



仿生多孔膜材料研究进展

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摘要 近些年来, 仿生多孔膜材料已经成为膜材料领域研究热点之一. 与传统多孔膜材料相比, 受生物膜独特孔道结构和分离特性启发的仿生多孔膜具有优异选择分离性、智能门控性和智能响应性等优点, 实现了多孔膜材料性能的突破并拓展其应用领域. 首先, 本文从生物膜的多尺度孔道结构出发, 讨论生物膜的孔道结构与其选择分离性、门控性和响应性的关系, 并系统介绍受生物膜启发的仿生纳/微米尺度多孔膜材料的构建思路、结构及性能. 其次, 介绍仿生多孔膜在水处理、能源、生物医学及物质检测等主要应用领域的最新研究进展. 最后, 讨论了仿生多孔膜材料在发展过程中所面临的挑战, 并展望了其发展趋势.

关键词 多孔膜, 仿生, 生物膜, 纳/微米尺度孔道

多孔膜材料基于孔径筛分作用和界面效应可以实现对物质的选择性分离^[1,2], 已经在水处理^[3,4]、能源^[5,6]、气体分离^[7]、生物医疗^[8,9]及物质检测^[10]等领域得到了广泛的应用. 例如, 纳滤膜基于纳米级孔径筛分效应和界面静电排斥效应, 可以实现对不同价态盐离子的选择性分离, 在硬水软化、重金属离子去除、药物分离及果汁浓缩等领域得到了广泛的应用^[11]. 又如, 微滤膜基于孔径筛分作用和界面超浸润特性, 实现对大分子有机物、蛋白质、微生物、油污染物的选择性分离, 在油水分离、工业废水处理、微生物分离及食品工业等方面得到了广泛的应用^[12,13]. 但是, 多孔膜材料在应用中仍然存在一定的问题, 如反渗透膜在海水淡化过程中存在能耗高的问题、纳滤膜对一价和二价金属离子选择分离性低的问题、微滤膜在油水分离过程中抗污染

性差的问题等, 限制了其进一步发展^[14~17].

自然界生物体内存在不同尺度孔径的生物膜, 经过长时间的进化, 展现出具有近乎完美的结构, 赋予其在特定环境中独特的选择分离性、智能响应性和智能门控性, 为解决多孔膜材料在实际应用过程中存在的问题提供了新的思路^[18,19]. 如, 细胞膜具有独特的纳米尺度水通道, 其仅让水分子通过而完全截留其他物质, 展现出对水分子的高度选择分离性^[19,20]. 受此启发, 研究人员开发出含有水通道蛋白的仿生多孔膜, 有助于解决多孔膜材料在海水淡化过程中存在的水传输速率低和能耗高等问题^[17,21]. 叶子中的微米尺度气孔可以在太阳光的照射下关闭而在无光照的时候打开. 受此启发, 研究人员开发出了具有光响应特性的智能响应性多孔膜, 其孔道结构可以在光刺激下关闭或打开, 实

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现动态可调控的物质传输,解决了传统多孔膜材料响应性差的问题^[22,23]。受肺泡中具有门控特性的微米尺度肺泡孔启发, Hou等人^[24]和Fan等人^[25]开发出以压力为驱动力的智能响应液体复合门控多孔膜,其能够实现膜孔道打开和关闭状态的动态可逆调控,在含油废水处理、多相分离、微流体及微型化学反应器等领域具有广阔的应用前景。因此,从生物膜独特的纳/微米尺度孔道结构入手,研究其与生物膜独特的选择性分离性、门控特性和物质传输特性的关系,开发出不同尺度孔道的仿生多孔膜,有助于解决多孔膜材料在实际应用过程中遇到的问题^[17,26,27]。

本综述主要从生物膜具有的独特纳/微米尺度孔道出发,重点讨论具有纳/微米尺度孔道生物膜的选择分离机理、门控特性及智能响应性,并系统介绍受生物膜启发的仿生纳/微米尺度多孔膜材料的构建思路、分离机理及应用领域(图1)。本文主要包括以下3个部分:(1)着重介绍了细胞膜和肾小球滤过膜中纳米尺度孔结构、叶子气孔和肺泡孔的微米尺度孔道结构及其选择分离性和物质传输特性,及受其启发的仿生纳/微米尺度多孔膜材料的研究进展;(2)主要介绍了近些年仿生纳/微米尺度多孔膜在水处理、能源、生物医学及物质检测领域的最新研究进展;(3)讨论了仿生多孔膜在发展和应用过程所面临的挑战,并提出了一些建议和展望。

1 仿生纳/微米尺度多孔膜

基于孔道尺度特征,可以将仿生多孔膜分为两类:仿生纳米尺度多孔膜和仿生微米尺度多孔膜。仿生纳米尺度多孔膜主要包括受细胞膜离子通道蛋白启发的

仿生离子通道多孔膜、受细胞膜水通道蛋白启发的仿生水通道多孔膜,以及受肾小球滤过膜启发的仿生血液透析多孔膜。仿微米尺度多孔膜主要包括受叶子气孔启发的仿生智能响应多孔膜和受肺泡孔启发的仿生液体复合门控多孔膜。

1.1 仿生纳米尺度多孔膜

在自然界中主要存在多种具有纳米尺度孔道的生物膜,当前研究主要集中在细胞膜和肾小球滤过膜。通过研究这些生物膜的孔道结构和选择分离机理,可以开发出新型仿生纳米尺度多孔膜,实现对小分子或盐离子的选择性分离和传输^[18]。

首先,细胞膜中纳米尺度的孔道主要有离子传输孔道和水传输孔道,而这些孔道主要存在细胞膜中蛋白质内,具体带有离子通道的蛋白被命名为离子通道蛋白,而带有水通道的蛋白被命名为水通道蛋白。离子通道蛋白主要嵌入或贯穿细胞膜内部离子通道蛋白的中间,存在供特定离子穿过的孔道(图2(a)),赋予细胞膜优异的离子选择特性、离子整流特性以及离子门控特性,是实现细胞能量转换、神经细胞膜电位调控及细胞间通信等功能的根本^[18,28]。根据离子选择性可以将离子通道分为钾离子通道、钠离子通道、钙离子通道等,而根据离子通道门控机制可以分为配体门控离子通道、电压门控离子通道及机械敏感门控离子通道^[29-31]。因此,受细胞膜中纳米尺度的离子通道启发构筑仿生离子通道多孔膜,将有助于解决传统多孔膜材料存在的离子选择性差和智能响应性弱等问题,促进纳米尺度多孔膜材料在海水淡化、能源、物质检测等

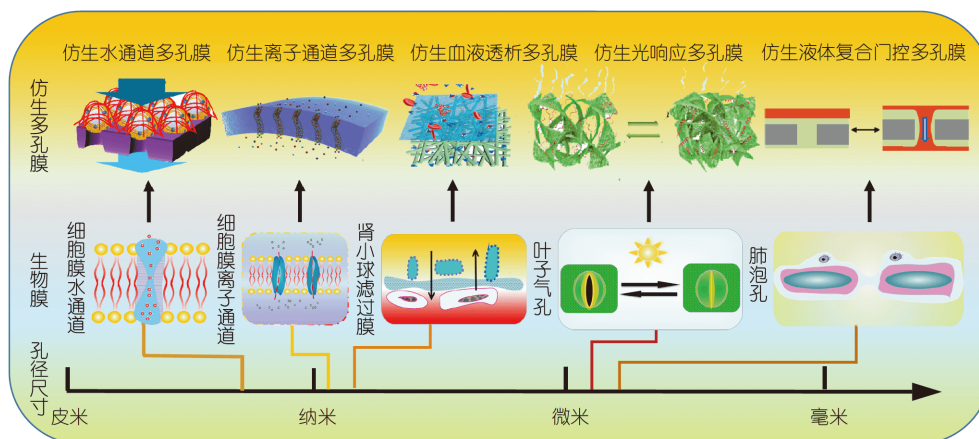


图1 不同尺度孔道生物膜及仿生多孔膜材料

Figure 1 Biological membranes and bio-inspired porous membranes with difference pore sizes

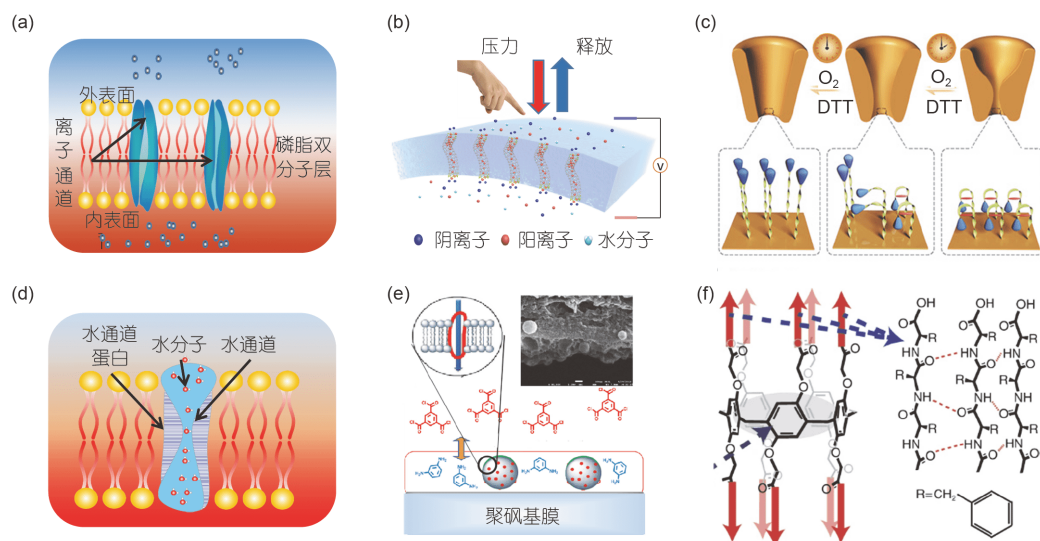


图2 生物多孔膜中纳米尺度孔道及其仿生多孔膜。(a) 细胞膜中离子通道结构示意图。(b) 动态曲率响应纳米通道膜结构及其离子传输行为^[32]。(c) 肽链CGGC改性纳米级多孔膜在二硫苏糖醇(DTT)刺激下的开孔与关孔过程^[33]。(d) 细胞膜中水通道蛋白结构示意图。(e) 含水通道蛋白基仿生多孔膜制备过程示意图及其断面形貌图^[34]。(f) 仿生人工水通道分子结构^[35]

Figure 2 Nanoscale pore of biological porous membrane and its bio-inspired porous membrane. (a) Schematic illustration of ion channels in cell membrane. (b) Structure of dynamic curvature nano-channel-based membrane and its anomalous ionic transport behaviors^[32]. (c) Closing and DTT-driven opening process of the peptides CGGC modified nano-porous membrane^[33]. (d) Schematic illustration of aquaporin in cell membrane. (e) Schematics showing the synthesis process and the cross section structure of aquaporin-based biomimetic membranes^[34]. (f) Molecular structure of bio-inspired artificial water channels^[35]

领域的进一步发展^[10,31]。

当前, 构筑仿生离子通道多孔膜的策略主要有: (1) 将纳米尺度孔道与基材结合成膜; (2) 直接通过化学刻蚀或激光刻蚀等技术在基材中构筑纳米尺度孔道再进行化学改性成膜。例如, 本课题组^[32]首先采用化学气相沉积法制备碳纳米管阵列, 再利用聚二甲基硅氧烷填充碳纳米管间隙, 获得具有纳米尺度离子通道的仿生柔性碳纳米管多孔膜。如图2(b)所示, 该膜内部具有垂直的碳纳米管孔道, 通过外界压力刺激可使碳纳米管通道曲率发生动态形变, 调控膜的离子传输速率, 以实现生物膜离子通道的机械门控特性。江雷课题组^[33]受细胞膜配体门控离子通道启发, 通过化学刻蚀法制备具有纳米级孔道基膜, 再将CGGC肽链接枝到该膜的孔道中, 制备具有类似配体门控离子通道特性的仿生纳米尺度多孔膜。在氧气(O₂)作用下, 膜孔道中的肽链发生分子构型转变以实现孔道关闭, 而在二硫苏糖醇(DTT)的作用下, 膜孔道又会可逆性打开, 实现可控的分子运输。

水通道蛋白广泛存在于多种动植物体内, 因具有超快的水分子传输效率(AQP-1型水通道蛋白可达 $3 \times 10^9 \text{ s}^{-1}$), 而且表现出对水以外所有物质优异的截留特性, 从而受到广泛的关注和研究^[36,37]。其中水通道结构

为沙漏结构(图2(d)), 在磷脂双分子层两侧开口较宽而通道中心较窄(约为0.28 nm), 对直径大于0.3 nm的水合离子可以完全截留^[38-40]。基于以上特性, 研究认为其有助于解决当前反渗透膜在海水和苦咸水脱盐过程中存在的产水率低、高浓度盐水脱盐率低及能耗高等问题^[41,42]。

当前, 主要通过以下两种方法构筑仿生水通道多孔膜: (1) 从生物细胞膜中提取水通道蛋白, 再将其嵌入多孔膜内^[21,34,43]; (2) 人工合成出具有类似细胞膜水通道结构的人工水通道分子, 再将其嵌入多孔膜内^[35,44]。例如, Zhao等人^[34]从大肠杆菌中提取Z型水通道蛋白(Aquaporin Z), 并将该水通道蛋白嵌入到反渗透膜的聚酰胺层中以制备仿生水通道反渗透膜(图2(e))。其水通量高于商品膜水通量的40%, 证明水通道蛋白有助于提高反渗透膜的脱盐性能。又如, Shen等人^[35]先合成出具有水通道结构的柱状芳香烃共聚物, 结构如图2(f)所示, 再将其嵌入到嵌段共聚物中, 最后通过层层自组合法制备仿生水通道多孔膜。该膜的水通道尺寸约为0.5 nm, 对分子量大于500的有机物具有很高的分离效率, 其水通量可以高达 $65 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ 。

肾脏是动物体的重要器官之一, 其主要功能是调节动物体内的水、无机盐离子及酸碱平衡, 从血液中

滤除代谢废物、毒物等物质并重吸收水及其他营养物质,肾脏的滤过功能主要是依靠肾小球滤过膜来实现^[46-48]。如图3(a)所示,肾小球滤过膜主要是由最里面的内皮细胞膜、中间的基底膜及外层的足细胞膜构成,其孔径尺度主要介于50~100 nm^[49,50]。受肾小球滤过膜对血液的独特滤过性能启发,研究人员希望开发出可以去除血液中有毒物质的仿生血液透析多孔膜,为肾功能发生病变的病人提供治疗^[51,52]。

受肾小球滤过膜的多层结构启发,当前构建仿生血液透析膜的主要思路为:先通过相转化法、静电纺丝等技术制备多孔支撑基膜,再通过等离子体活化、化学改性、表面涂覆等技术在基膜表面构筑抗凝血、抗蛋白吸附和高选择性的活性分离层,获得具有纳米尺度孔道的仿生血液透析多孔膜^[9,45]。Jiang等人^[45]先通过相转化法制备聚醚砜中空纤维多孔基膜,再经过等离子体活化处理,将5-磷酸吡哆醛和超支化聚甘油先后接枝到基膜表面得到仿生血液透析多孔膜。如图3(b)所示,该膜表面涂覆的5-磷酸吡哆醛可以有效降低血液中的血同型半胱氨酸含量,而超支化聚甘油可以增强膜的血液相容性并有效保护5-磷酸吡哆醛免于受靶细胞的攻击。Fan等人^[8]受肾小球滤过膜和天然吸

附海绵启发,开发出可以用于血液透析的仿生多孔膜系统。具体该膜系统的血液透析过程如图3(c)所示,基于仿生多孔膜的滤过作用和仿海绵微球的吸附作用,可以有效去除血液中的有毒物质,如尿素、肌酐、溶菌酶和 β 2-微球蛋白等。Yu等人^[9]采用静电纺丝法制备高孔隙率的聚丙烯多孔基膜,再通过化学交联法在基膜表面化学交联一层亲水的聚乙烯醇层,以制备仿生聚乙烯醇/聚丙烯腈多孔膜(图3(d))。该膜具有类似肾小球滤过膜的有毒物去除和蛋白截留功能,同时还具有良好的渗透性和血液相容性。

1.2 仿生微米尺度多孔膜

当前,在生物体内具有科学意义的微米尺度多孔生物膜主要有肺中肺泡膜和叶子气孔,受其启发,研究人员开发多种具有智能响应特性或智能门控特性的仿生微米尺度多孔膜。

肺泡孔在肺的呼吸作用中起到关键的作用,在肺泡扩张时其会完全张开而在肺收缩时则关闭。肺泡孔基于毛细力作用而使表面活性液体稳定填充在其孔道内(图4(a)),其可在一定压力驱动下迅速开启或关闭,是一种独特的液体复合门控孔道^[54,55]。受其启发,Hou等

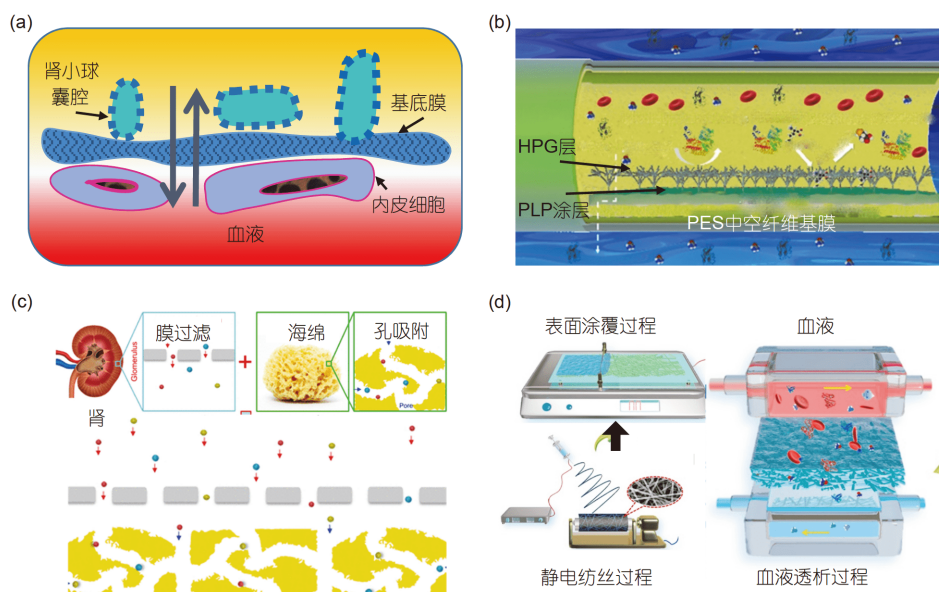


图 3 微米尺度生物多孔膜及其仿生多孔膜。(a) 肾脏中肾小球滤过膜结构示意图。(b) 分级5-磷酸吡哆醛(PLP)-g-超支化聚甘油(HPG)/聚醚砜(PES)多孔膜血液透析过程^[45]。(c) 受肾小球膜和海绵启发的仿生多孔膜系统结构图^[8]。(d) 用于血液透析的仿生聚乙烯醇(PVA)/聚丙烯腈(PAN)多孔膜制备过程^[9]

Figure 3 Micro-scale biological porous membrane and its bio-inspired porous membrane. (a) Illustration of glomerular filtration membrane in kidney. (b) Hemodialysis process of hierarchical PLP-HPG layer-functionalized PES porous membranes^[45]. (c) Bio-inspired porous membrane system of a molecule-filtrating glomerulus inspired by the glomerulus and natural sponge^[8]. (d) Illustration for the fabrication procedure of bio-inspired PVA/PAN composite porous membrane for hemodialysis^[9]

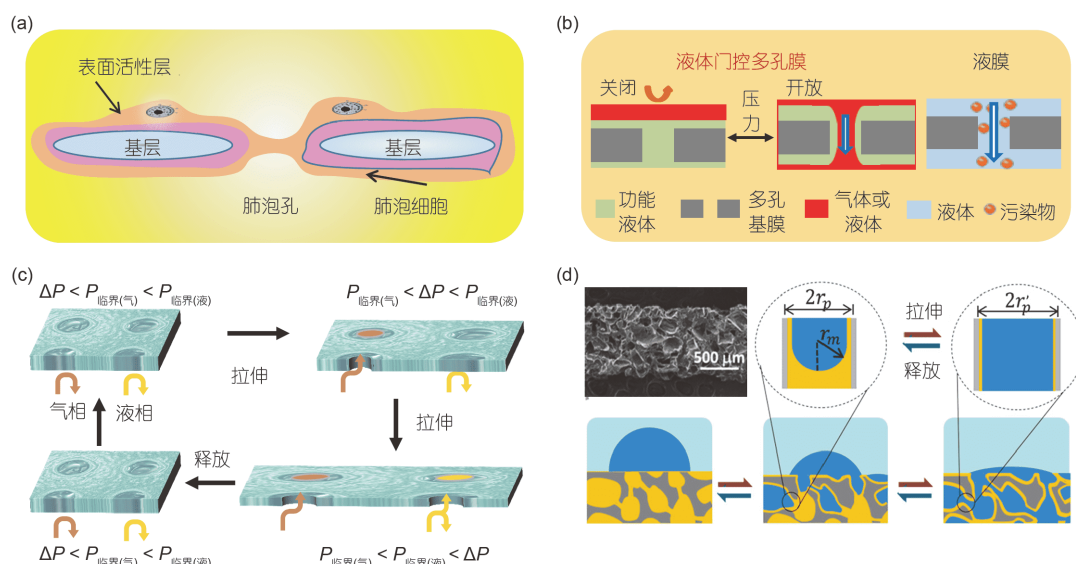


图4 肺泡膜及其仿生多孔膜。(a) 肺泡孔结构示意图。(b) 传统液膜和液体门控多孔膜分离过程示意图^[24]。(c) 弹性液体复合多孔膜动态气液传输过程^[53]。(d) 仿生弹性液体注入膜断面形貌结构及其水下呼吸气泡过程^[54]

Figure 4 Alveolar membrane and its bio-inspired porous membrane. (a) Schematic diagram of alveolar pore in alveolus. (b) Separation process of traditional liquid membrane and liquid-based gating membrane^[24]. (c) Dynamic gas and liquid transport process of liquid gating elastomeric porous membrane^[53]. (d) Cross section and on-demand bubble manipulation process of bio-inspired elastic liquid-infused membrane^[54]

人^[24]首先提出了仿生液体复合门控膜概念(图4(b)): 利用毛细力作用, 将液体稳定填充在多孔膜孔道内部, 形成一种能够可逆性开关的液体复合门控膜。与传统的利用化学势梯度或浓度差驱动的液膜不同, 液体复合门控多孔膜是把液体作为动态结构材料与多孔基膜复合, 实现压力驱动的液门开关。仿肺泡孔液体门控膜的构建策略主要为: 先制备具有微米尺度孔道的多孔膜, 再基于毛细作用力将功能液体填充到多孔膜的孔道内部, 以得到液体复合门控多孔膜^[24,54,56]。例如, 本课题组^[53]采用激光刻蚀技术制备了弹性有机高分子多孔膜, 再将功能液注入到该膜的孔道中, 获得一种在恒压环境下可以实现多相输运与动态分离的弹性液体复合门控复合膜。如图4(c)所示, 在恒压环境中, 因为气体和液体的跨膜临界压力值均大于恒压的压力值, 气液均不能通过膜孔道。当动态拉伸弹性液体门控膜时, 因膜孔增大导致气体的临界跨膜压力低于恒压压力而通过液门孔道。当进一步拉伸时, 气体和液体的临界跨膜压力值均大于恒压压力值, 此时气液两相均能通过液门, 当释放拉伸应力后又可以恢复初始状态。江雷课题组^[54]受肺泡孔的收缩性启发, 采用盐模板法制备了弹性有机硅多孔膜, 将全氟润滑剂作为功能液注入到多孔膜中, 得到一种弹性液体注入多孔膜。该液体注入多孔膜可以动态实现对气泡的吸附、释放及运输, 具体

过程如图4(d)所示。在水下环境中, 气泡会随着该膜孔的拉伸膨胀而浸入膜孔内, 当释放拉伸应力时, 气泡又会因膜孔的收缩而被挤出。

气孔在植物的碳同化、呼吸、蒸腾等气体代谢过程中, 主要作为二氧化碳、氧气和水蒸气的通路。在光照作用下, 气孔因保卫细胞内水势下降而关闭, 阻止物质交换。无光或弱光时, 气孔因保卫细胞内水势升高而自动打开, 进行物质交换(图5(a))^[58]。叶子气孔的这种在太阳光照射下的关闭而在无光时打开的特性为开发智能响应多孔膜提供了思路^[22,59]。

当前受气孔启发的智能响应仿生多孔膜的构建思路主要是将具有光或热刺激响应性的聚合物或分子引入到多孔膜。如, Zhang等人^[23]基于冷冻干燥技术和激光加工技术, 利用具有光热特性的石墨烯和热响应特性的聚(*N*-异丙基丙烯酰胺)的协同作用, 开发出一种具有光响应特性的石墨烯/聚(*N*-异丙基丙烯酰胺)多孔膜。如图5(b)所示, 在强阳光照射下, 该膜的微孔会发生收缩闭合, 膜的光热蒸腾产水速率降低; 而在弱光时, 该膜的微孔膨胀打开, 膜的光热蒸腾产水速率增加, 实现依靠太阳光照强度变化调控膜的光热产水速率。Li等人^[57]将对一氧化氮(NO)分子具有特异响应性的铁卟啉接枝到聚对苯二甲酸乙二醇酯膜的孔道内, 制备一种智能响应的钾离子传输多孔膜。如图5(c)所示, 当气体

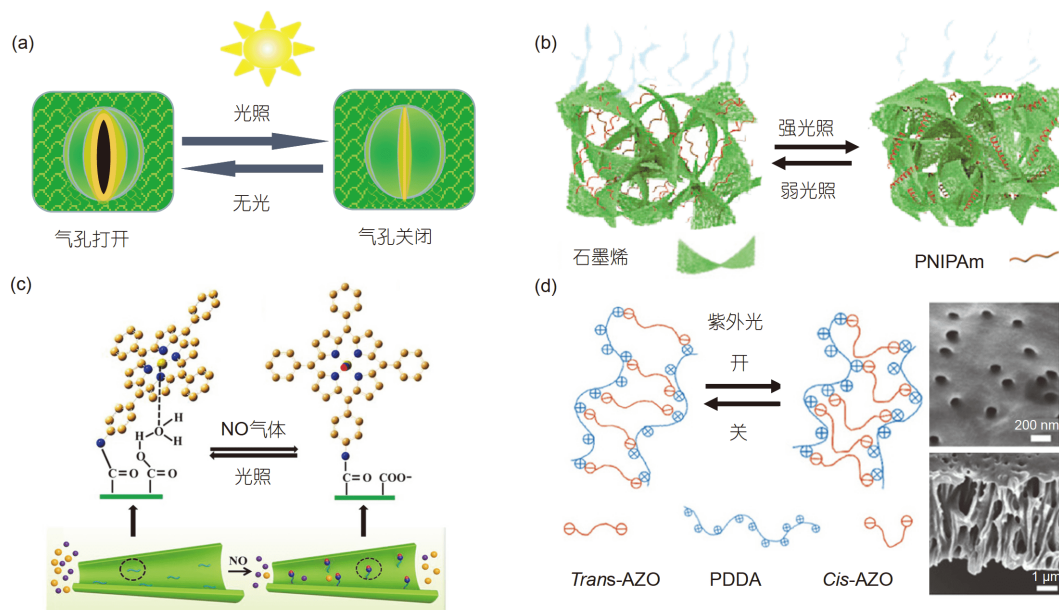


图 5 叶孔及其仿生多孔膜. (a) 叶片气孔在光照和无光作用下的开合过程. (b) 仿叶子气孔微结构石墨烯/聚 N -异丙基丙烯酰胺(mG/PNIPAm)在不同光强射下的膜孔开/关过程^[23]. (c) 铁卟啉分子接枝膜在一氧化氮(NO)/光照刺激下的开合与关闭过程^[57]. (d) 偶氮苯聚合物的结构及其在紫外光照射下收缩和释放过程^[22]

Figure 5 Stomatal and its bio-inspired porous membrane. (a) Schematic illustration of the opening/closing process of stomatal under intense and no sunlight irradiation. (b) Pore structure turned on/off process of a leaf-like mG/PNIPAm membrane under solar irradiation of different intensity^[23]. (c) Opening/closing process of ferroporphyrin molecules gated membrane under NO/light active^[57]. (d) A reversible contraction and expansion change of AZO-PDDA supermolecules at UV region^[22]

信使分子NO通过膜孔时,与膜孔内的铁卟啉结合时,促使膜的孔道打开,钾离子能快速通过膜的孔道.当停止NO刺激改为太阳光照射时,NO从亚硝酰基铁卟啉中解离,膜孔道重新关闭,此时钾离子不能穿过膜孔.Chun等人^[22]受叶子气孔启发,将具有紫外光响应性的偶氮苯聚合物接枝到聚碳酸酯膜孔道内以制备一种智能光响应多孔膜.在紫外光照时,该膜孔内的偶氮苯聚合物会发生收缩,使膜孔增大,导致该膜对离子的传输速率增加;当无紫外光照时,偶氮苯分子链会重新铺展而使膜孔减小,导致离子传输速率降低,实现了光响应可控的物质传输过程(图5(d)).

2 仿生多孔膜材料的应用

向自然学习开发出的仿生多孔膜材料为解决当前多孔膜材料在水处理、能源、生物医学及物质检测等应用领域中存在的问题提供了新的思路和解决方案,促进多孔膜材料的进一步发展.

2.1 水处理领域

当前多孔膜材料虽然在水处理领域得到了广泛的

应用,但在实际应用过程中有很多问题亟待解决,如产水率低、能耗高、分离选择性差及抗污染性差等问题^[60-62].通过借鉴具有独特选择分离性、极高传输速率及良好门控特性的多孔生物膜开发出的仿生多孔膜材料,为解决这些问题提供了新的方向.

针对正渗透膜在海水淡化过程中脱盐率不高的问题, Fuwad等人^[21]制备的含Z型水通道蛋白仿生聚碳酸酯复合多孔膜(图6(a))具有比商品正渗透膜更高的水通量性能,其对NaCl的截留率高达 $97.8\% \pm 0.7\%$,水通量达到 $(7.45 \pm 0.62) \text{ L m}^{-2} \text{ h}^{-1}$.研究表明,这主要归功于该膜中存在的水通道蛋白. Saeki等人^[63]制备的含有短杆菌肽A的仿生反渗透膜(图6(b))对NaCl的截留率高于97%,其水通量约为 $11.08 \text{ L m}^{-2} \text{ h}^{-1} \text{ MPa}^{-1}$,高于商品反渗透膜的水通量(约为 $10 \text{ L m}^{-2} \text{ h}^{-1} \text{ MPa}^{-1}$).针对多孔膜在海水淡化过程中能耗高的问题, Zhang等人^[23]受叶子气孔启发制备的仿生石墨烯/聚(N -异丙基丙烯酰胺)多孔膜在弱光下(低于1个太阳光强度)产水率约为 $1.66 \text{ kg m}^{-2} \text{ h}^{-1}$,而在强光下(1~2个太阳光强度)的产水速率约为 $0.24 \text{ kg m}^{-2} \text{ h}^{-1}$,实现以太阳光为能源的海水淡化过程(图6(c)),并可以根据光照的强弱可控地调节

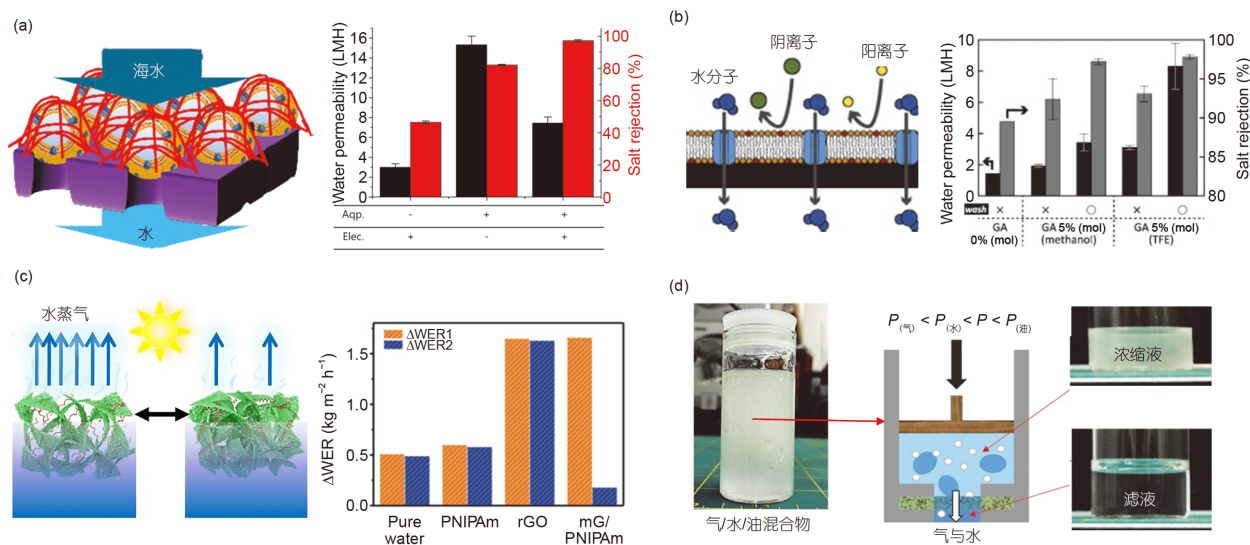


图 6 仿生多孔膜在水处理领域的应用。(a) 聚碳酸酯(PCTE)基水通道蛋白复合膜脱盐性能^[21]。(b) 仿生反渗透膜脱盐性能^[63]。(c) 仿叶子气孔 mG/PNIPAm 多孔膜光热脱盐性能^[23]。(d) 液体门控多孔膜油水分离过程^[24]

Figure 6 Application of bio-inspired porous membrane in water treatment. (a) Desalination performance of PCTE-based aquaporin composite membrane^[21]. (b) Desalination performance of a biomimetic RO membrane^[63]. (c) Photothermal desalination performance of a leaf-like mG/PNIPAm membrane^[23]. (d) Oil/water separation process of liquid-gating porous membrane^[24]

产水速率。针对传统多孔膜在油水分离过程中存在的油水分离效率低和抗污染性差的问题，Hou 等人^[24]开发的仿生液体复合门控多孔膜基于压力调控可以实现高效的油水分离(图6(d))，当施加的操作压力大于水的跨膜压力而小于油的跨膜压力时，水可以通过该膜系统而油则被截留，实现高效的油水分离。

2.2 能源领域

淡水和海水之间蕴藏着巨大的盐梯度能，其被认为是一种很有前景的可持续再生清洁能源，据估算从流入海洋的每立方米河水中可以提取出2.3 MJ的盐梯度能^[67-69]。目前，膜反向电渗析技术是收集该能源的主要技术之一。但是，作为该技术核心的离子交换膜存在离子选择性差及传输效率低的问题，阻碍了该技术能源转化效率的进一步提升。近年来，以细胞膜离子通道为基础的仿生离子通道多孔膜的快速发展，有望解决该技术存在的这一问题。

Wang 等人^[64]受细胞膜离子通道启发制备的仿生离子通道杂化膜，具有类似细胞膜离子通道的快速离子传输特性和离子选择性(图7(a1))。在电场作用下，基于该膜构建的盐差能转化电能器的输出功率高达 1.6 W m^{-2} (图7(a2))。Chen 等人^[65]制备的含氮化硼(BN)仿生离子通道多孔膜在盐梯度下可以产生较高的电流(图7(b1))，在10个膜组件串联时可以输出1.32 V电压，

单个膜组件的输出功率可达 0.6 W m^{-2} (图7(b2))。Ji 等人^[66]分别制备了带负电的石墨烯膜(n-GO)和带正电的石墨烯膜(p-GO)。通过将这对带相反电荷的石墨烯膜耦合到电化学池中(图7(c1))。在双向跨膜化学梯度的驱动下，阳离子通过带负电荷的n-GO膜的离子通道向一侧迁移，而阴离子则通过带正电的p-GO膜的离子通道向反方向迁移。在两个电极之间产生叠加的电化学势差和离子通量，可以输出接近 0.77 W m^{-2} 的功率，比使用商业离子交换膜(约 0.50 W m^{-2})高出约54%(图7(c2))。

2.3 生物医学领域

多孔膜材料在血浆分离、蛋白质分离、血液透析等生物医学领域得到了广泛的应用，但是其在使用过程中存在血液相容性差、选择分离效率低等问题。因此，研究人员希望借鉴生物膜良好的选择性和生物相容性，开发出具有优异选择分离性和血液相容性的仿生多孔膜，解决多孔膜材料在生物医学应用过程中存在的以上问题。

针对传统医用导管存在的功能性单一、无响应性差、抗凝血性能差等问题。本课题组^[70]基于仿生液门复合控多孔膜技术开发出具有优异血液相容性的液体复合门控多孔膜基医用导管(图8(a1))。该膜主要是由PVDF多孔基膜和功能门控液组成。当该门控导管的内

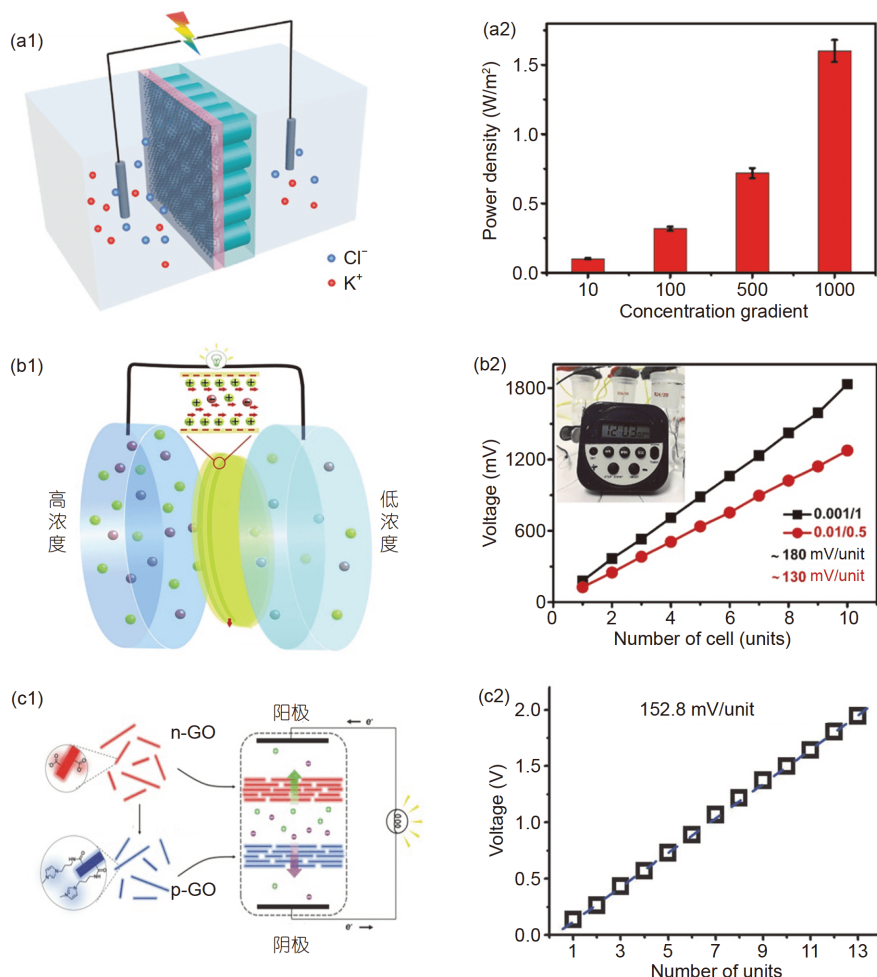


图7 仿生多孔膜在能源领域的应用。(a1) 自制的化学能转化电能检测 I - V 装置示意图。(a2) 不同盐度梯度下输出功率密度^[64]。(b1) ABN30膜分离NaCl溶液串联电池示意图。(b2) 电压与所用ABN30装置数量的关系^[65]。(c1) GOM基纳米流体能量收集装置。(c2) GOM-NRED单元电池串联数量与产生电压的关系^[66]

Figure 7 Application of bio-inspired porous membrane in energy. (a1) Schematic illustration of the self-made electrochemical device for I - V measurement. (a2) Power densities of the hybrid under a series of salinity concentration gradient^[64]. (b1) Schematic of tandem cells with NaCl solutions separated by an ABN30 membrane. (b2) Representation of voltage as a function of the number of ABN30 units used^[65]. (c1) GOM-based nanofluidic energy harvesting device. (c2) Multiple GOM-NRED unit cells connected in series generate a voltage^[66]

外压差变化时, 其内的门控液体厚度也会随之发生动态变化, 使液体门控导管具有类似于血管收缩舒张的自适应形变功能(图8(a2))。另外, 该膜因其功能液体具有极低的表面能而表现出良好的抗凝血性能, 还可以在管壁上不同位置定点释放功能性药物分子, 全面提升传统医用导管的功能性、安全性等。Fan等人^[8]将具有仿肾小球滤过功能的多孔膜与海绵启发的微纳米颗粒结合制备了一个血液净化膜系统(图8(b1), (b2)), 该系统对血液中尿素、肌酐和 β 2-微球蛋白的去除率分别为86%、86%和65%。针对常规血液透析膜的生物相容性差的问题, Yu等人^[9]开发的具有优异亲水性和血

液相容性血液透析PVA/PAN多孔膜(图8(c1)), 其在0.1 MPa操作压力下的渗透通量可达 $290.5 \text{ L m}^{-2} \text{ h}^{-1}$ 。该膜可以去除血液中82.6%的尿素和45.8%的溶菌酶, 保留血液98.8%的牛血清白蛋白, 并且比常规的聚砜血液透析膜具有更强的抗蛋白吸附性能(图8(c2))。

2.4 物质检测领域

膜技术作为一项高效、低成本及操作简单分离技术, 已经在物质检测领域得到广泛应用。将生物膜对物质独特的响应性与多孔膜的分离性结合可以开发具有高效物质检测性能的仿生多孔膜材料, 实现对特定物

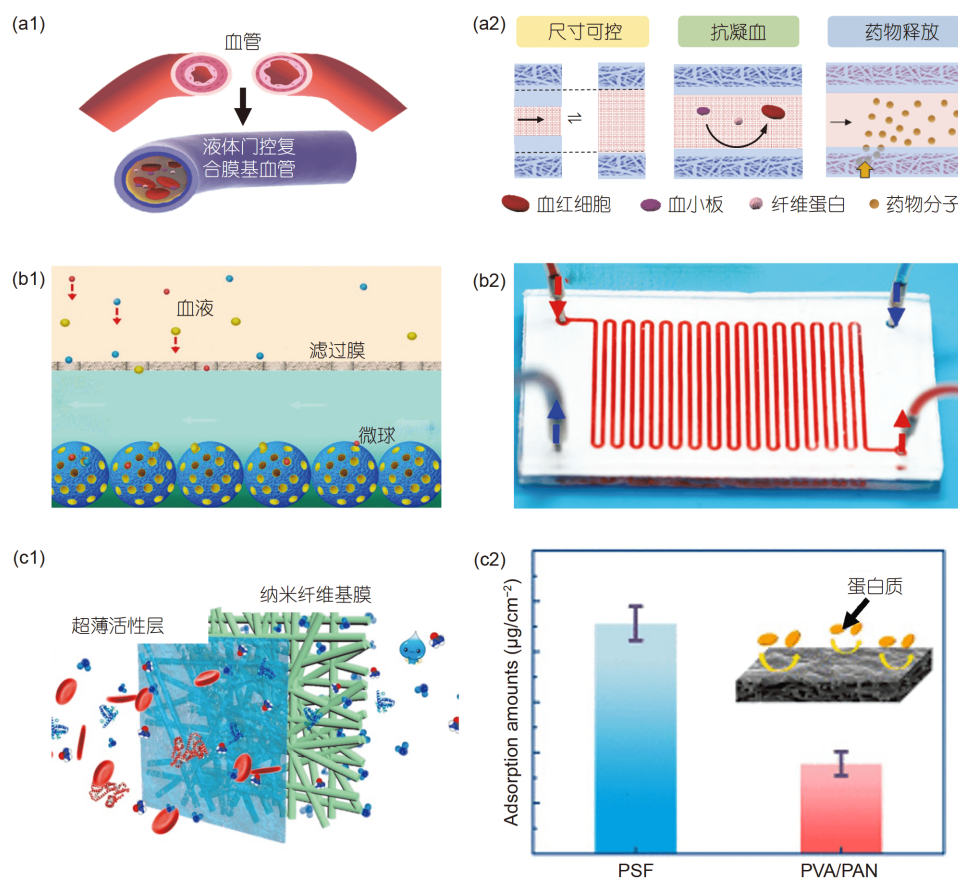


图 8 仿生多孔膜在生物医学领域的应用. (a1) 仿肺泡液体门控医用导管结构示意图. (a2) 膜的可变尺寸、抗凝血性能和药物释放性能^[70]. (b1) 仿生生物分子清洗装置设计原理示意图. (b2) 模拟流体对流进入仿生生物分子清洗装置微通道^[8]. (c1) 血液透析膜的结构示意图. (c2) 膜对BSA的吸附量^[9].

Figure 8 Application of bio-inspired porous membrane in biomedicine. (a1) Schematic diagram of liquid gating membrane-based catheter. (a2) Tube sizes, anticoagulation property and drug release of the membrane^[70]. (b1) Schematic of the design principle of the bio-inspired biomolecule cleaning device. (b2) Photograph of the simulated fluid convectively entering into the serpentine microchannels of the bio-inspired biomolecule cleaning device^[8]. (c1) Schematic of hemodialysis membrane. (c2) BSA adsorption amounts onto the membranes^[9].

质的高效检测。

针对膜技术在物质检测过程存在的灵敏度低、响应性差等问题,本课题组^[25]通过在仿生液体门控多孔膜系统中引入双亲分子,开发出了一种无电可视化液体门控物质检测技术。如图9(a1)所示,该技术的检测机理为功能门控液体中双亲分子(如十二烷基苯磺酸钠)与待检测物质特异性相互作用导致的界面张力变化信息转化为气体跨膜临界压力阈值变化信息。例如,在金属离子检测过程中,金属离子与功能分子发生偶极诱导相互作用,导致双亲分子在界面上重新排布,宏观上表现为界面张力发生变化,最终影响气体跨膜临界压力阈值,而高价金属离子的临界压力阈值降低效果明显高于一价离子。因此,基于以上独特的响应性,可以实现对物质的无电、可视化检测,如图9(a2)所示。基于

细胞膜对离子的独特响应特性,Mo等人^[10]制备了具有纳米尺度离子孔道的ZIF/PAA多孔膜。如图9(b1)所示,在电场作用下,金属离子穿过ZIF-8的纳米孔道时,基于ZIF-8独特的金属离子吸附特性,对应不同金属离子会产生不同的电信号,其中铅金属离子(Pb^{2+})穿过ZIF-8的纳米孔道时产生的电信号最强。因此,该膜可以实现对 Pb^{2+} 的检测,并且对 Pb^{2+} 的检测极限低至 0.03 nmol L^{-1} (图9(b2))。

3 总结与展望

综上所述,仿生多孔膜材料近些年来已经得到了快速的发展,有效地弥补了传统多孔膜材料在应用过程中的短板,拓展了其应用领域。但是仿生多孔膜材料距离规模化生产及应用存在一定距离,因为仿生多孔

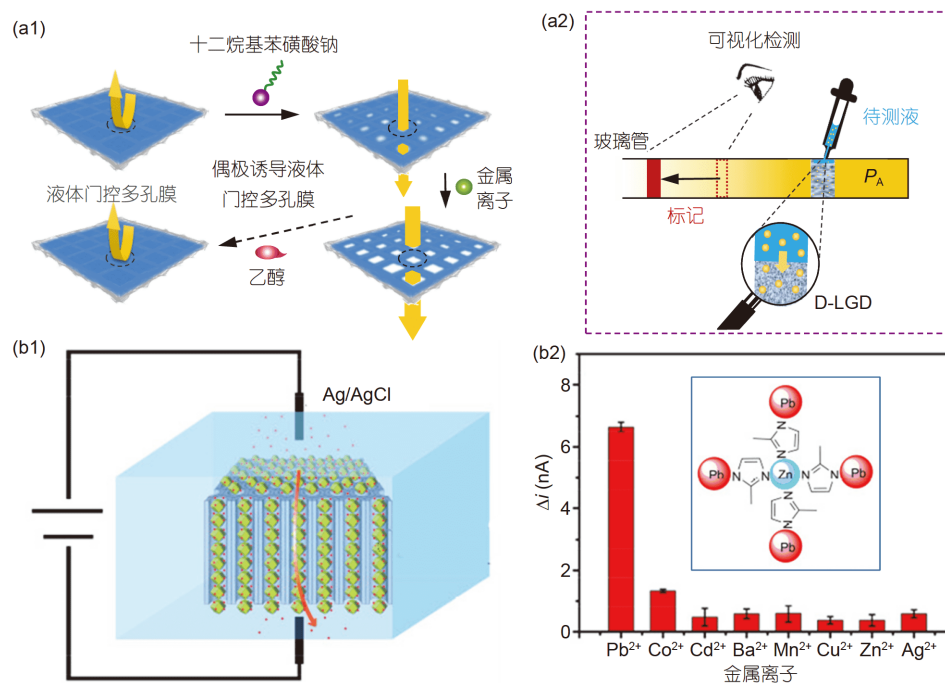


图9 仿生多孔膜在物质检测领域的应用。(a1) 偶极子诱导液体门控型多孔膜检测系统(D-LGD)的检测机理。(a2) D-LGD在可见化学检测中的应用^[25]。(b1) 采用多孔阳极氧化铝膜检测铅离子(Pb^{2+})的仿生传感器。(b2) 对应于其他金属离子和 Pb^{2+} 的电流变化(ΔI)^[10]

Figure 9 Application of bio-inspired porous membrane in detection. (a1) Illustration of the mechanism of the dipole-induced liquid-gating-based porous membrane system for detection (D-LGD). (a2) Application of D-LGD in visible chemical detection^[25]. (b1) Biosensor prepared by porous anodized aluminum membrane was used to detect Pb^{2+} . (b2) Current change (ΔI) corresponding to other metal ions and Pb^{2+} ^[10]

膜材料在以下几方面仍然存在一些问题亟待解决: (1) 理论基础方面. 当前基于分析检测技术的限制, 仅对生物膜的组成、结构和分离机理进行了初步的研究. 因此, 需要通过提升检测手段进一步分析生物膜组成、结构及分离机理之间的内在关系, 构建生物膜模型, 为仿生多孔膜材料研究提供理论指导. (2) 功能性方面. 当前设计的仿生多孔膜仅能实现生物膜的部分功能, 功能比较单一, 需要向更高级、全面的生物膜功能进行仿生, 开发出具有多功能的仿生多孔膜. (3) 制备方

面. 当前仿生多孔膜材料虽然在很多领域取得了良好的应用效果, 但是由于其制备工艺复杂、成本高及稳定性差等问题, 限制其规模化生产及应用. 因此, 应该寻求更简单、低成本的仿生技术, 实现规模化生产仿生多孔膜材料. 随着更多新型多功能材料的出现和制备技术的不断发展, 基于多学科融合开发出具有高选择性、快智能响应性及高稳定性的仿生多孔膜将成为未来的发展趋势. 总之, 向多孔生物膜学习, 多孔膜材料一定能取得更加快速的发展和更广阔的应用.

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Summary for “仿生多孔膜材料研究进展”

Progress in bio-inspired porous membranes

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Porous membranes are one of the research hotspots in the domain of separation materials and have been widely utilized in various fields (such as water treatment, energy, biological medicine) due to their high separation efficiency, low cost, and non-secondary pollution, etc. However, there still exist many challenges in their application process, such as low selectivity, poor anti-fouling, poor responsive, critically limiting its development. Recently, bio-inspired porous membranes offer a brand-new idea for solving those problems and are becoming one of the most exciting research areas in porous membranes field. Compared with traditional porous membranes, bio-inspired porous membranes, designed by imitating the unique pore structure and separation performance of biological membranes, display distinct advantages in selectivity performance, antifouling property, intelligent gating property, and intelligent response property. This critical review detailedly presents the recent developments of bio-inspired porous membranes with multi-scale pore structure. In this paper, the first section mainly gives a brief introduction to the pore structure, separation selectivity, and response property of both nano-scale biological membranes and micro-scale biological membranes. Among numerous nano-scale biological membranes, the most concerning to date include cell membrane with ion channel, cell membrane with aquaporin, and glomerular filtration membrane. Ion channel embedded within cell membrane provides ion selectivity, ion current rectification, and ion gating characteristics to cell membrane. Inspired by this, novel porous membranes with excellent ion selectivity are developed for regulating ion permeation. Meanwhile, aquaporin can let the passive transport of water across cell membrane while simultaneously excluding ions, offering a new strategy for designing bio-inspired porous membrane for high efficiency desalination. Glomerular filtration membrane with the nano-scale pore can prevent the filtration of blood cells and platelets, and removes most of nano-scale molecules from the blood. Inspired by its unique filtration property, various bio-inspired porous membranes have been designed for clinical kidney replacement treatment. Additionally, the micro-scale porous membranes in nature mainly contain alveolar membrane with intelligent gating property and leaf stomata membrane with intelligent response property. By imitating the alveolar membrane, liquid gating porous membranes with intelligent gating property are developed, achieving for controlling the movement of fluids, vapours, and solids. Learning from leaf stomata membrane, intelligent responsive porous membranes are also designed, and can be applied in controllable transmission. For preparing the high performance of bio-inspired porous membranes, the designed strategies are also introduced in detail. The second section has further outlined the applications of bio-inspired porous membranes in water treatment, energy, biological medicine, and substance detection. In water treatment field, aquaporin-based bio-inspired porous membrane possesses better desalination performance than traditional porous membrane. In energy field, bio-inspired porous membranes offer unique ions selectivity and ions transport, thus enhancing energy conversion efficiency in salinity gradient energy capture. In biological medicine field, alveolar-like liquid gating composite membranes can be utilized for fabricating artificial blood vessel with anticoagulation and drug release properties. Moreover, bio-inspired porous membranes with porous support and thin separation layer are demonstrated as hemodialysis membranes for the treatment of kidney disease. In substance detection, visual and low power consuming chemical detection of specific analytes can be achieved by liquid gating composite membrane. In the last section, future challenges in theory, preparation technology, and application of bio-inspired porous membrane are presented, providing an overview of the various solutions. Ultimately, continued efforts to relative research and materials development will promote the rapid development of bio-inspired porous membrane.

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