

生物活性材料修饰的聚醚醚酮在口腔种植领域的潜在应用

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摘要

口腔种植作为牙列缺损的重要修复方法之一, 目前在临床上得到广泛应用, 种植材料的优化对于种植成功具有重要意义。聚醚醚酮(polyetheretherketone, PEEK)是聚芳醚酮家族中的一种半结晶形态热塑性聚合物, 相比于传统的口腔种植材料, PEEK具有与骨组织相近的弹性模量、化学稳定性和射线可透性等一系列优异的性能, 能够避免金属腐蚀以及应力遮挡效应导致的骨吸收。然而, PEEK表面的生物惰性可能会导致骨结合不良以及感染等问题, 不利于其在口腔种植领域的应用。因此, PEEK的表面改性日益成为研究的热点之一。研究人员探索了多种生物活性材料的改性方法, 以有效增强PEEK及其复合材料的成骨和抗菌性。因此, 本文主要综述了近年来生物活性材料改性后的PEEK的研究进展, 对PEEK的成骨及抗菌策略进行了分析, 并对改性后的PEEK在口腔种植领域的应用进行了展望。

关键词

聚醚醚酮, 牙种植体, 表面修饰, 生物活性, 成骨, 抗菌, 弹性模量, 种植材料

The Potential Application of Polyetheretherketone Modified by Bioactive Materials in the Field of Oral Implantology

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Abstract

As one of the important repair methods for dentition defects, oral implant is widely used in clinic. The optimization of implant materials is of great significance to the success of implants. Polyetheretherketone (PEEK) is a semi-crystalline thermoplastic polymer in the polyaryletherketone family. Compared with traditional oral implant materials, PEEK has a series of excellent properties such as elastic modulus, chemical stability and ray permeability similar to bone tissue, which can avoid metal corrosion and bone resorption caused by stress shielding effect. However, the biological inertia of PEEK surface may lead to problems such as poor osseointegration and infection, which is not conducive to its application in the field of oral implantology. Therefore, the surface modification of PEEK has increasingly become one of the research hotspots. Researchers have explored a variety of modification methods of bioactive materials to effectively enhance the osteogenic and antibacterial properties of PEEK and its composites. Therefore, this paper mainly reviews the research progress of PEEK modified by bioactive materials in recent years, analyzes the osteogenesis and antibacterial strategies of PEEK, and looks forward to the application of modified PEEK in the field of oral implantology.

Keywords

Polyetheretherketone (PEEK), Dental Implant, Surface Modification, Biological Activity, Osteogenesis, Antibacterial, Elastic Modulus, Implant Materials

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1. 引言

随着口腔种植技术的不断发展以及材料的不断优化，口腔种植作为牙列缺损的重要修复方法之一，因兼顾功能及美学等一系列优点，目前在临床上得到广泛应用。能否获得长期稳定的治疗效果是种植修复是否成功的关键。早期骨整合不良、感染以及植体应力疲劳导致的骨吸收等均会导致种植失败[1]。种植材料的优化对于种植成功具有重要意义，生产性能更佳、更加耐用的种植材料成为当前的研究重点。

PEEK 是聚芳醚酮家族中一种半结晶形态热塑性聚合物[2]，与传统植入物相比，其弹性模量(3~4 GPa)与人皮质骨(18 GPa)更为接近，并且还具有优异的耐高温性、抗疲劳性、抗腐蚀性、机械性能和射线可透性，因此被广泛应用于口腔领域[3]。但 PEEK 表面具有疏水性，导致细胞蛋白的黏附和骨传导能力较差，因此被归类为生物惰性材料，这导致了 PEEK 在植入领域受到了很大的限制[4]。

研究人员探索了多种 PEEK 改性方法，以有效增强 PEEK 表面的生物活性。主要为通过物理方法改变 PEEK 表面形貌或在其表面引入化学基团或生物活性材料以及与其他物质共同制备 PEEK 复合材料[5]。其中，具有特定而复杂成分和结构的生物活性材料可以在分子水平上靶向调节细胞的生理行为，构建有利于细胞生长、增殖和分化的微环境[6]，有效提高 PEEK 表面的生物活性，增加其成骨和抗菌作用。因此，本文主要综述了近年来不同功能以及不同类型的生物活性材料改性 PEEK 的研究进展，对改性 PEEK 的成骨及抗菌策略进行了分析，并对改性后的 PEEK 在口腔种植领域的应用进行了展望。

2. PEEK 生物活性材料修饰——生长因子

种植体周围骨结合的成功取决于生长因子的固定效率和递送行为，而生长因子的固定效率和递送行

为又受种植体表面特征的影响[7]。通过在 PEEK 表面引入不同类型的生长因子,提高诱导成骨分化和刺激新骨形成的能力,从而实现 PEEK 植入物与周围牙槽骨之间的骨整合。

骨形态发生蛋白(bone morphogenetic protein, BMP),尤其是骨形态发生蛋白-2 (bone morphogenetic protein-2, BMP-2),是目前用于骨再生中研究最多和最有潜力的骨诱导生长因子[8]。BMP-2 目前已被美国食品药品监督管理局批准用于临床[9]。研究人员将 PEEK 与羟基磷灰石复合物作为植入物,并将 BMP-2 负载于表面,结果证明 BMP-2 的负载导致植入区域骨结合效率显著增加[10]。然而,BMP-2 的半衰期较短,并且 BMP-2 在短时间内局部高释放量可能会激活破骨细胞,导致种植体周围骨结合能力变差,因此如何建立 BMP-2 的缓释体系具有重要意义[11]。研究人员利用明胶水凝胶构建 BMP-2 的缓释体系,并利用聚多巴胺(polydopamine, PDA)的粘结性将其修饰在 PEEK 表面,结果证明,修饰后的 PEEK 能够保证 BMP-2 的持续释放并具有促进干细胞成骨分化的能力[12]。BMP-2 还可以与其他物质共同作用于 PEEK 表面,协同促进骨结合。将胰岛素样生长因子-1 (insulin-like growth factor 1, IGF-1)和 BMP-2 双生长因子涂层固定在 PEEK 表面,与单独修饰 BMP-2 相比,可显著增强材料的生物活性[13]。并且,与单一生长因子相比,多种生长因子联合使用可以避免单一生长因子使用剂量过高而引起的种植体松动、组织肿胀和炎症等副作用[14]。Chubrik 等人将 BMP-2 和促红细胞生成素加载在 PEEK 上,能够提高骨和血管的生成能力[15]。此外,还可以利用 BMP-2 与肝素的结合能力,将 BMP-2 通过肝素固定在 PEEK 表面,能够实现与肝素协同增强细胞的增殖及成骨分化的生物活性[16]。

除 BMP-2 外,血小板衍生生长因子-BB (platelet-derived growth factor-BB, PDGF-BB)是 PDGF 家族的一种亚型,已被证明 PDGF-BB 可以通过刺激破骨细胞产生骨保护素来抑制破骨细胞的生成,增强骨再生能力[17]。在一项研究中,通过构建 PDGF-BB 与核晶素双生长因子的缓释微球,并利用 PDA 将微球加载在 PEEK 表面,结果表明,修饰后的 PEEK 支架具有良好的生物相容性,能够促进细胞迁移,增强骨髓间充质干细胞(bone marrow mesenchymal stem cells, BMSCs)的软骨形成分化,并在体内促进软骨再生[18]。

3. PEEK 生物活性材料修饰——细胞外基质

细胞外基质(extracellular matrix, ECM)是所有生物系统的基础,在动物 ECM 中,蛋白质(例如胶原蛋白、弹性蛋白)和蛋白多糖(与糖胺聚糖链共价连接的蛋白质)大量存在[19]。其中,胶原蛋白是人体中最普遍存在的蛋白质,因其生物相容性、生物降解性和细胞粘附性而广泛用于组织工程和再生医学[20]。通过 PDA 涂层将 I 型胶原蛋白固定在多孔 PEEK 表面上,可以提高 PEEK 的生物相容性,促进骨形成[21]。还有研究将具有甲基丙烯酰基的胶原蛋白通过表面引发的光聚合共价连接到 PEEK 表面,当人骨髓来源的间充质干细胞在胶原固定的 PEEK 表面上培养时,能够增强细胞的粘附和增殖,诱导成骨分化后,其表面的细胞显示出比 PEEK 表面更高的成骨细胞的矿化以及相关基因的表达[22]。

硫酸软骨素(chondroitin sulfate, CS)和透明质酸(hyaluronic acid, HA)等糖氨基聚糖是 ECM 的重要多功能成分,可通过聚集间充质细胞并支持其分化来刺激骨整合[23]。其中 CS 是一种存在于软骨、骨、皮肤、韧带和肌腱等结缔组织中的硫酸化糖氨基聚糖,能诱导成骨细胞粘附和分化[24]。有研究通过 PEEK 表面的仿生 PDA 涂层持续释放 CS 和左氧氟沙星来实现 PEEK 表面双功能化,显著改善了细胞生长和粘附、细胞活力和成骨分化,并有效抑制了大肠杆菌和金黄色葡萄球菌的生长[25]。将镁-硫酸软骨素复合物构建在磺化的 PEEK 表面,制备后的材料具有良好的亲水性和生物活性,能够促进骨和血管生成相关基因的表达,进而促进骨整合和血管生成[26]。

HA 是一种天然糖氨基聚糖,是影响细胞行为的重要细胞外基质,在生物医学领域有着广泛的应用[27]。Aso Abdulghafor 团队将 HA 涂覆在 PEEK 表面,表面涂层增强了 PEEK 植入材料的生物功能特性,改善了成骨细胞的增殖、分化和矿化潜力[28]。HA 还可以作为负载富血小板血浆(platelet-rich plasma, PRP)

和神经生长因子(nerve growth factor, NGF)的水凝胶涂层, 结果表明, PEEK 表面负载 PRP 和 NGF 的 HA 水凝胶涂层降解缓慢, 并且可以持续释放包括 NGF 在内的各种生长因子, 促进植入物的骨整合以及新骨和新生血管的生成[29]。

4. PEEK 生物活性材料修饰——肽

肽作为结构简单的氨基酸序列, 具有良好的生物学特性和安全性[30]。其中, 成骨肽、粘附肽以及一些融合肽等均被应用于 PEEK 的表面修饰。

将成骨多肽序列 BEE1 聚合并接枝到 HA/CF/PEEK 复合材料表面, 生成 HA/CF/PEEK-BEE1 复合材料。体外及体内实验表明, 成骨肽修饰的复合材料更加有助于细胞的黏附和增殖并且能更好地促进骨缺损处的骨矿化及新生[31]。玻连蛋白序列的粘附肽及其逆向倒置的二聚体结合了天然玻连蛋白的生物活性和稳定性, 将其构建在 PEEK 表面, 能够增强细胞的增殖能力, 并且具有更高的钙沉积[32]。定位在 BMP-2 生长因子上的序列可显著增强人成骨细胞的矿化和基因表达, 将其接枝在 3D 打印的 PEEK 表面, 生成的复合材料不仅被证明无任何细胞毒性, 还提高了细胞增殖速率和钙沉积量, 增强了成骨基因的表达[33]。除单独修饰外, 有课题组构思了一种多功能异质结构纳米涂层, 其中包含了多种金属元素以及促进骨和血管生成的融合肽, 将其修饰在 PEEK 表面能够协同促进细胞粘附和增殖, 并促进血管生成和成骨相关基因的表达, 从而在体外表现出优异的血管生成分化和成骨诱导能力, 进而改善骨整合[34]。

5. PEEK 在口腔修复领域的应用及展望

近年来, 由于 PEEK 材料性能的不断优化及各种生物复合材料的创新, PEEK 逐渐被应用于口腔修复的临床领域。

PEEK 在口腔修复治疗中应用广泛, 如活动义齿、固定义齿及种植体基台等。对于固定义齿来说, PEEK 的弹性模量为 3~4 GPa, 低于氧化锆和钴铬合金的弹性模量(220 GPa), 因此, PEEK 为基牙提供了更多的应力抵抗力, 防止基牙断裂, 并且与氧化锆相比, PEEK 需要的制造时间更少, 并且重量比氧化锆更轻[35]。

在种植治疗过程中, 首选的传统材料是钛及其合金, 然而由于钛的弹性模量(110 GPa)与骨的弹性模量(1~30 GPa)之间存在显著差异, 可能会导致牙槽骨负担过重, 进而导致骨吸收[36]。PEEK 作为一种植入材料, 具有良好的物理及机械性能, 并且由于弹性模量与骨组织更加相近, 能够优化咀嚼力在种植体周围的分布, 减少应力集中导致的骨吸收。近年来, 关于 PEEK 材料改性优化的研究逐渐增多, 被生物活性材料改性的 PEEK 的生物相容性及抗菌性不断增强, 我们有理由相信, 在不远的将来, PEEK 在种植领域能够具有更广阔的应用前景。

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