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· 综述 ·

柔性传感器在口腔健康监测应用的研究进展

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【摘要】 口腔健康与面部美观、咀嚼发音和全身疾病紧密相关, 柔性传感器能通过健康监测完善口腔诊疗中存在的不足。本文就近年来柔性传感器在口腔健康监测方面的研究应用做一综述, 为进一步研发口腔方面的柔性传感器提供参考。柔性传感器的结构基础包括柔性基底、可拉伸电极和活性层, 各部分通过材料的选择设计以适应口腔环境; 传感器的传感机制涉及电学、光学、电化学、免疫学, 其中电化学生物传感器和光学传感器在口腔领域的应用尤为突出。监测的信号包括正畸力、咬肌力、呼吸湿度、种植体温度等物理信号, 唾液代谢物、口腔气体等化学信号和牙周病、口腔癌标志物等生物信号。目前柔性传感器在口腔这一特殊环境中, 仍面临着诸多挑战, 未来的研究方向包括提升传感器在口腔中的生物相容性、耐湿和柔性贴合能力, 引入温度不敏感材料和保护膜提升稳定性, 引入人工受体和传感器阵列提升选择性等。此外, 多学科合作对于突破当前瓶颈、实现更精确的疾病诊断和健康监测至关重要; 寻找与口腔相应疾病相关的高浓度特异性生物标志物是传感器健康监测的关键。通过这些努力, 柔性传感器有望在口腔健康监测领域实现更广泛的应用。

【关键词】 传感器; 柔性电子; 床旁检测; 生物监测; 牙周病; 龋病; 可穿戴电子设备; 生物传感器

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Research progress on flexible sensors in oral health monitoring HUANG Jingwen, HAN Shuang, ZHENG Yi, MA Ning. Department of Periodontics, Hospital of Stomatology, Jilin University, Changchun 130021, China
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【Abstract】 Oral health is closely related to facial aesthetics, mastication, pronunciation, and systemic diseases. Flexible sensors can improve current deficiencies in clinical diagnosis and treatment through oral health monitoring. This paper reviews the research on and application of flexible sensors in oral health monitoring in recent years, providing a reference for the further development of flexible sensors in the oral field. The structural basis of flexible sensors includes a flexible substrate, stretchable electrodes, and an active layer, and each part is designed through material selection to adapt to the oral environment. The sensing mechanisms of sensors involve electricity, optics, electrochemistry, and immunology, among which electro-chemical, biological, and optical sensors are particularly prominent in the oral field. The monitored signals include physical signals such as orthodontic force, bite force, respiratory humidity, and implant temperature; chemical signals such as saliva metabolites and oral gases; and biological signals such as periodontal disease and oral cancer markers. At present, flexible sensors still face many challenges in this special oral environment. Future research directions include improving the biocompatibility, moisture resistance, and flexible fitting ability of sensors in the oral cavity; using temperature-insensitive materials and protective films to improve stability; and introducing artificial receptors and sensor arrays to improve factors such as selectivity. In addition, multi-disciplinary cooperation is crucial for breaking through current bottlenecks and achieving more accurate disease diagnosis and health monitoring.

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In the field of stomatology, finding specific biomarkers related to corresponding oral diseases is the key to sensor health monitoring. Through these efforts, flexible sensors are expected to gain more extensive applications in the field of oral health monitoring.

【Key words】 sensors; flexible electronics; point-of-care testing; biological monitoring; periodontal diseases; dental caries; wearable electronic devices; biosensors

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口腔疾病如龋病、牙周病和口腔癌等,不仅影响咀嚼言语功能和面部美观,还和糖尿病、高脂血症等全身性疾病相互作用^[1-2]。传统的口腔医疗检查容易出现检查时间长、结果误差大、患者难以配合的情况。即时检测(point-of-care testing, POCT)技术能够通过检测平台现场捕获疾病数据,从分子诊断水平辅助医生进行实时判断,提高医疗效率^[3]。柔性传感器进一步融合了柔性电子技术,能够贴合人体并承受机械变形,将信号识别、转换、输出、处理^[4],通过穿戴的方式提供实时长期的疾病数据监测,解决患者需要多次复诊的难题。经过特殊设计和材料选择的口腔柔性传感器,能适应口腔的特殊环境,检测与正畸力、种植体、龋病及牙周炎相关的信号,为口腔健康与疾病的早期诊断提供依据、后续治疗提供帮助^[5]。随着技术的不断更新,它有望为人们提供便捷个性化的医疗健康方案^[6]。本文拟从柔性传感器的基本结构、材料选择、传感机制、传感器在口腔领域的应用、柔性传感器面临的挑战和突破进行阐述。

1 柔性传感器的基本结构和材料选择

1.1 柔性基底

柔性基底是柔性传感器区别于传统刚性传感器的组成部分,它能提供与人体曲面紧密贴合的平台,使得柔性传感器便于佩戴。因此,柔性基底需要具备轻薄、柔性、拉伸性好、绝缘耐腐蚀等性能^[4],应用于口腔还需要具有良好的生物相容性、耐湿性、耐变形能力等。

作为广泛应用的低成本柔性材料,聚二甲基硅氧烷(polydimethylsiloxane, PDMS)、聚酰亚胺(polyimide, PI)等有机弹性聚合物具有机械性能可调、化学惰性、绝缘的特点,既能作为柔性基底又能作为电极弹性体材料^[7]。水凝胶是一种具有三

维聚合物网络结构的材料,也适合作为柔性基底应用于口腔。常用的水凝胶例如聚丙烯酰胺水凝胶、聚乙烯醇水凝胶在力学性能上与人体软组织匹配,还具有生物相容性和亲水性等优点^[8-9]。

1.2 可拉伸电极

可拉伸电极是监测和传输电信号的组成部分,能在形变时保持信号传输的连续稳定^[10]。常用材料有导电聚合物、碳基材料、金属及半导体材料,目前可拉伸系统主要以弹性体材料或特殊电极结构的形式实现^[11]。

碳基材料例如碳纳米管、石墨烯,通过提升导电性与机械稳定性,提高传感器在不同应用场景的适应性,识别疾病的生物标志物^[12]。碳纳米管以其高长径比、高载流子迁移率和可拉伸性用于电极和活性层的设计^[13]。例如,Zhang等^[14]设计的锰铁掺杂氮的碳纳米管通过显著提高乳酸酶的电催化效率,提高了检测唾液的乳酸的灵敏度。

除了碳基材料,功能化的金属及金属纳米粒子也具备识别与传输信号的能力,Kim等^[15]制备的功能化银纳米片通过触发局部表面等离子体共振来检测牙周病的标志物碱性磷酸酶。除此之外,二维过渡金属碳化物、氮化物或碳氮化物($M_{n+1}X_nT_m$, MXene)以其导电性、高亲水性、可调的电化学性能和柔韧性广泛应用于柔性电子领域^[16]。金属-有机框架(metal-organic frameworks, MOFs)也因其电化学和光学性质,常用于提高灵敏度和选择性^[17]。

1.3 活性层材料

活性层材料是传感器的核心,能将被测量的目标如咬合压力、种植体周围温度转换为信号^[18]。当活性层设计为换能器与生物元件如酶、抗体组合时,能实现从分子水平检测临床生物信号^[19],例如Zhang等^[20]设计了固定抗体的氧化铌/碳化钛纳

米复合材料传感器用于检测牙周病标志物。活性层还能通过集成能量收集机制实现自供能,如摩擦电、压电、热电、生物燃料电池或光伏效应^[21]。

具有导电性能的聚合物如聚吡咯(polypyrrole, PPy)、聚苯胺(polyaniline, PANI)等,因其可调的导电性,适合作为传感器的活性层材料或者充填材料^[22]。采用多孔PANI和PDMS复合微结构的柔性压力传感器通过构建空心结构和微突起表面结构,大大提高了灵敏度和线性度^[23]。

活性层的设计如孔隙率、厚度、应力分布、形态和尺寸能够影响传感器的整体结构特性,比如机械稳定性、导电性。目前许多研究着力于提高传感性能,平衡机械柔韧性和导电性。一种方法是选择兼具良好机械柔韧性和导电性的材料,比如聚(3,4-乙烯二氧噻吩)-聚苯乙烯磺酸、将导电材料与柔性聚合物结合的材料等^[24];另一种方法是设计特定的微结构,如蛇形、螺旋形或波浪形,提高柔软性并保持导电性^[25]。

2 传感器的传感机制与口腔领域的应用

口腔领域的传感器,根据监测信号可分为4类:电生理、肌肉运动及受力、牙齿运动及受力、温度、湿度、血管动力学等物理信号;唾液、龈沟液中的电解质和代谢物、呼气气体等化学信号^[26];口腔环境中的基因组学、蛋白组学、代谢组学相关标志物的生物信号,以及多信号集成。另外,根据机制的不同分为生物传感器和电子传感器^[27]:生物传感器通过将固定化的生物活性分子定位到构建的电极上来获取生物、化学活性信号;电子传感器直接采用具有电性能的材料实现对物理化学信号的测量,例如用压电弹性衬底捕获人体机械信号^[28]。

柔性传感器可以监测正畸力、种植体温度、咬肌电生理以及获取部分生化信号,在口腔领域有广泛应用。下文将分类阐述口腔领域的主要传感器以及柔性传感器的应用。

2.1 电化学传感器

电化学传感器经氧化还原反应将目标分析物的信息转换为可测量的电信号^[29],通过安培法、电位法和伏安法监测。Zhang等^[14]设计的电化学生物传感器的微芯片,可以在30 s内定量精确测量唾液中的乳酸浓度,为牙周炎提供初步数据。Ara-kawa等^[30]研发的涂有醋酸纤维素膜的电化学传感器,可以实时检测唾液中的葡萄糖,间接反映血糖情况与龋齿、牙周病风险,从而非侵入性地监测与

管理糖尿病、控制糖尿病型牙周炎。目前的电化学传感器仍然面临着电极污染、电噪声、电子集成和电源供应等挑战,在口腔领域多为POCT,尚不能安全长期地进行监测^[17]。

2.2 光学传感器

与传统的电化学传感器相比,光学传感器因其高灵敏度、快速响应和抗电磁干扰的能力,在临床疾病检测中更具优势^[31]。光学传感器利用光学原理检测参数,在心率、血氧饱和度等生理信号的监测方面发挥了重要作用^[32]。

依赖于能够改变光学检测元素的化学或生物反应,光学传感器也可以监测生物标志物。目前,已经研究出用以检测循环肿瘤细胞(circulating tumor cells, CTCs)的表面增强拉曼散射(surface-enhanced raman scattering, SERS)便携式平台,成功做到无需富集癌症检测指标^[33]。Wang等^[34]进一步报道了以SERS作为底层传感原理的柔性光学传感器,攻克了柔性超材料纳米结构在形变下难以保持活性的技术难关,实现生物体内多目标分子的无损、实时、在线监测。

表面等离子体共振传感器,是用金属纳米结构在特定频率下与入射光相互作用产生的局域电磁场增强效应作为底层原理的光学传感器,能提供目标生物分析物的光学无标记检测^[35]。在口腔医学领域,Li等^[36]研发的金-银核壳纳米聚二甲基硅氧烷可穿戴护齿器,利用金-银核壳纳米棒与挥发性硫化化合物的局部反应,通过改变纳米棒表面等离子体共振实现了颜色变化,从而准确定位龋齿。另外,还有基于荧光猝灭的传感器、结合光学分析的侧流免疫分析等可以用于监测唾液中的标志物,如药物滥用标志物、口腔癌早期标志物等^[37]。

2.3 生物传感器

生物传感器可基于电化学、光、声波等原理,通过目标与生物分子识别元件的结合、换能器的转换来检测生物信号。生物传感器的分类是基于该分析装置的生物识别元件与传导元件直接接触的空间特性^[38],因此它也可能属于复杂的电化学、光学传感器,经典的生物传感器有酶传感器、免疫传感器。酶传感器利用酶作为生物识别元件,通过测量与目标分子发生特异性反应产生或消耗的电活性物质,实现对目标物质的定量检测^[39]。免疫传感器则是利用抗原和抗体之间的特异性识别功能来检测样品中的抗体或抗原,通常由生物识别分子(如抗体或抗原)和信息转换器(如光敏元

件)组成,优势在于高特异性与定量检测^[40]。Dong等^[41]提到了一种聚噻吩-C70有机光电探测器的光学微流控生物传感器,通过吸收光谱法来检测唾液中的白细胞介素-8、白细胞介素-1 β 和基质金属蛋白酶-8,在牙周病等监测方面具有重要意义。Joe等^[42]成功研发了一种基于适配体的便携式电化学生物传感器,通过适配体二元组的夹心型结合方式,利用辣根过氧化物酶(Horseradish peroxidase, HRP)标记的二级适配体与目标分子结合形成三明治复合物,进而通过HRP催化3,3',5,5'-四甲基联苯胺二盐酸作为电子介体产生电流信号,以检测存在于牙龈沟液中的牙周病生物标志物——人牙源性成牙本质细胞相关蛋白(human odontogenic ameloblast-associated protein, ODAM),实现早期牙周病的快速诊断。目前,由于生物传感器的复杂机制以及口腔环境的严苛条件,检测酶、免疫标志物等生物标志物在口腔领域的柔性传感器研究数量不多。

2.4 其他物理信号传感器

根据电传感器的电阻、电容、压阻等传感原理设计的压力传感器,常应用于测量咬合力、牙齿的侧向力、口腔—舌压力、机械疼痛阈值、吞咽力等口腔内的压力,在临床辅助进行咀嚼效率的监测、睡眠磨牙症的诊断、吞咽困难等口腔功能障碍的分类等^[43]。Hu等^[44]研发的一种柔性六维力敏电阻压力传感器,利用分布在柔性印刷电路板上的压力敏感导电片,通过不同方向的刺激产生的接触变形来检测六维力。该传感器应用于检测正畸治疗所受的力和力矩,辅助医生获取更合适的矫正力。O'Hare等^[45]开发了一种安装于上颌后牙颊侧黏膜的微电子机械系统压阻式压力传感器,并结合机器学习算法来诊断磨牙症。该传感器被嵌入在医用级硅胶中,直接感受从硅胶表面传达到膜片的咬肌力,并将信号传输到移动设备。传感器的监测结果与肌电图相似,因此具备成为长期家庭监测平台的可能。

Kim等^[46]报道了一种基于电阻温度系数原理的牙种植体温度传感器,该传感器使用了柔性PI基底和微加工工艺,通过包裹围绕牙科种植体的基台实时监测植入物的温度变化,可以在植入失败时发送早期预警信号,从而有助于实时诊断感染性疾病。Thiyagarajan等^[47]研究的一种基于电容原理、使用多壁碳纳米管和PDMS复合的屏印互指电极的纸基湿度呼吸传感器,以无创、低成本的形

式监测呼吸模式,具有家庭式监测与矫正口呼吸不良习惯的潜能。

3 口腔柔性传感器的技术挑战与突破

为了在口腔这一复杂环境中实现监测,柔性传感器不仅需要适应口腔的潮湿环境、酸碱度、肌肉舌头的灵活运动,还需要提高生物相容性和便携性,延长监测时间^[5]。

因为口腔环境潮湿和唾液流动,生物感受器的酶容易丧失其生物识别功能^[48],封装不仅能够提升设备的稳定性,还能防止不必要的损耗。生物污染物的积累也会影响传感器的稳定性,采用保护膜可以有效防止生物污染^[49];人工受体的使用不仅能提升信号采集的稳定性,还能减低加工难度从而降低成本^[50]。

咀嚼时的温度变化会影响传感器材料的稳定性,引入温度不敏感材料可以避免影响。应用自修复材料如在水凝胶中加入丝素蛋白或单宁酸、增强内部化学力^[51-53],能提高材料的自愈能力,提升柔性电子产品的整体贴合性能^[54]。另外,口腔和颅面生物化学环境以及口腔频繁运动带来的机械应力增加了对特定目标信号监测的难度。采用具有强附着力的导电水凝胶作为基底^[55],并利用离子添加剂来提高材料的拉伸性和导电性,同时降低其杨氏模量,可以实现更好的传感精度^[56]。

开发基于人工受体的高选择性酶的技术^[57],利用选择性传感器阵列来识别不同的分析物并通过数据分析来确定混合物成分^[4],可以提高传感器的选择性。此外,对于物理信号的测量,设计传感器的几何结构,使其在特定方向的外力下产生响应,有助于精确的力学分析^[58]。高灵敏度对于准确监测口腔和颅面状态至关重要,尤其是在口腔生物液中许多生物指标的浓度远低于血液的情况下^[59]。

小型化是口腔柔性电子设备发展的另一个关键方向。利用3D打印和薄膜溅射技术,结合半固体/固体电解质,可以构建微尺度的柔性电子器件^[60]。此外,基于柔性半导体材料的传感器,如晶体化硅纳米膜,可以实现超薄的传感器设计^[61]。

口腔内的细菌种群对安装的柔性电子产品构成潜在威胁。在器件表面应用抗菌涂层可以降低细菌的附着率;或者通过集成自热电极的热量来消除细菌^[62]。

4 小结与展望

传感器技术和POCT技术在临床上已显现巨大的应用潜力,尤其是那些结合了柔性电子技术的设备,因其适应性和长期监测能力而在口腔医学领域受到重视。近十年来,柔性传感器在实验研究中取得了进展并应用于运动健康领域,如与智能手表耦合监测指标,但是在医疗领域的应用仍具有巨大的提升空间。通过共同研究,专家们提出了“3S”原则以突破柔性传感器的发展瓶颈——稳定性(stability)、选择性(selectivity)和灵敏性(sensitivity)

[4]。针对口腔的柔性传感器,Wang等^[5]进一步提出了“5I”原则:隐蔽化(imperceptibility)、智能化(intelligence)、个性化(individualization)、集成化(integration)和低成本化(inexpensiveness)。除此之外,柔性传感器的研究需要多学科的合作,这不仅涉及材料科学、微纳米加工技术、新兴的人工智能技术,还需要对口腔疾病标志物的深入探索。遵循上述设计原则和多学科合作,未来开发的口腔柔性传感器才能实现更精确的疾病诊断和健康监测(图1)。

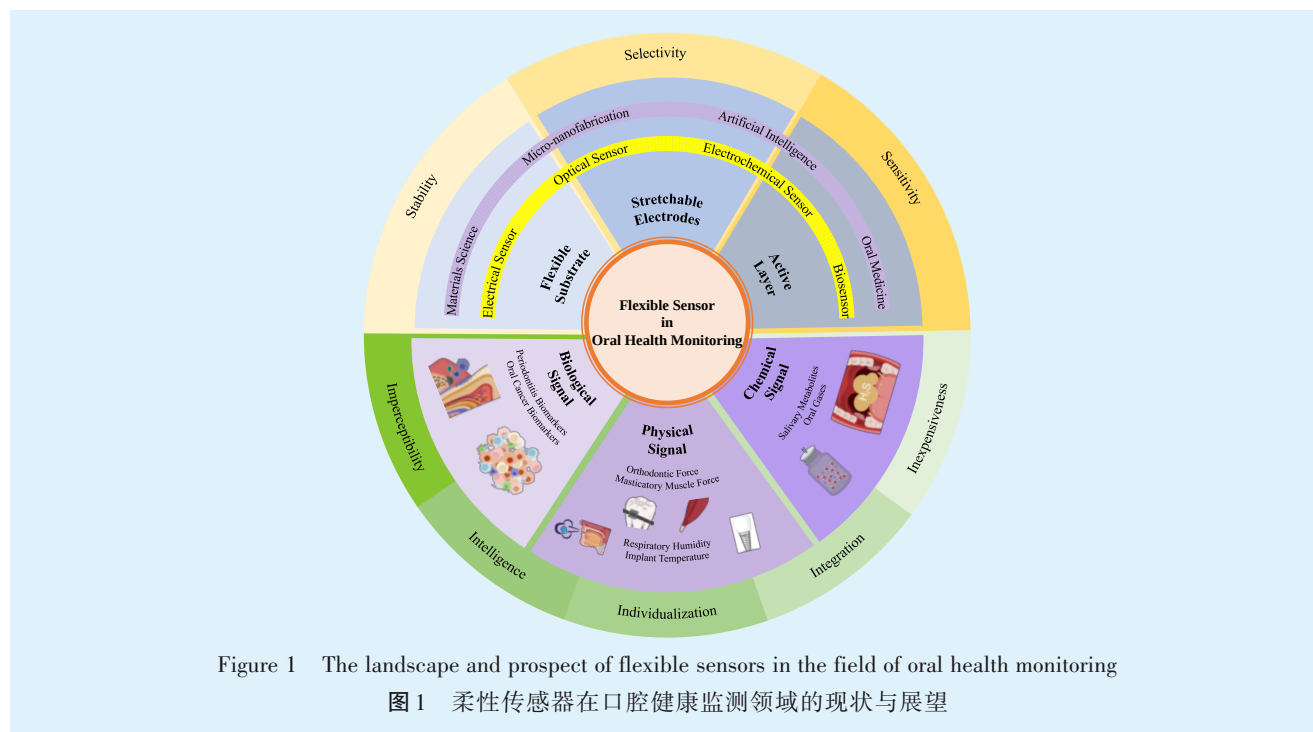


Figure 1 The landscape and prospect of flexible sensors in the field of oral health monitoring

图1 柔性传感器在口腔健康监测领域的现状与展望

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