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滨海环境下水泥基材料有机硅防护涂层的研究进展

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摘要:有机硅涂料通过对侵蚀介质的传输抑制作用以及诱导结晶作用,赋予了水泥基体优越的表面防护效果,显著提升了混凝土结构的耐久性能与安全可靠性.本文归纳了有机硅涂料的分子结构特征及其对胶凝材料的作用机理,全面阐述了有机硅涂料对水泥基材料耐久性能的提升效果,探讨和展望了有机硅涂料在工程应用中存在的主要问题以及未来发展趋势,旨在为水泥基材料耐久性防护的深入研究与应用提供重要的理论指导与工程参考.

关键词:有机硅涂料;水泥基材料;表面防护;结晶作用;抗侵蚀

中图分类号:TQ324.2^{+}1 **文献标志码**:A **doi**:10.3969/j.issn.1007-9629.2024.04.007

Research Progress on Silane Protective Coatings of Cementitious Materials in Coastal Environment

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Abstract: Silane coatings provide a variety of superior surface protection to cement substrates by corrosion transport inhibition and crystallization induction, improving the durability and safety reliability of concrete structure. The physical and chemical properties of silane coatings, the binding mechanisms for cementitious materials were summarized, while the effectiveness of silane coatings in improving the durability of cementitious materials were comprehensively reviewed. Additionally, the main issues and future development trends in engineering applications were analyzed and proposed, aiming to provide significant theoretical guidance and engineering reference for the in-depth research and application of durability protection of cementitious material.

Key words: silane coating; cementitious material; surface protection; crystallization; corrosion resistance

中国海洋强国战略的提出极大地促进了广大沿海地区基础设施建设的快速发展.混凝土作为一种被广泛使用的复合胶凝材料,其耐久性决定了滨海环境下混凝土结构的服役寿命与安全性.混凝土的耐久性受到各种恶劣自然环境的制约,包括海水侵蚀、碳化、冻融破坏、紫外老化、风化作用和海浪冲刷等[1-2].有机硅涂料表面处理以其防护性能优越、施工便捷与应用范围广的显著特点,已经成为近年来水

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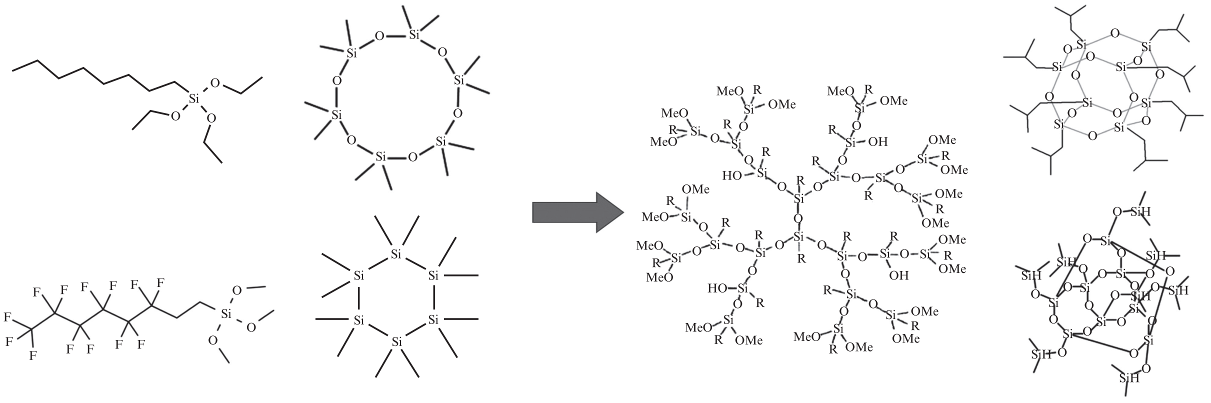
泥基材料耐久性提升领域的研究热点之一.硅烷作为一种渗透反应型的高分子聚合物涂料,水解后生成的聚硅氧烷链可以通过羟基间缩合反应和氢键作用与水泥水化产物(主要为水化硅酸钙(C-S-H))形成较强的物理与化学结合,从而在水泥基材的表面、毛细孔与凝胶孔道内部形成长效稳定的疏水吸附层与抗渗结晶体[3-4],抑制各种侵蚀介质与物理化学因素对水泥水化产物微结构的持续性破坏[5].有机硅涂料渗透进混凝土后会在水泥基体与界面过渡区(ITZ)形成完整的聚合物保护膜,增强粗、细骨料与水泥浆体之间的黏结作用.近年来,有机硅涂料已经被广泛地应用于海岸工程、水利工程、民用与工业建筑工程、交通建设和环境保护等各个领域.

1　硅烷的分子结构特征

硅烷分子以Si—O键为主链,其侧链通常为烷基或芳香基团.根据其空间排列方式或侧基的不同,线性硅氧烷与环状聚硅氧烷分子可以聚合形成不同相对分子质量的立体异构聚合物,包括交联聚硅氧烷与笼形聚硅氧烷等,如图1所示.



Linear siloxane　Ring siloxane　Crosslinked polysiloxane　Cage polysiloxane

图1　硅烷分子结构示意图

Fig. 1　Schematic diagram of silane and siloxane molecular structure

硅烷分子中硅原子的电子云较为松散,其与水解后形成的羟基键角为109.5°,并且由于硅羟基具有较高的松弛特性,因而通过取代反应形成带电离子或偏极性分子的势垒相对较低.在无定型态的硅氧烷分子中,以硅原子为中心的稳定分支结构与羟基自由端随着溶剂的挥发固化聚合形成了立体网状交联结构.聚硅氧烷分子中Si—O键的键能为452 kJ/mol,远高于其他聚酯类涂料中C—C键的356 kJ/mol[6].聚硅氧烷交联网络中的Si—O—Si键具有高达130°～160°的键角与0.164 nm的键长[7],其分子链能够形成自由内旋,并具有极高的柔顺性.聚硅氧烷分子间相互作用力小且其处于饱和配位的硅原子很难发生自发分解,因而具有良好的化学惰性与粒子凝聚能力[8].

2　有机硅涂料对胶凝材料的防护机理

2.1　硅烷对侵蚀介质的传输抑制作用

硅烷分子在渗透过程中水解形成的羟基回转半径很小,能够保证烷烃链的自由舒展与定向排列,显著降低了聚硅氧烷界面的偶极矩并产生了强疏水作用.聚硅氧烷分子以静电相互作用为驱动力,以单分子吸附的形式结合在水泥水化产物上[9],同时水泥的水化反应促进了界面聚硅氧烷片段的融合(见图2).

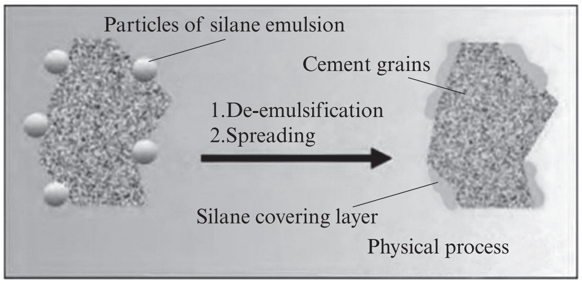


图2　聚硅氧烷组分在水泥水化产物的表面吸附与融合

Fig. 2　Surface adsorption and fusion of polysiloxane components with cement hydration products[10]

聚硅氧烷层中Si—O—Si键的带宽广且电阻性强[11],能够有效抑制侵蚀阶段阴极与阳极之间的电子隧穿,形成完整的侵蚀介质扩散屏障.聚硅氧烷分子具有较低的储能模量与很强的界面定向诱导能力,促使凝胶孔道壁上形成了有序沉积的疏水膜[12],这种分子级别的屏障具有非常高的化学稳定性和孔径适配性[13].C-S-H凝胶孔道中聚硅氧烷段的空间分布具有短程随机性,增大了基体孔隙的体积分形维数与迂曲度,延长并阻断了侵蚀介质在混凝土中的

传输路径[14].

2.2　硅烷在胶凝材料内部的渗透结晶作用

聚硅氧烷分子渗透进水泥基体中，与C-S-H、$CaCO_3$、$Ca(OH)_2$等水化产物通过二次水化反应形成不溶性硅酸钙螯合物[15]，进而填充基体表层的孔隙与微裂隙，提高了水泥基材料凝胶的结构密实性．聚硅氧烷分子中的羟基与水化产物中硅氧四面体的氧原子形成氢键，并与Na^+、Ca^{2+}、Al^{3+}形成配位键，促使游离的硅酸盐单体聚集在聚硅氧链周围参与成核反应[16]．聚硅氧烷分子与$Ca(OH)_2$、$Al(OH)_3$反应形成的硅酸盐聚合物具有较小的粒径，能够在毛细孔隙中作为成核位点不断生长成致密的硅酸盐吸附层，导致基体中大口径贯通孔比例的降低与封闭孔比例的提高．硅氧烷分子通过桥接水泥基体与混凝土中的粗、细骨料，改善了界面过渡区的微结构完整性，如图3所示．

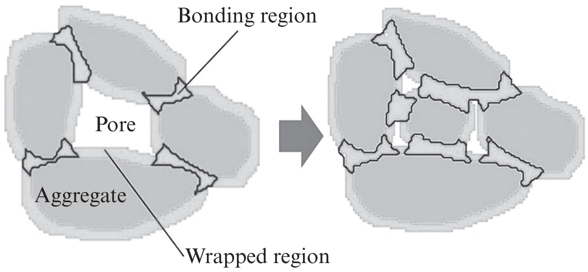


图3　聚硅氧烷对水化产物的孔隙封闭与结晶诱导
Fig. 3　Pore sealing and crystallization induction of hydration products by polysiloxane[17]

同时，硅烷水解生成的水分子参与硅酸三钙(C_3S)和硅酸二钙(C_2S)的水化反应，聚合为具有连续桥接位点的硅酸盐结晶层．硅烷单体在水泥碱性环境下会形成化学梯度势较大的线形硅氧烷聚合物[18]，这有利于Ca^{2+}与Al^{3+}在硅氧链上的吸附结合，并改善硅酸盐聚合物的链段缺陷和应力集中．聚硅氧烷诱导下的硅酸盐产物是一种典型的具有较高断裂韧性的有机-无机交联网络[19]，不仅可以降低侵蚀介质在水分子团中的析出和吸附，还可以抑制各种降解作用下晶体表面颗粒的松动和位移．

3　有机硅涂料对水泥基材料的防护性能

3.1　防水性能

优越的防水性能与抗渗性能是有机硅涂层的重要优势．C-S-H凝胶孔道中水分子的传输形态类似于半月板形[20]，其传输规律符合Lucas-Washburn方程．孔壁上的聚硅氧烷团簇正是通过阻断水分子与硅酸盐链非桥接氧之间的氢键作用来构筑防水层[21]，进而降低扩散中的水分子团内部的位移变化梯度．硅烷憎水层能够显著降低水分子在快速渗透阶段的扩散驱动力，导致水泥基体毛细孔隙内部出现毛细逆气压[22]，致使硅酸盐凝胶的毛细吸水作用过早地进入到稳定渗透阶段．张文娟等[23]研究发现，水分子的传输会在聚硅氧烷团簇的黏附力与排斥力的作用下得到显著抑制[24]（见图4)，并且水分子的渗透深度会随着聚硅氧烷中羟基单元游离程度的提高而减小[25]．相关研究表明[26-27]，有机硅涂料处理后水泥基材料的静态毛细吸水量与渗流速率至少降低了80%．水泥基材料的水灰比越低，硅烷涂覆量越高，有机硅涂层的防水提升效果越显著[28]．

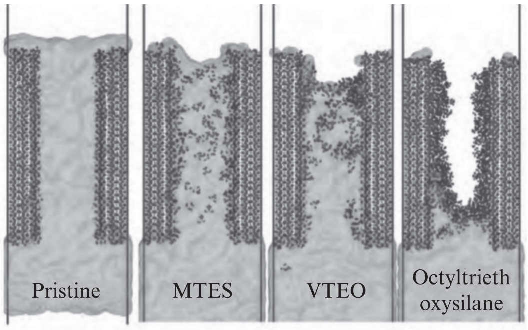


图4　有机硅涂料作用下凝胶孔道中水的传输抑制
Fig. 4　Inhibition of water transport in gel channels with treatment of silane coating[24]

聚硅氧烷诱导下形成的二次水化产物能够堵塞胶凝材料表面的部分毛细孔与微裂缝，阻碍水的润湿与饱和渗透[29]．喻建伟等[9]发现有机硅涂层降低了水泥基体中连通性毛细孔的体积率，并提高了凝胶孔的体积率．聚硅氧烷通过交联反应形成的硅酸盐矿物具有团簇状的形貌特征，其作为成核位点会增大硅酸盐晶粒尺寸，并为后续的二次水化反应提供了沉积模板[30]．此外，硅氧烷可以与钙矾石等铝硅酸钙矿物缩合反应生成水化硅铝酸钙(C-A-S-H)凝胶[31]，通过补偿体积膨胀引起的微裂缝伸展来提高水泥基材料的抗渗性．

3.2　抗氯盐侵蚀性能

滨海环境下进入水泥基材内部的氯离子一部分被化学结合并生成Friedel’s盐，一部分被物理吸附继而引起C-S-H凝胶层间界面电荷平衡的紊乱[32]．氯离子极易在混凝土表面与界面过渡区富集，水泥基体内外的饱和梯度相差越大，氯离子越容易向毛细孔内部渗入[33]．有机硅涂层通过疏水以及表层密实作用抑制了毛细压力作用下氯离子向微孔隙中的迁入[34]，使氯离子在水泥基体中的扩散系数显著降低，如表1所示．

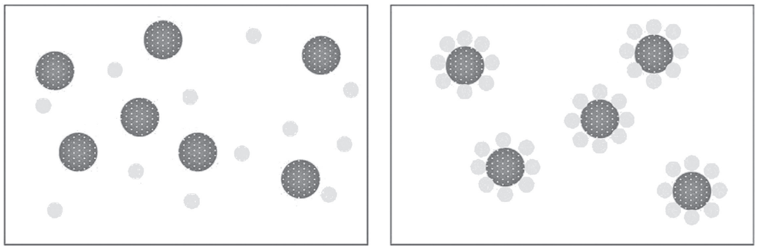
表1　有机硅涂料处理后混凝土材料的抗氯盐侵蚀性能

Table 1　Chloride resistance of concrete treated with silane coating

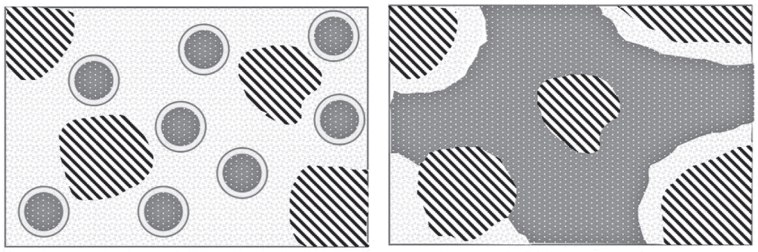
Coating type	Concrete strength	Curing age/d	Chloride ion diffusion coefficient decrease/%	Reference
Conventional polysiloxane coating	C30	28	72.20	[35]
	C40	28	31.63-89.06	[26, 36-39]
	C45	28	85.00	[40]
	C50	28	33.67-88.78	[26-27, 35, 37-39]
	C60	28	58.40	[41]
Nano-modified silane coating	C40	28	72.64-87.61	[34, 42-44]
	C50	28	83.34-85.87	[27, 34]
Siloxane copolymer coating	C30	28	83.30	[35]
	C40	28	66.76-88.97	[29, 32, 37-39, 42, 44-45]
	C50	28	70.45-85.94	[32, 35, 37-39, 45]
	C60	28	50.79-78.48	[41]

快速氯离子迁移试验表明,防水抗渗效果越好的有机硅涂料,其表面处理后的混凝土抗氯离子侵入的效果也越好[36].聚硅氧烷抗渗膜提高了侵蚀产物形成的反应能量势垒,维系了C-S-H凝胶层状结构的稳定性.

张馨元[27]指出,有机硅涂层的抗氯盐侵蚀性能主要来源于硅烷的渗透结晶作用.有机硅涂料处理后水泥基材内部氯离子的传输路径被延长或阻断,同时聚硅氧烷分子通过诱导硅酸盐晶核的形成降低了水泥基体内连通孔隙的比例[46],并大幅降低了混凝土内部的电荷迁移系数[47].聚硅氧烷分子对水泥水化反应的促进作用,等同于提高了水泥基体中氯离子的扩散衰减系数[48],限制了硅酸盐结晶层界面两侧的离子浓度差.Li等[41]认为,由聚硅氧烷网络交联的硅酸盐晶粒能够长期维持胶凝材料内水蒸气的进出平衡,限定了凝胶孔道内氯离子的溶解浓度变化区间.此外,硅氧烷组分通过改善和增强水泥-骨料过渡界面区的结构强度与黏结力[49](见图5),缓解了因硅酸盐层断裂而导致的溶出型破坏.



(a) Just mixed　　(b) Aggregation of polymers



(c) Coverage with polymeric reticulation　　(d) Denser structure

● Cement particle; ○ Polymer; ▨ Aggregate

图5　聚硅氧烷对混凝土中界面过渡区的增强效应

Fig. 5　Enhancement effect of polysiloxane on interfacial transition zone in concrete[49]

3.3　抗硫酸盐侵蚀性能

硫酸盐侵蚀是海洋环境下混凝土材料发生侵蚀破坏的重要原因之一[50-51].一方面,硫酸盐通过脱钙、脱铝反应导致C-A-S-H凝胶的解聚与断裂[52];另一方面,钙矾石、石膏等硫酸盐侵蚀产物通过结晶膨胀引起混凝土的贯穿性开裂[44].有机硅涂层主要基于疏水作用与诱导结晶填充作用,通过抑制和阻断SO_4^{2-}的侵蚀路径与渗透驱动力来维系水泥基体微结构的

完整性.聚硅氧烷抗渗层使得孔隙中SO_4^{2-}的浓度过早达到稳定饱和,并对SO_4^{2-}产生更加显著的位阻排斥效应与黏滞效应.聚硅氧烷网络中尺寸紧密的高支化链段间足以形成对水合硫酸根离子的径向屏蔽[43],有效阻碍了SO_4^{2-}与C-A-S-H凝胶中Ca^{2+}、Al^{3+}的静电吸引.重要的是,聚硅氧烷渗透结晶层具有足够的内聚力,能够有效传递内部积累的膨胀应力,缓解侵蚀产物造成的微裂缝尖端处的应力集中[53].此外,硅烷处理后混凝土内部多为封闭的无害孔或少害孔[54],避免了侵蚀产物局部积累导致的水化产物降解连锁反应.

3.4　对内部钢筋的防护性能

钢筋的耐久性防护包括对对钢筋的锈蚀防护以及混凝土保护层的抗开裂防护[55].首先,有机硅涂层从降低收缩驱动力和提升胶凝材料结构强度两方面提升了混凝土的抗侵蚀性能.聚硅氧烷会在水泥毛细孔道壁上形成连续致密的疏水网络,间接阻断了水、O_2、CO_2与离子的侵入通道,降低了钢筋表面钝化膜的腐蚀电流密度[56].聚硅氧烷通过提升钢筋中相邻锈蚀坑点间水泥基体的密实程度,降低两点间的腐蚀电位差[57],有利于缓解钢筋的电化学腐蚀.张东方等[40]指出,有机硅涂料处理后钢筋钝化膜的侵蚀速率不仅显著降低,并且会更早地进入锈蚀稳定发展阶段,其开路电位能够始终保持在−200 mV以上.此外,聚硅氧烷分子通过水分蒸发抑制、水化热调控及收缩变形补偿降低了混凝土保护层的局部开裂,并利用聚合物网络的桥接作用削弱了钢筋界面处水泥浆体的应力集中与剪切滑移[58].

图6为有机硅涂料处理后混凝土内部钢筋表面形态.由图6可见,硅烷处理后混凝土中钢筋表面的锈蚀情况得到了显著的改善.聚硅氧烷网络对钢筋锈蚀物的结晶膨胀起到约束作用[59],通过减少外围水泥浆体孔隙与缺陷的比例抑制了钢筋表面因侵蚀性离子扩散而产生的电荷迁移.

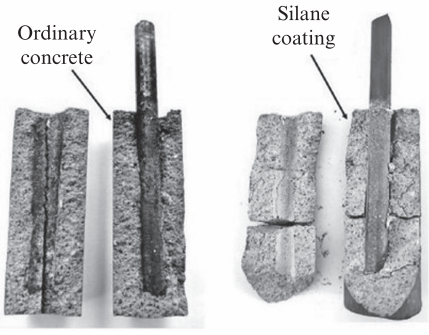


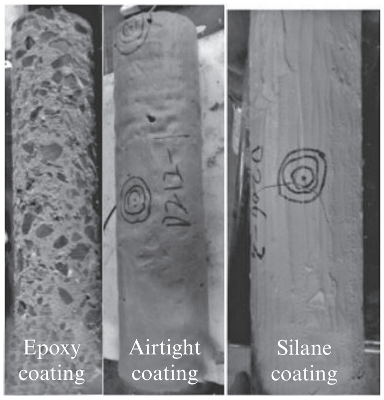
图6　有机硅涂料处理后混凝土内部钢筋表面形态
Fig. 6　Surface morphology of steel bar inside concrete treated with silane coating[40]

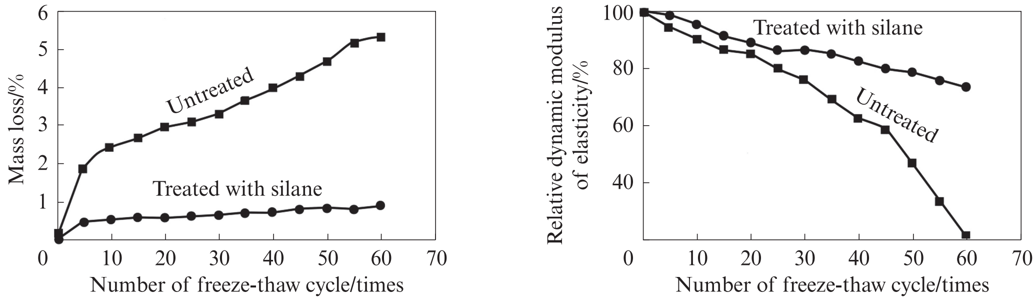
3.5　抗冻融劣化性能

冻融破坏对水泥基材料微结构的影响包括孔隙水结晶导致的体积膨胀、未结冰水向微裂缝浸湿引起的渗透水压、不同水化产物在温度应力作用下产生的损伤破坏[60].有机硅涂层主要围绕改善水泥基材料的孔隙率与孔隙饱水度来提升其抗冻融性能[61].有机硅涂料为水泥基体构筑了具有更高比例闭合小孔的密实结构,通过增大毛细孔渗透水压阈值来显著降低孔隙保水度.同时,赵炜等[62]发现硅烷憎水网络在经历长期冻融循环后仍可以维持混凝土中水蒸气良好的“呼吸性”,使得微孔隙中的毛细压力不至于引起水侵入量的突然增大.赵长勇等[63]也指出,聚硅氧烷结晶沉积层可以同时降低由结冰体积膨胀引发的静水压与冰水蒸气压.朱方之等[64]发现,有机硅涂层在冻融剥蚀初始期对于混凝土材料的防护效果是尤为显著的.相关实验表明,与其他表面屏障型防护涂料相比,经历100次冻融循环后有机硅涂料处理的混凝土试样未发生显著的剥落破坏现象(见图7(a)),有机硅涂料处理后的混凝土材料保持了相对较低的质量损失率与较高的相对动弹模量(见图7(b)、(c)).

冻融循环过程中水泥基体凝胶结构的损伤与断裂属于脆性破坏,而聚硅氧烷交联网络能够优化微孔隙端部的受力状态,通过局部的塑性形变传递结晶膨胀压力[66].聚硅氧烷诱导下的水化产物是一种具有高比热容与低线膨胀系数的有机-无机复合聚合物,能够提高粗、细骨料与胶凝材料之间的界面结合强度,从而降低混凝土材料的温度敏感性以及热量梯度差.与C-S-H凝胶的层状结构相比,聚硅氧烷分子由于内旋程度更高且链段间的氢键作用更弱[67],能够适应冻融循环条件下分子形态的变化以及由于水分迁移和重排所形成的压应力[45],并对水泥基材料微孔隙中冰晶膨胀的发展具有一定的约束作用.此外,高菁[68]认为吸附在凝胶孔道壁上的聚硅氧烷分子团簇会削弱孔隙冰晶与凝胶结构间的黏结力与界面剪切力,并通过位移滑动与摩擦耗能削弱结冰水对毛细孔壁的挤压与剪切作用.

3.6　抗磨性能

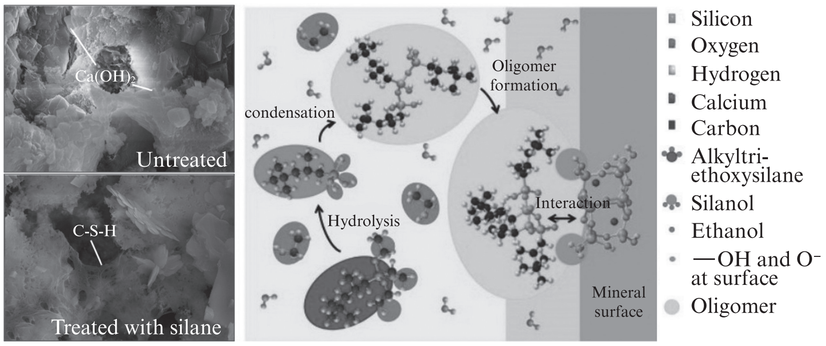
滨海浪溅区海浪的反复冲蚀作用,以及气候变化、风化、酸雨等户外环境因素[33],会引起水泥基材料的表面开裂、剥落以及胶凝物质的降解,从而严重影响混凝土结构的安全性与服役寿命.图8为有机硅涂层抗磨机理示意图.

(a) Sample morphology after freeze-thaw deterioration[65]



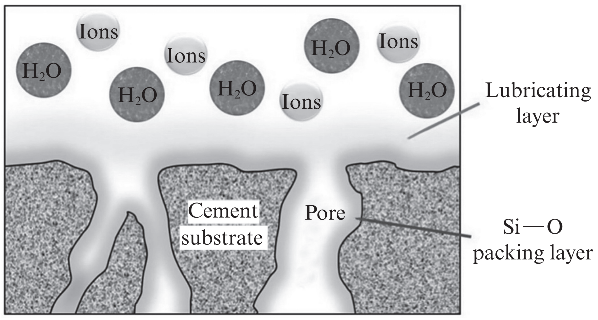
(b) Mass loss[64]　(c) Relative dynamic modulus of elasticity[64]

图7　硅烷处理后混凝土抗冻融性能

Fig.7　Freeze-thaw resistance of concrete after silane treatment



(a) Reinforcement of cement microstructure by siloxane



(b) Silane lubrication layer on the surface of cementitious materials

图8　有机硅涂层抗磨机理示意图

Fig. 8　Anti-wear mechanism of siloxane coatings[69]

有机硅涂料表面处理主要通过4个方面提升水泥基体表面的抗磨性：

(1)化学反应增强.硅氧烷分子与水泥基体活性组分中的游离氢氧根通过缩合反应形成强效的化学键结合[14],改变了C-S-H凝胶结构的层间交联形式与物化性质.在提高水泥基材抗化学腐蚀性能的同时,使其沿切线方向的晶格规整度与力学性能有所提高[70].

(2)沉积层构建.受到氢键作用与范德华力的影响,聚硅氧烷会在水泥基体表面逐渐吸附,并且反应形成铝硅酸盐沉积层[58],在填充与堵塞水泥材料毛细孔和凝胶孔的过程中有效阻挡了外部颗粒的楔入式进入,削弱了磨粒作用下凝结结构的开裂趋势.

(3)表面硬化.硅氧烷组分与水泥基体反应产生了具有较低钙硅比的硅酸盐晶体[71],并通过消耗水化产物的方式提高了水泥熟料的水化程度与抗剥落性能.陈旭[38]的研究表明,有机硅涂料处理后水泥的平均维氏硬度可以达到40 MPa以上,并呈现出较低的离散性.图8(a)呈现的水泥微结构强化机制实现了前3个方面对抗磨性能的提升作用.

(4)润滑作用.具有极低表面张力的有机硅化合物可以在胶凝材料表面形成连续且致密的耐磨润滑层,能够有效减少外力、侵蚀介质以及环境变化所引起的摩擦损耗[72].此外,内部沉积的硅氧化物可以填充细微裂纹与微凹凸面,降低水泥基材料表面的摩擦系数[73],如图8(b)所示.朱懋江等[74]研究发现,喷涂硅烷后的混凝土面层在干燥与润湿状态下的摩擦系数均有所降低.

4 实际应用中有机硅涂料存在的主要问题

相较于其他聚合物防护涂料,有机硅涂料在大规模混凝土结构表面防护工程的应用推广进展比较缓慢.首先,有机硅涂料的制备与单位面积施工成本比其他成膜型聚合物涂料高30%以上.同时,涂层施工前必须确保混凝土表面干净和相对光滑,否则硅烷结晶层很容易出现开裂、脱落等问题,特别是有机硅涂料对既有混凝土结构的防护保养效果相对较差.有机硅涂料在水泥基材料中的渗透深度仅为3~12 mm,对于大体积混凝土结构以及具有低钢筋保护层厚度钢筋混凝土结构的防护效果相对有限.

有机硅涂层在有压水环境与极端老化环境中,其疏水膜与结晶层极易发生软化与断裂,从而丧失对胶凝材料的防水抗渗性能与抗侵蚀性能.尽管有机硅涂料通过渗透结晶作用具有缺陷修补功能,但由于聚硅氧烷分子的吸附位点与硅酸盐成核速率存在较大的离散性,因而通常只能修复宽度0.2 mm以下的细小裂缝以及直径0.3 mm以下的表层孔洞,严重降低了有机硅涂料对严寒气候下水泥基材料抗冻融性能的提升效果.此外,聚硅氧烷分子中的Si—O—Si键在吸收能量后的弯曲振动频率较高,在多重物理因素以及反复荷载的耦合作用下有机硅涂层会发生明显的性能劣化现象,造成混凝土材料大面积的开裂与剥落,极大地损害了滨海环境下有机硅涂层的结构稳定性以及水泥基体的力学性能与服役寿命.

5 结论与展望

有机硅涂料通过表面疏水、传输抑制与诱导结晶的方式显著提升了水泥基材料的防水性能、抗离子侵蚀性能、防钢筋锈蚀性能、抗冻融劣化性能与抗磨性能.有机硅涂料的研究与应用不仅可为国家和企业带来巨大的经济利益,还具有重大的社会效益,在未来拥有巨大的应用发展空间.

今后有机硅防护涂料的发展可以从以下几方面工作展开:

(1)改进有机硅涂料的制备流程与合成工艺,调整硅烷单体与化学助剂的配比,在降低有机硅涂料应用成本的同时充分改善其疏水性与防水性能稳定性.聚焦高黏结性与高渗透性有机硅涂料的研发,使其适用于复杂环境下不同水泥基材料表面的快速防护.

(2)推进聚合物杂化以及矿物掺杂等调控手段在制备有机硅涂料中的应用,构建兼具表面屏蔽、传输与渗透结晶的多重防护机制,实现全服役周期内有机硅涂层抗渗性能、抗侵蚀性能与抗冻融性能的显著提高.

(3)基于纳米改性手段提升有机硅涂料对硅酸盐基体微观结构的调控水平,着重于改善硅烷结晶层的强度以及胶凝材料的物相组成与孔结构,从而在宏观层面上实现力学性能与抗磨性能的进一步提升,维持复杂气候变化下对内部钢筋的长效防护.

(4)通过聚合物氢键化、愈合剂外援等方式,赋予有机硅涂料自感知修复功能,规避承载变形、化学腐蚀与冲蚀作用下内部裂缝发展的风险,显著提升有机硅涂层的耐久性与服役寿命.

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