以聚乳酸溶液为壳层，环状糊精溶液为芯层，姜黄素包封于芯层中进行静电纺丝，所得到的纳米纤维表现出良好的水溶性和缓释效果。乳液电纺是近年新型的一种电纺方式，通过在电纺溶液中制备油包水或者水包油乳液实现亲水或疏水功能性成分的载运，通过控制电纺参数还可能得到核壳结构的纳米纤维[19]。如Deng等[20]利用阴离子型、阳离子型和非离子型表面活性剂包埋姜黄素后溶解于明胶溶液中进行电纺，增加了姜黄素的溶解性，有利于其功能性的表达。

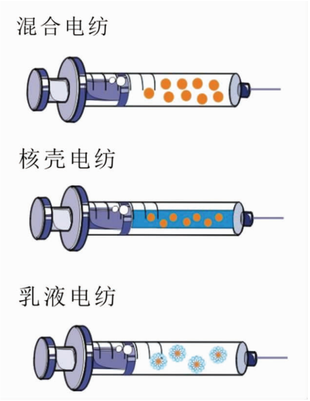

图4 含姜黄素纳米纤维制备方式：混合电纺、核壳电纺、乳液电纺

Fig.4 Various methods to fabricate curcumin encapsulated nanofibers: hybrid, core-shell, and emulsion electrospinning

3.2 电纺原料

3.2.1 合成高分子 表2为利用合成高分子制备姜黄素纳米纤维的相关研究的电纺方式，纳米纤维平均直径及其应用方向。主要应用于姜黄素包埋的合成高分子材料有聚己内酯（PCL）、聚氧乙烯（PEO）、聚乳酸（PLA）、聚乙二醇（PEG）、聚羟基乙酸（PLGA）、聚乙烯吡咯烷酮（PVP）。通过合成高分子制备得到的姜黄素纳米纤维具有相对较好的机械性能，因此大多应用于组织工程领域，起到愈伤修复、抗肿瘤、抗炎等效果。

3.2.2 天然高分子 表3列举了利用天然高分子或者天然高分子/合成高分子混合体系包埋姜黄素的纳米纤维直径、包封率和潜在应用。蛋白类主要有明胶、玉米醇溶蛋白、丝素蛋白等，多糖类主

表2 合成高分子-姜黄素纳米纤维制备相关研究

Table 2 Relevant researches about fabrication of curcumin encapsulated synthetic polymer nanofibers

高分子	电纺方式	平均直径	应用	参考文献
PLGA	混合电纺	100~300 nm	抗癌	[21]
PCL	混合电纺	620 nm	抗癌	[22]
PEO/PLA	混合电纺	588~703 nm	抗癌	[23]
聚甲基丙烯酸-2-羟乙酯	混合电纺	20~110 nm	愈伤修复	[24]
PVP	混合电纺	485 nm	抗肿瘤	[25]
PVP	乳液电纺	500 nm~2.84 μm	缓释	[26]
PLA，PLA/PVP，PLA/PEG	混合电纺	1 800 nm	抗凝血	[27]
壳:PLA 核:环状糊精	核壳电纺	695 nm	抗氧化	[18]
PVP	混合电纺		抗凝血	[28]
PLGA	混合电纺	400 nm	愈伤修复	[29]
PLA	层层叠加	900~1 500 nm	缓释	[30]
聚二恶烷酮	混合电纺	1.1~1.8 μm	组织工程	[31]
壳层:PVA 核层:PCL	核壳电纺	156~205 nm	组织工程	[32]
聚醚砜(polyethersulfone)	混合电纺	0.5~2.5μm	缓释	[33]
PCL	乳液电纺	20~550 nm	缓释	[34]
Eudragit RS 100	混合电纺	-	抑菌	[35]
壳:PLA 核:PVA	混合电纺	2 800 nm	缓释	[36]
PEG-PLA-PCL共聚物	纱线电纺	172~889 nm	缓释	[37]
PCL/copolymer F-108	混合电纺	613~800 nm	愈伤修复	[38]

要由乙基纤维素、壳聚糖、透明质酸、海藻酸钠、黄芪胶等。已有研究中测得的姜黄素包封率大都超过90%，理论上来说静电纺丝的过程是一个无物质损耗的过程，即使制备过程中有液滴损耗也不会改变固形物的比例，因此研究中出现的包封率的差异也可以归因于包封率测定方法的不同。有的研究者会将纳米纤维先进行冲洗，将附着于纤维表面的姜黄素去除掉再进行包封率测定，因此得到相对较低的包封率值。

表3 天然高分子-姜黄素纳米纤维制备相关研究

Table 3 Relevant researches about fabrication of curcumin encapsulated biopolymer nanofibers

高分子	平均直径/nm	包封率/%	应用	参考文献
乙基纤维素	314~340	90.8~100	组织工程	[39]，[40]
玉米醇溶蛋白	231~297	77.5~83.2	组织工程	[41]
壳聚糖/PLA	84~232		愈伤修复	[42]
玉米醇溶蛋白	438~543		抑菌	[43]
丝素蛋白	50~300		缓释	[44]
丝素蛋白/P(LLA-CL)	293~461		抑菌	[45]
玉米醇溶蛋白	482~586		纳米传感	[46]
苋菜分离蛋白-普鲁兰多糖	134~248	72.6~93.3	缓释	[47]
明胶/PCL	123~133		抑菌	[48]
黄芪胶/PCL	164~191		愈伤修复	[49]，[50]
乙基纤维素	104		纳米传感	[51]
玉米醇溶蛋白	-	82.4~86.7	活性包装	[52]

(续表3)

高分子	平均直径/nm	包封率/%	应用	参考文献
明胶	–	–	愈伤修复	[53]
明胶	368	94.6~95.8	缓释	[20]
丝素蛋白	982~1 297	–	缓释	[54]
黄芪胶/PVA	181~216	–	组织工程	[55]
乙基纤维素/PVP	1 528	–	抑菌	[56]
玉米醇溶蛋白	615~785	96.89~98.67	抑菌	[57]
明胶/PVA	200~250	–	经皮给药	[58]
扁桃胶/PVA	127~169	92~95	提升生物利用度	[59], [60]
乙基纤维素	780	–	抑菌	[61]
谷朊蛋白	258~375	80.7~84.8	抑菌和抗氧化	[62]
明胶,玉米醇溶蛋白	230,360	86~89	活性包装	[63]
黄原胶/壳聚糖	750~900	69.4	提升生物利用度	[64]
壳聚糖/PVA	200~1 250	–	组织工程	[65], [66]
魔芋葡聚甘露糖/玉米醇溶蛋白	–	–	抑菌和抗氧化	[67]
丝素蛋白	100~200	–	缓释	[68]
环状糊精	220~165	98.8~99.3	口腔快速给药	[69]
海藻酸钠/PEO	212~250	–	–	[70]
壳聚糖/胶原蛋白/PVA	–	–	组织工程	[71]
透明质酸	149~799	–	–	[72]
明胶/PCL	1 548	–	愈伤修复	[73]

4 姜黄素纳米纤维应用

4.1 缓释体系

通过静电纺丝技术将姜黄素包埋后所要达到的主要目的是缓释，主要分为体外缓释和体内缓释。将姜黄素的抑菌和抗氧化功能赋予某些包装材料后，缓释的效果能够使其在较长的时间发挥功效。体内释放分为食品体系和药品体系。当姜黄素作为一种功能性成分被食用时，缓释的效果能够使其最大限度的达到小肠以被吸收，甚至到达大肠起到肠道菌群调节的作用。当姜黄素作为一种药品时，缓释效果能够使其靶向到达要作用的位点，起到抗炎、抗肿瘤的效果。

4.2 组织工程

姜黄素由于具有抗炎、抗肿瘤和抗凝血等功效被广泛研究应用于愈伤敷料和组织工程支架制备。Chen 等[74]发现 PLA/姜黄素纳米纤维膜延长了血液凝固时间，并且随着姜黄素浓度的提高血液凝固时间延长。抗凝血活性的材料有利于提升组织工程血管移植成功的机会。Ramalingam 等[24]制备了含姜黄素的聚(二羟基乙基甲基丙烯酸酯)纳米纤维，表现出对耐甲氧西林金黄色葡萄球菌(MRSA)和超广谱 B 内酰胺酶(ESBL)的抑菌性，可作为多种耐药菌引起的传染病伤口愈合的贴剂。

4.3 活性包装

活性包装是指在传统包装中添加抗氧化剂、抑菌剂、指示剂等,使得包装具有抑菌抗氧化、吸湿、缓释香气、吸收异味等功能特性。静电纺丝是制备活性包装极具前景的一种技术。纳米纤维能够为包装提供纳米级的反应空间，巨大的比表面积能够大幅提高感应物质的精度和速率。姜黄素作为一种既具有抗氧化性又具有抑菌性的分子具有应用于食品活性包装的潜力。Yilmaz 等[52]制备了玉米醇溶蛋白/姜黄素纳米纤维膜并将其直接电纺到接种了灰霉和扩展青霉的苹果上，发现玉米醇溶蛋白/姜黄素能够显著延缓苹果的腐烂。

4.4 微纳米传感器

姜黄素的结构在酸性和碱性环境下会发生酮

和烯醇式互变异构。其在酸性条件下呈黄色，碱性条件下呈红色，因此可以作为pH指示剂和金属络合剂。姜黄素对铁离子有非常好的结合能力，被Saithongdee等[46]应用于制备铁离子浓度测定试纸。其研究结果表明，用质量分数5%的姜黄素/玉米醇溶蛋白制备的膜在pH值为2的溶液中进行Fe^{3+}的测定，显示出良好的颜色变化，且无明显干扰。其光学检测限为0.4 mg/L，低于饮用水的最大可接受浓度。Raj等[51]制备了乙基纤维素/姜黄素纳米纤维膜用于水体中铅离子的检测，具有线性的色度变化。

5 结论与展望

已有的大量研究将姜黄素包埋于各种不同的高分子纳米纤维体系中，研究了纳米纤维膜物理化学性质的差异，对其抑菌性、抗氧化性、抗炎效果等进行了表征。通过改变纳米纤维的组成成分、形态结构和成分间的交联状态能够改变其包埋的姜黄素的释放行为及功能发挥。但是大部分的研究仅限于体外的生物活性测试。针对纳米纤维膜包埋功能性成分的体内研究也相对较少，因此探究姜黄素纳米纤维在体内的代谢过程及其生物利用率是未来的发展方向。

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Research Progress of Curcumin Entrapment by Electrospinning

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Abstract Electrospinning，as a nanofiber fabrication technology，has attracted more and more attention. A few literatures for fabrication of curcumin encapsulated nanofiber or nanoparticles has been reported in recent years. The review will introduce the principle and affecting factors of electrospinning and fabrication of curcumin encapsulated nanoparticles. The fabrication of curcumin encapsulated nanofibers will be summarized in the aspect of electrospinning methods and electrospinning materials (synthetic polymer and biopolymer). At last，the application of curcumin encapsulated nanofibers on controlled delivery，tissue engineering，active package，and nano sensor will be discussed.

Keywords electrospinning; curcumin; nanofiber