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【金属腐蚀与防护技术专题】　　**专题主持人:廖伯凯**

光热防冰超疏水表面研究进展

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摘　要: 由环境因素导致工业设备、输电线路等出现低温结冰的现象是亟待解决的问题。近年来,超疏水防冰涂层发展迅速,但与此同时也暴露出不可忽视的弱点,在极端环境下超疏水表面微纳结构易与冰形成“互锁”现象,导致防冰性能衰减甚至消失。而具有光热效应的超疏水涂层能够在光照条件下迅速升温,“解锁”微纳结构与冰粘附,这类表面为防冰提供了一种新的研究思路。文章首先介绍了固体表面结冰机理,阐释了固体表面液滴的凝结行为,总结了不同种类光热材料与超疏水表面的制备方法,归纳了几类具有光热效应超疏水表面的性能与优势;最后,简要阐述了光热超疏水表面防冰机理,并指出了当下光热防冰超疏水表面的不足与未来研究方向的展望。

关键词: 光热材料; 超疏水表面; 防结冰

中图分类号: TQ63　　**文献标志码:** A

Research advancements of photothermal anti-icing superhydrophobic surfaces

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Abstract: In the contemporary emphasis on maintaining social stability and industrial safety, the issue of low-temperature icing, induced by environmental factors affecting transportation, large-scale machinery, and power transmission lines, stands as an urgent challenge requiring immediate attention. Over the past few years, the rapid development of traditional Superhydrophobic Surface(SHBS) anti-icing coatings has been notable; however, these coatings have also revealed significant limitations, particularly their tendency to lose hydrophobic and anti-icing properties under extreme conditions. In contrast, Superhydrophobic(SH) coatings with photothermal effects have demonstrated sustained effectiveness in harsh environments, offering a promising avenue for further research. This paper begins by elucidating the mechanisms of surface icing, detailing the process by which liquid droplets condense on substrate surfaces. It then reviews various photothermal materials and traditional methods for preparing SHBS. Subsequently, the paper illustrates several composite preparation techniques for SH-

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BS with photothermal effects through specific examples. Finally, after a concise explanation of the photothermal SH anti-icing mechanism, the paper highlights the current limitations of photothermal SH anti-icing surfaces and outlines potential directions for future research.

Key words: photothermal material; superhydrophobic surface; anti-icing

冬季的积雪与冰面是人们生活中一道美丽的景色。然而,结冰对许多大型设备与工业设施来说具有灾难性,易造成严重的安全问题和巨大的经济损失[1-4]。例如在热交换器表面形成的冰会使制冷系统的效率降低50% ~70%,绝缘子上严重的结冰将破坏绝缘子的电绝缘性[5],从而威胁到电力传输系统的安全[6-7]。飞机机翼结冰会影响气流分布,影响升力,可能导致机翼或船体表面出现破损,危及人员和设备安全[8-9]。冰的堆积也会对输电线路造成严重的影响,因极端冰雪天气,仅 2008 年的 2 个月就对数以万计的居民生活造成不便,直接或间接经济损失高达 1 100 亿元[10]。

传统除冰方法,如电热除冰、化学除冰及机械除冰,尽管在实际应用中发挥出一定作用,却不可避免地伴随着能耗高、效率低及环境污染等诸多问题[11-12]。在此背景下,受自然界荷叶自洁现象的启发,发现超疏水表面(Superhydrophobic Surface, SHBS)具有优异的拒水性[13]、显著的结冰延迟效应[14]以及较低的冰附着强度[15-16]。凭借其疏水性能与低冰附着力表面,不仅能使水滴快速滑落、延缓结冰,降低对额外能源的依赖,而且还有利于冰层自然脱落。与传统方法相比,SHBS 避免了使用大量化学防冰液,从根本上有效地减少了生产的污染排放,对环境十分环保,故而将 SHBS 视为一种更好的、颇具潜力的防冰材料[17-18]。然而,在高湿与极寒的环境下,SHBS 的微纳结构易于结冰[19],并因“互锁”效应导致冰的粘附强度剧增[20]。甚至,除冰过程往往会破坏这些精细的微纳结构,进而使 SHBS 失去其超疏水性[21-22]。

为了克服这一难题,科研人员近年来在热性质材料的研发上取得了系列进展,他们巧妙地将光热材料[23-24]与超疏水表面相结合,开发出一种全新的功能性超疏水表面。这种表面通过保持其温度始终高于冰点,不仅能够有效地延缓冰的形成,而且能够直接融化已形成的冰或霜[25-26]。这一创新设计不只赋予了材料卓越的防冰与除冰性能[27-29],更是克服了传统超疏水材料在极端环境下的局限性,因而被视为新一代的疏冰材料。本文对光热防冰超疏水表面进行系统阐述,以期为建筑、交通、能源、电力等行业防冰工作提供参考。

1　固体表面结冰机理

在低温条件下,水蒸气在固体表面凝结并转化为冰的过程,即表面结冰,是一个复杂且受多种因素影响的物理现象。其核心在于水分子在固体表面上的吸附、冷凝、聚集以及最终形成冰晶的过程。首先,当水蒸气分子接触到低于其饱和蒸汽压的固体表面时,会因失去能量而吸附在固体表面形成一层薄薄的液膜或微小的液滴。这一过程显著受到固体表面特性的影响,如粗糙度、润湿性和化学组成等。粗糙的固体表面提供了更多的吸附位点,有利于水蒸气的快速凝结。而后,随着温度的进一步降低,这些液滴中的水分子开始有序排列,形成冰晶的初始结构,这一过程被称为冰晶成核。成核机制主要分为均相成核和异相成核两种。均相成核发生在纯净的液滴中,需要克服较大的能量势垒,通常在更大的过冷度下才能发生;而异相成核则更容易在含有杂质或缺陷的液滴中发生,因为这些区域提供了较低的成核势垒,使得在较小的过冷度下也能形成冰晶核。冰晶形成后,随着温度的持续降低和时间的推移,冰晶会逐渐生长并扩散到整个液滴中,最终形成完整的冰层[22]。图 1 所示为冰晶形成过程。这一过程受到多种因素的影响,包括温度梯度、湿度、压力以及表面特性等[22, 30]。综上所述,表面结冰机理是一个涉及多个步骤和因素的复杂过程。深入理解这一过程对于开发高效的防冰和除冰技术,以及优化相关工业设备的性能具有重要意义。

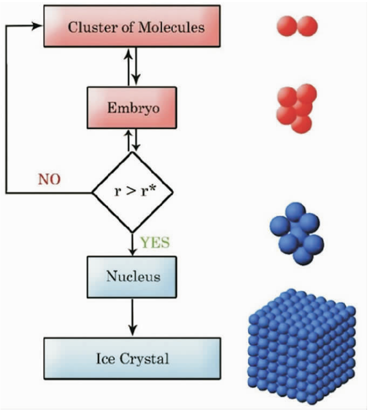


图 1 冰晶形成示意图(r 和 $r*$ 分别为胚胎的半径和临界半径)[22]

Fig. 1 Schematic diagram of ice crystal formation (r and $r*$ are the radius and velocity of the embryo, respectively Boundary radius)[22]

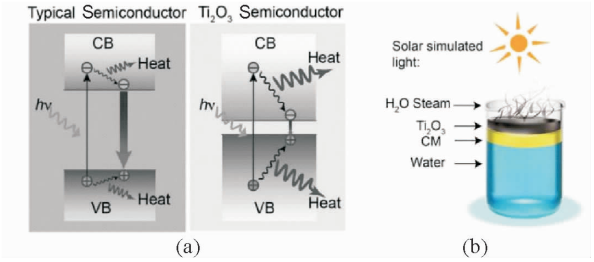
2 光热材料

太阳能以其清洁性、可持续性和丰富性等优点而展现出替代传统能源的巨大潜力。它既可以被直接利用,也可以转换为其他形式的能源,如光热转换即为光能转换为热能[31-32]。

为了满足实际应用的需求,理想的光热材料[33-35]具备在整个太阳光谱范围内的高吸收率,并具备高效的光热转换效率。光热材料能够有效地将太阳能转换成热能,并在除冰领域得到了创新应用[36-37]。利用其光热特性,这些材料能够捕获太阳热量来防/除冰,从而缓解恶劣环境中冰层积聚所带来的问题。

2.1 无机光热材料

无机光热材料按维度可分为零维、一维、二维和三维。零维主要包括纳米粒子,如金纳米粒子具有表面等离子体共振效应,能够在特定波长光照下高效地将光能转化为热能,在癌症光热治疗、光热防冰表面和光热发电等领域中有着广泛应用[38-40];一维比如碳纳米管和纳米线,碳纳米管导热和光吸收性能优异,可将光能转化为热能并迅速传导,如可以与超疏水材料复合,赋予其光热转化能力,而纳米线如硅纳米线可用于太阳能电池等领域;二维通常指纳米片和石墨烯等,石墨烯载流子迁移率和光吸收能力极高,其光吸收和热传导性能可通过控制厚度和层数进行调节[41-43];三维通常具有复杂结构和较大比表面积,能够有效地吸收和散射光线,提高光热转换效率,如多孔金属氧化物、金属有机框架材料等[44-45],Wang 等[46]首次展示了 Ti_2O_3 纳米颗粒的高性能光热转换特性,图 2(a)为典型半导体和 Ti_2O_3 半导体光激发过程示意图,图 2(b)为 Ti_2O_3 光催化水蒸发中的应用[46],证实了 Ti_2O_3 纳米颗粒优异的光热性能。



(a) (b)

图 2 Ti_2O_3 纳米颗粒与传统半导体对比与应用设计示意图

Fig. 2 Ti_2O_3 nanoparticle and traditional semiconductor comparison and application design diagram

注:图 2 中,(a)为正常半导体(左)和窄带隙 Ti_2O_3(右)中电子空穴的产生和弛豫示意图;(b)为太阳能水蒸汽蒸发设计示意图,其中纤维素膜(CM)负载 Ti_2O_3 纳米颗粒[46]。

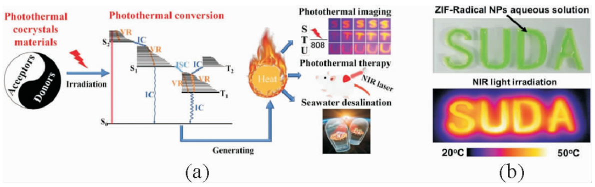
2.2 有机光热材料

有机光热材料主要包括有机小分子染料、超分子复合物和共轭聚合物等。有机小分子染料如吲哚菁绿、普鲁士蓝、噻二唑衍生物等在近红外区吸收强、摩尔消光系数高,能有效吸光转化为热能,其中,普鲁士蓝的摩尔消光系数与纳米金属粒子同一数量级,比碳纳米管、硫化铜等光热转换剂高 2~3 个数量级且光热转换稳定性好[47-48];超分子复合物以卟啉环为代表,由双层卟啉分子和磷酸脂质体自组装而成,卟啉分子密度高,消光系数大,光吸收和光热转换性能优异;共轭聚合物通常由有机小分子单体氧化聚合得到,如聚苯胺、聚吡咯、聚噻吩和聚多巴胺等,在近红外区光吸收性能好,共轭结构促进光生载流子产生和传输实现高效光热转换[49-50]。通过对共晶体材料的光热机理研究(图 3)。Li 等[43]将有机自由基分子铆钉进金属有机框架纳米颗粒(Zeolitic Imidazolate Framework-8,ZIF-8)中,使其在水中稳定存在且光热转换效率高,体外抗菌研究证实其近红外光照射下抗菌活性高效[51]。

2.3 复合光热材料

复合光热材料是将不同种类的光热材料或光

热材料与其他功能材料进行复合,以综合各组分的优势,实现性能的优化和功能的拓展[52-54]。如图 4 所示,Wei 等[55]利用激光加工聚酰亚胺薄膜的方法,制备出石墨烯/氧化铜复合材料,这种亲水性的石墨烯光热涂层在 1 个太阳照射下获得了 2.54 kg/m^{2}h 的蒸发速率,有望用于海水淡化和废水处理。这种将金属纳米颗粒与石墨烯复合的方法,可以结合金属纳米颗粒的局域表面等离子体共振效应和石墨烯的高比表面积、热导率良好等优点,极大地提高光热转换效率和热量传递性能,使得防冰表面温度迅速上升,有效地防止表面结冰[56]。

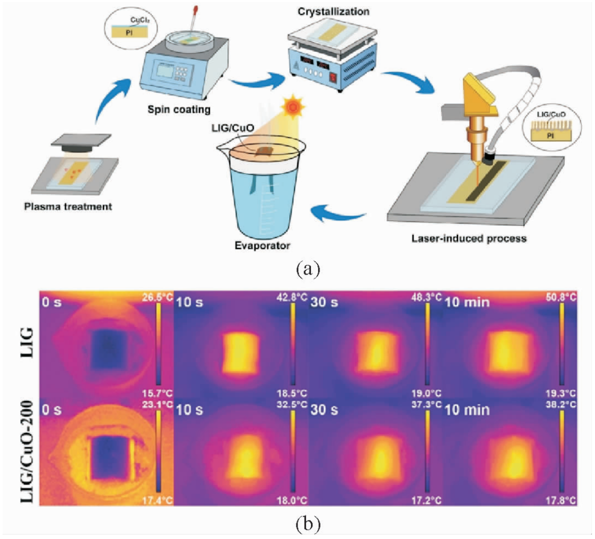


(a)　(b)

图 3　有机分子材料光热转换设计示意图

Fig. 3　Schematic diagram of photothermal conversion design of organic molecular materials

注:图 3 中,(a)为共晶材料的光热机理及应用示意图[50];(b)为注入 SUDA 图案的 ZIF-自由基纳米粒子水溶液的照片及其在 808 nm 激光照射下的相应红外热图像[51]。



(a)

(b)

图 4　石墨烯/氧化铜复合材料工艺流程与光热性能对比图

Fig. 4　Graphene/copper oxide composites process flow and photothermal properties comparison

注:图 4 中,(a)为用连续波 CO$_2$ 激光在不同浓度 CuCl$_2$ 溶液包覆的 PI 膜上制备激光诱导石墨烯/CuO 复合材料的工艺;(b)为 LIG 和 LIG/CuO-200 分别在照射 0 s、10 s、30 s 和 10 min 时的红外图像[55]。

3　光热超疏水表面

3.1　光热超疏水表面的制备

光热超疏水表面的制备通常是以超疏水表面为基础,将具有光热效应的光热材料以特定的方式与传统超疏水表面结合,使其具有光热效应。

受壁虎皮肤和猪笼草启发,Wang 等[57]采用一步激光法制备基于碳纳米管复合材料的微纳米分级结构超疏水光热表面(Superhydrophobic Photothermal Surface,SH-PS),通过注入硅油得到超滑光热表面(Slippery Photothermal Surface,S-PS)。该表面在多项性能测试中,包括静态和动态抗冰性能测试、光热除冰性能测试以及液滴操控能力测试,都表现出十分优异的性能。

Zhang 等[58]则是用化学蚀刻法在 AZ31 镁合金表面制备等离子蚀刻黑色陶瓷作为基层,负载聚二甲基硅氧烷(Polydimethylsiloxane,PDMS)纳米粒子(PDMS NPs)作为表层的复合涂层,以实现超疏水和光热防冰。该涂层在 200 mW/cm^{-2} 照射强度,温度可迅速升至 69.4 ℃,具有理想的光吸收和光热性能。而 Si-O-Si 的微观结构和极性键提供了优异的超疏水性,使表面接触角达到 156°。

如图 5a 所示,Wu 等[59]运用一种极为简便、高效的方式制备并收集蜡烛烟灰,将收集的烟灰用其他的光热材料进行掺杂或复合,提高其光热转化效率。然后涂覆二氧化硅壳增强,接枝低表面能聚二甲基硅氧烷(PDMS)使其表面超疏水。结果表明,在 −50 ℃的环境温度下,表面不会结冰。该方法是基于廉价的蜡烛烟灰制备光热超疏水表面,并利用蜡烛烟灰提供了层状微纳米结构和光热性能。

另外,也可采用喷涂、旋涂等方法,将含有碳粉、碳纳米管、石墨烯等碳材料的超疏水涂层溶液喷涂到基材上,从而获得具有防冰性能的光热超疏水表面[60-61]。如图 5b 所示,Wang 等[62]通过化学气相沉积等方法制备多孔石墨烯材料,将其与低表面能物质和粘结剂等混合均匀,形成复合涂料,并以喷涂、旋涂或浸涂等方法将复合涂料涂覆在基底表面,经过干燥、固化等工艺,在基底上形成多孔石墨烯基光热超疏水表面。如图 5c 所示,

Wei 等[56]基于十八烷基三氯硅烷的水解缩合反应,通过掺杂碳纳米管制备涂层溶液,由此形成的涂层有着独特微观结构,表面存在大量纳米级凹凸结构,配合低表面能的硅烷链,使涂层具备超疏水性能。然后将其喷涂在固体基材上,从而得到了光热超疏水表面。

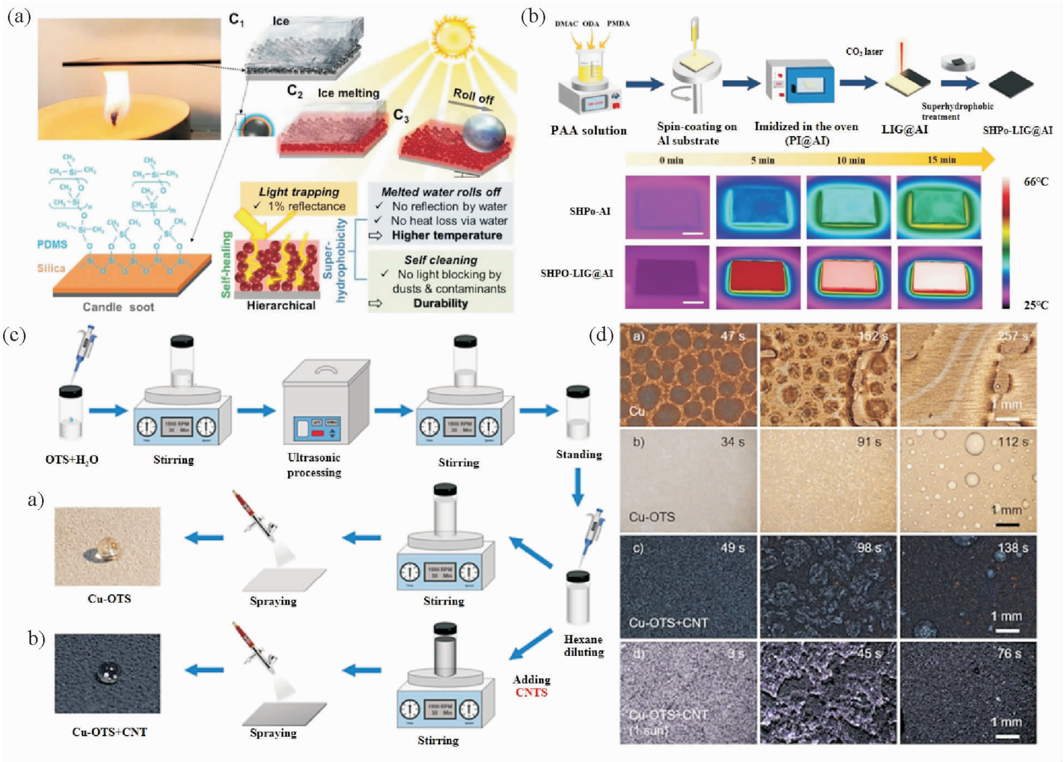


图 5　多种方法制备具有光热效应的超疏水涂层示意图

Fig. 5　Schematic diagram of preparing superhydrophobic coatings with photothermal effect by various methods

注:图 5 中,(a)为基于蜡烛烟灰的超疏水光热防冰表面的制备及原理图[59];(b)为 PI@ Al 和 SHPo-LIG@ Al 制造工艺示意图与 1 个太阳照射下的光热性能对比[62];(c)为超疏水涂层和光热超疏水涂层的制备方案;(d)为其他基底与 Cu-OTS 基底的光热性能测试对比[56]。

3.2　光热超疏水表面防冰进展

在使超疏水涂层具有光热效应的过程中,通常将复合的光热材料按照其光热转换机制的不同,大体可分为金属纳米材料、半导体材料和碳基材料。金属纳米材料因其高效的光热转化效率、高强度和优异的热稳定性被认为是良好的光热转换材料[63-64]。而金和银纳米粒子常被用于为超疏水表面提供光热效应。图 6a 所示的制备过程为 Wang 等[65]利用银镜反应制备超疏水棉线,借助其超疏水特性以及良好的光热转换能力,在近红外光照射下,通过光热转换使油中的水蒸发,以此辅助实现高效(分离效率达 99% 以上)的油水分离,为油水分离提供了新的思路与方法(图 6b)。如图 6c 所示,Tao 等[66]将 Au/TiO_2 复合材料作为等离子体光热材料沉积在聚四氟乙烯(Polytetrafluoroethylene,PTFE)表面,用于光热超疏水涂层,在 300 mW/cm^2 光照下,等离子体光热膜的温度上升 25 ℃以上,具有良好的防/除冰性能。但金银等粒子因其价格高昂,限制了实际使用的可能性。因此,铝、铜等粒子被广泛应用,通过特定方式的改性或复合后,在表现出超疏水性的同时,也能赋予表面优异的光热转化性能[55, 67]。

相比于金属粒子,基于半导体的光热转化材料宽光谱吸收特性、成本较低等优点且光热性能并不弱于金属粒子。Xiang 等[68]通过将 GO@Fe_3O_4 混合 PDMS/PF 润滑剂注入纳米草 CuO(PSSS)中,得到一种防/除冰表面(图 7c)。由于润滑油的低表面能和 GO@ Fe_3O_4 的光热效应,该表面不仅在延迟结冰性能上表现优异且还具有较高的光热转化效率。如图 7d 所示,在红外照射下,表面温度在 120 s 内急剧上升至 71.7 ℃,使得冰和润滑剂在高温下迅速融化。Ma 等[69]利用氮

化钛和聚四氟乙烯(TiN-PTFE),制备一种光热超疏水薄膜用于防冰和除冰,等离子体 TiN 纳米棒具有局部表面等离子体共振(Localized Surface Plasmon Resonance,LSPR)性能,将光能转化为热能,而低表面能的 PTFE 则最大限度地提高了超疏水性。图 7a、图 7b 为其防冰与光热性能的展示,半导体基光热材料的光生载流子复合、量子效率较低等问题也不可忽视[70]。

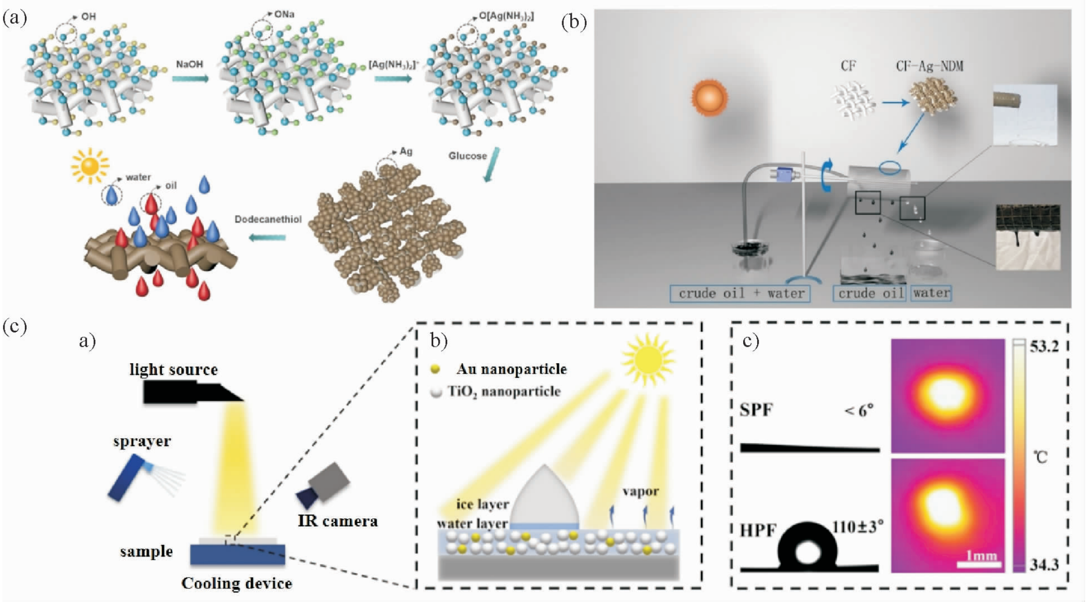


图 6　金银粒子在光热超疏水领域的工作原理及性能对比示意图

Fig. 6　Schematic diagram of working principle and performance comparison of gold and silver particles in the field of photothermal superhydrophobicity

注:图 6 中,(a)为 CF-Ag-NDM 的制备及其在阳光下的油水分离的示意图;(b)为 CF-Ag-NDM 在光照下的工作机理示意图[65]。(c)中,a)为实验系统草图;b)为除雾除冰等离激元光热过程草图;c)为两种不同等离激元光热膜在 3 个太阳照射下水滴的静态接触角和温度分布[66]。

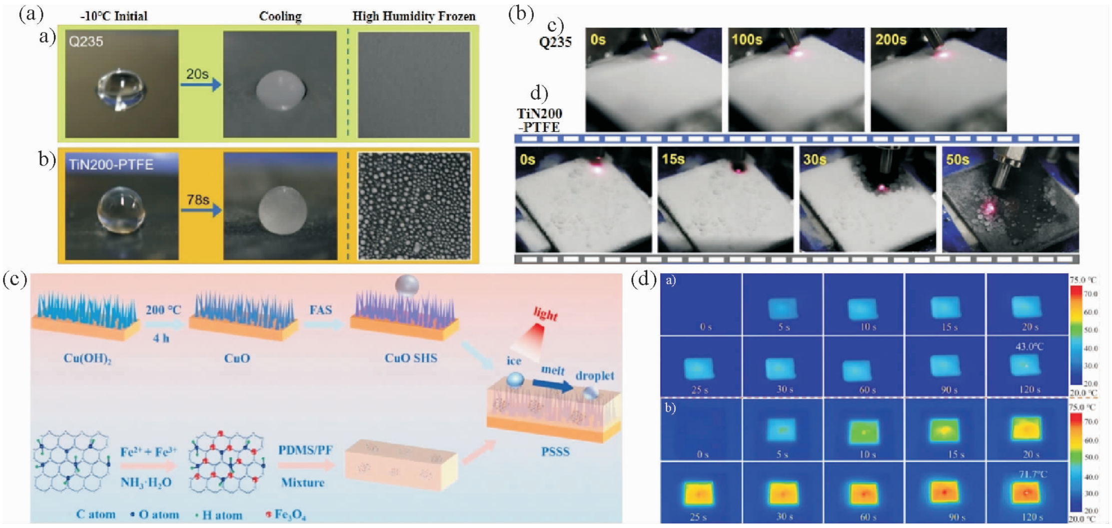


图 7　光热超疏水涂层与传统超疏水涂层性能对比及工艺流程示意图

Fig. 7　Comparison of performance and process diagram of photothermal superhydrophobic coating with traditional superhydrophobic coating

注:图 7(a)中,a)为裸 Q235 基片和 b)为 TiN200-PTFE 表面冻结前后的水滴照片;图 7(b)中,c)为裸 Q235 基片和 d)为 TiN200-PTFE 表面光热除冰过程的光学图像[69]。图 7(c)为 PSSS 制作过程的示意图;图 7(d)中,a)、b)分别为 GO 和 GO70@ Fe3O4 混合 PDMS/PF 注入 PSSS 的光热性能[68]。

碳基材料在解决杂质含量高、粒度难以控制、碳纳米管容易团聚等缺点后,以其独特的结构、优异的化学稳定性和较低的使用成本,可成为光热转化应用的最理想材料之一。Xie 等[71]通过结合碳材料的黑体特性和微纳分层结构,并利用电化学沉积和硅烷化处理方法制备了低成本、耐用、高效的光热超疏水材料。图 8c、图 8d 所展示的不同表面对比中,在 100 mW/cm^2 的太阳光照射下,表面温度可升至 90 ℃,结冰延迟时间长达 3 600 s。如图 8a、图 8b 所示,Hu 等[72]将有机金属框架(Metal-Organic Framework,MOF)与石墨烯通过水热合成等化学合成方法,将两种物质复合得到了 MOF/石墨烯气凝胶(MEGA),其水接触角(Water ContactAngle,WCA)为 152.7°,且在光照下,MEGA 表面温度可迅速上升至 80 ℃。

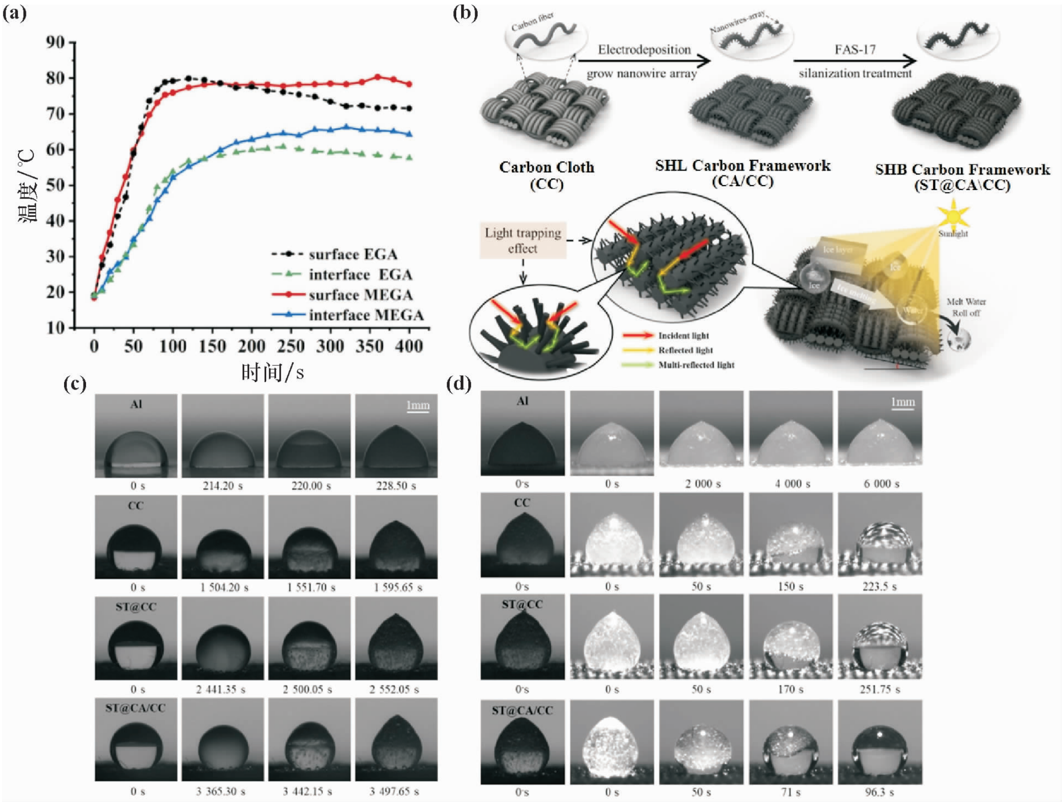


图 8 碳基材料在光热超疏水涂层应用中的性能对比及工作原理示意图

Fig. 8 Comparison of properties and schematic diagram of working principle of carbon-based materials in the application of photothermal superhydrophobic coating

注:图 8 中,(a)为光照射下 EGA 和 MEGA 的表面和界面温度[72];(b)为光热超疏水涂层的制备、光热效应原理、光热超疏水涂层的除冰机制;(c)和(d)分别为铝、CC、ST@ CC 和 ST@ CA/CC 表面上水滴的结冰过程和光热除冰过程[71]。

在上述讨论中,由图 5d、图 7、图 8 所展示的除冰过程中及数据可见,在延迟结冰与除/防冰等性能上,依托于光热材料复合的超疏水涂层相比传统超疏水涂层表现更加优异。

4 光热超疏水防冰机理

考虑到传统超疏水表面在极端条件下存在的不足,越来越多的团队将光热材料与超疏水表面进行集成[73-74]。

超疏水表面因其高接触角、低滚动角和抗粘附性能而成为解决结冰问题的有效方法。微/纳米级的超疏水表面确保水滴保持在 Cassie-Baxter 湿润状态,从而产生对水的排斥效果。因此,微纳结构的设计对于超疏水表面至关重要,如图 9a、图 9b 所示依据相关模型,通过特定结构增大接触角、减小滚动角,增强疏水性能,实现自清洁;同时,还能增加涂层附着力与耐磨性,提升机械稳定性[75-76]。此外,这些结构形成热障,能抑制热传递,延缓界面处的非均相成核,从而延缓了冰的形

成[77-78]。但在极端环境下(极寒高湿),普通超疏水表面往往会失去其正常的疏水、防冰等性能,甚至会造成冰附着强度增大。

对于具有光热效应的超疏水表面,在太阳光照下,表面温度能以更快的速度升至冰点以上,有效地阻止表面液滴凝结和润湿状态转变,使其在极端寒冷且高湿的条件下仍保持超疏水性和防/除冰性能。太阳能光子转化为热能的光热除冰是低碳、高效、可持续的解决方案,能克服超疏水被动除冰的部分缺点。此外,提高表面材料吸收率、设计多孔结构可促进光子吸收利用以增加热量转化[42,75-79]。Zhao 等[80]研究表明,构建在有限空间内重复反射光的微结构和多孔结构可以提高表面的吸光能力,并且在低太阳照度下实现更大的表面温升,从而具有更强的自适应防冰功能。

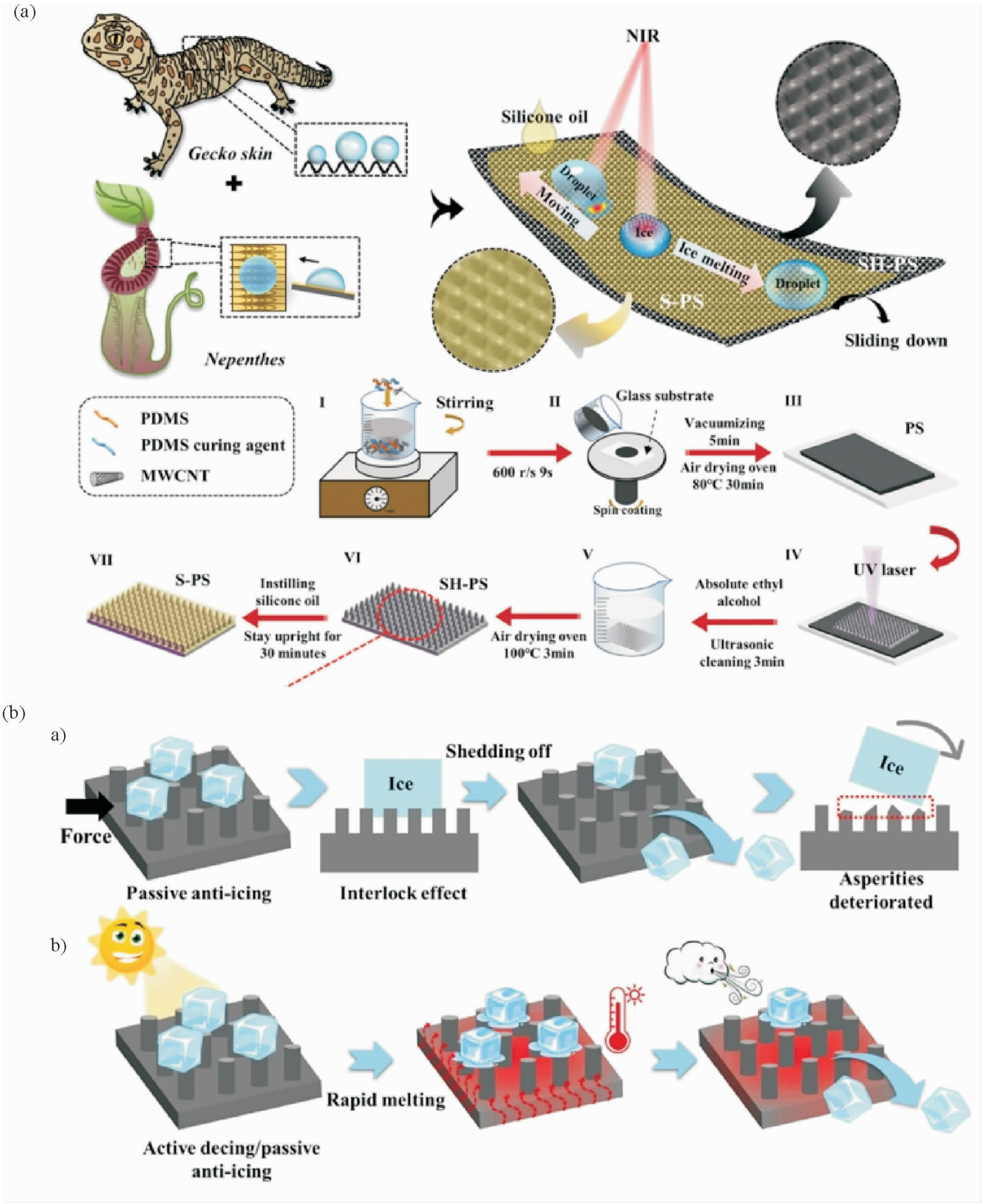


图 9　光热超疏水涂层工作原理及工艺流程示意图

Fig. 9　Schematic diagram of working principle and process flow of photothermal superhydrophobic coating

注:图 9 中,(a)为 s-ps 的原理图及制备过程[57];(b)中,a)和 b)分别是被动和主动除冰/被动防冰表面示意图[75]。

5 总结与展望

本文首先概述了固体表面结冰机理,随后对光热材料与传统超疏水涂层的制备方法进行简要总结,基于上述内容,归纳了具有光热效应的防冰超疏水涂层的制备方法,阐述了传统超疏水涂层与光热材料复合的优异性能,最后简要概述了光热超疏水防冰机理。

超疏水涂层无疑是防护大型工业设备和设施的重要选择,然而普通超疏水表面在极端条件下(极寒高湿)难以持久发挥其防冰作用,而具有光热效应的超疏水表面依靠自身光热效应,不仅改善了传统超疏水表面被动防冰的能力,甚至填补了主动防冰功能的空白,使其在多个领域具有广阔的应用前景。但目前光热防冰超疏水的实际应用还有待提升,笔者认为以下3个难点值得探索:①以更加简便、高效的方式大规模制备具有光热效应的超疏水涂层。传统光热超疏水表面制备过程十分复杂,不仅步骤繁多,操作也极为不易,因而难以高效且大规模地制备。②开发高稳定性、经济性的复合光热材料。目前的复合超疏水涂层往往会导致光热材料复合后稳定性变差,甚至几乎失去光热效应。如何解决这一问题,成为光热超疏水材料能否工程化应用的关键因素。③建立一套光热超疏水涂层综合衡量标准。目前的研究中,大多数实验团队对性能测试方法、结果分析难以完全一致,从而导致评估结果不能得到统一,严重影响了行业的相互交流与进展。因此,推动设立一套衡量标准,对于实现定量与定性表征十分关键。

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