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Anti-icing/De-icing Mechanism and Application Progress of Bio-inspired Surface for Aircraft

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Abstract: Icing on the surface of aircraft will not only aggravate its quality and affect flight control, but even cause safety accidents, which is one of the important factors restricting all-weather flight. Bio-inspired anti-icing surfaces have gained great attention recently due to their low-hysteresis, non-stick properties, slow nucleation rate and low ice adhesion strength. These bio-inspired anti-icing surfaces, such as superhydrophobic surfaces, slippery liquid-infused porous surfaces and quasi-liquid film surfaces, have realized excellent anti-icing performance at various stages of icing. However, for harsh environment, there are still many problems and challenges. From the perspective of bio-inspiration, the mechanism of icing nucleation, liquid bounce and ice adhesion has been reviewed together with the application progress and bottleneck issues about anti-icing in view of the process of icing. Subsequently, the reliability and development prospect of active, passive and active-passive integrated anti-icing technology are discussed, respectively.

Key words: mechanical manufacturing and automation; anti-icing of aircraft; superhydrophobic surface; slippery liquid-infused porous surface; electrothermal coating

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0 Introduction

Ice accretion has become an urgent challenge in aerospace, energy and mechanics fields, which greatly harms the reliability and safety of equipment. Due to the impact of raindrops under low temperature condition or the supercooled droplets in the atmosphere during flight, ice will easily accumulate on aircraft surface and greatly change the aerodynamic shape of aircraft[1-2]. The most serious parts of aircraft for ice accumulation include the leading edges of the wings and empennages, the propellers and hubs on fixed-wing aircraft, the engine nacelles, the rotor blades on helicopters, and the windshields[3]. Thus, various anti-icing strategies have been proposed and applied to aircraft, including mechanical deicing, thermal output anti-icing and antifreeze liquid technologies[4-5]. The above strategies relying on external energy input are generally called traditional active anti-icing technologies. However, there are several disadvantages that cause the traditional active anti-icing strategies unsuitable for present aircraft requirements, such as enormous energy consumption, elaborate supporting systems and serious environmental pollution. This imposes restrictions on the development and application of active anti-icing systems.

Due to low-hysteresis, non-stick properties (rolling angle (RA) lower than 5°), slow nucleation rate and low ice adhesion strength, superhy-

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drophobic surface (SHS) and slippery liquid-infused porous surface (SLIPS) have been deemed as two of the most excellent candidates for new generation anti-icing surface in aerospace[6]. SHS is inspired from the surface of lotus leaves[7], butterfly wings[8], rice leaves[9], water strider legs[10], etc., and the latter from Nepenthes[11]. Both SHS and SLIPS mainly rely on interface characteristics to realize anti-icing properties, thus called passive anti-icing strategies, which can effectively assist the existing anti-icing systems getting rid of energy limitation and environmental pollution. However, the passive bio-inspired anti-icing surfaces are extremely easy to lose efficacy due to microstructure breakage or chemical property degradation in harsh flight environment.

To solve the shortcomings of passive and active anti-icing strategies, researchers combine the novel active anti-icing surfaces and bio-inspired low adhesion surfaces, and the combination effectively reduces the anti-icing energy consumption and improves the interface performance. Therefore, the integrated anti-icing strategies have become an important trend in the development of bio-inspired anti-icing system in aerospace[12-16]. Herein, this review will systematically introduce recent anti-icing research from the aspect of theories, strategies and progresses based on bio-inspired anti-icing surfaces. In view of the processes of icing on the surface of aircraft, the existing strategies are divided into two types: (1) Lessening ice accumulation by slackening ice nucleation or shortening the contact time of droplets, (2) weakening the ice adhesion by transforming the contact interface between ice and substrate. The mechanism of ice nucleation, liquid bounce and ice adhesion has been reviewed together with the application progress and bottleneck issues about anti-icing, such as durability, energy consumption and multi-function compatibility. We hope this review will contribute to profoundly understanding anti-icing mechanism and provide reliable guidance for the next-generation anti-icing system.

1 Anti-icing Mechanism

1.1 Slackening ice nucleation and shortening droplet contact time

Icing refers to the process that droplet spontaneously forms ice cores internally, and ultimately grows with reduction of temperature[17]. Gibbs' hypothesis points out nucleation is necessary during phase transitions, such as freezing[18-19]. The nucleation process of droplet can be classified into homogeneous nucleation and heterogeneous nucleation, and the latter is more ubiquitous in real life. The homogeneous nucleation occurs at the temperature approximately below -40 ℃[20]. The classical nucleation theory is shown as

$$\Delta G_N^* = \frac{\left(\pi\gamma_{\text{l-v}} r^{*2}\right)\left(2-3\cos\theta+\cos^3\theta\right)}{3} \tag{1}$$

$$\cos\theta = \frac{\gamma_{\text{s-l}}-\gamma_{\text{s-v}}}{\gamma_{\text{l-v}}} \tag{2}$$

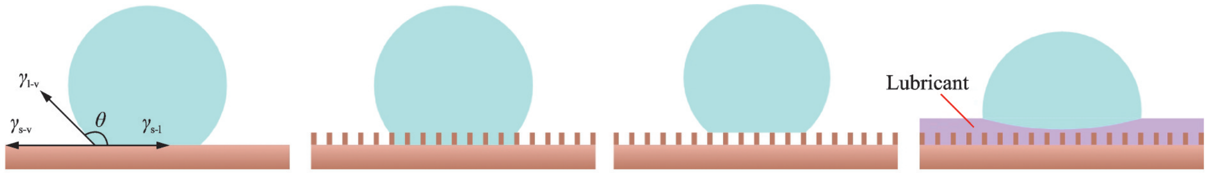
where $\gamma_{\text{l-v}}$, $\gamma_{\text{s-l}}$, $\gamma_{\text{s-v}}$ are the surface tension coefficients of liquid-gas, solid-liquid and solid-gas, respectively; r^* is the radius of curvature of droplets, and θ the contact angle (CA) of droplets on the surface, as shown in Fig.1(a). Eq.(1) shows that the free energy required for icing is decreased with the decrease of the CA, which demonstrates the significance of CA for icing. Thus, the CA is intuitive indicator for assessing the ice resistance of a surface (Fig.1(a)). SHS exhibits the ability for inhibiting the heterogeneous nucleation of droplets on the surface due to larger CA in static environment[21-22], which can be demonstrated by Wenzel model (Fig.1(b)) and Cassie-Baxter model (Fig.1(c))[23-24]. From the perspective of energy, the edge or defect region exhibits a lower energy barrier for heterogeneous nucleation, resulting in easier icing and then subsequently expanding outwards in the form of ice bridges[17, 25]. As for SLIPS, lubricant liquid infiltrates into the micro/nano-structure instead of air and generate continuous and homogeneous lubricant phase on the surface[26] (Fig.1(d)). This will result in homogeneous nucleation of droplets with higher energy barrier, which delays or even inhibits the heterogeneous nucleation of static droplets on the surface[27-29].

In static environment, ice mass and area will be effectively reduced by inhibiting heterogeneous nucleation. However, for dynamic icing process, such as the real flight condition, the supercooled droplets hit the cold surface at a high speed and nucleate instantly, resulting in ice accretion. The droplet impacting and freezing processes on the cold surface can be divided into four stages (Fig. 1(e)): Contact and nucleate; ice nucleus growing; recede and oscillate; and completely frozen. Droplet within a certain range of Weber number (*We*) will rebound when impacting on bio-inspired surfaces, such as SHS and SLIPS.

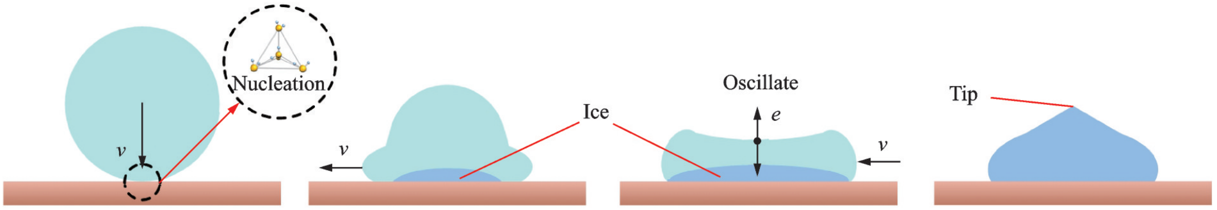
In cold environment, droplet rebounding or sticking dynamics is associated with viscosity and nucleation[30]. Maitra et al.[31] found that increased viscous effects significantly influenced all stages of impact dynamics, in particular, the impact and meniscus impalement behavior. Recently, Zhang et al.[32] proposed that the phenomenon of liquid adhesion on the surface under low temperature was caused by ice nucleation rather than enhanced viscous effect. As same as SHS, lubricant film on SLIPS is easily deformed and extruded until being function failed when droplet impacting on the surface with high *We*. The loss of interface characteristic results in droplet pinning on the surface and increases the contact area between the droplet and the substrate, which enhances heat transfer efficient, leading to heterogeneous ice nucleation easily[33]. Bird et al.[34] proved that a small fraction of heat was transferred between the droplet and the surface during the droplet impacting and rebounding, and quantitatively estimated how much heat was transferred. This phenomenon promotes droplet nucleation and causes the droplet to ultimately fail to bounce. Furthermore, when a relatively hot droplet impacts on a cold surface, the droplet easily condenses and eventually fully infiltrates within the cavities at the micro-nano structure leading to bouncing failure[35].

Thus, strengthening droplet bounce behaviors and obstructing heat transfer between droplet and substrate can avoid droplet nucleation and decrease ice accretion on the surface. Superhydrophobic surface exhibits special droplet bounce behavior, which endows the interface with great anti-icing ability[36-39]. The behavior mainly includes the coalescence and jumping of condensed droplets and bouncing behaviors of microdroplets impacting on the surface[8, 40]. Microstructures on the surface can offer high velocity and energy conversion efficiency for droplets during the process of spontaneous coalescence and jumping. Liu et al.[41] designed a micro-anisotropic SHS, using the work of adhesion as the steering force for leaping microdroplets, and achieved the leaping of droplets in a guided lateral direction on SHS without any external forces. Lu and Wang et al.[42-43] studied the energy conversion efficiency and the droplet velocity during the bouncing process on various microstructures respectively. Liu et al.[44] observed that a special pancake bounce phenomenon occurred as the millimeter-sized droplets impacting on the nanostructure-modified conical pillar array surface when *We* was 10. The structure could further reduce the contact time to 3.4 ms (Fig.1(f)). The capillary energy stored at the bottom of the droplet can be converted into kinetic energy, which is enough for the droplet to bounce in the shape of pancake[45-46]. Bird et al.[47] demonstrated that liquid mass could be redistributed when impacting on SHS with ridged structures leading to the morphology transformation of droplet hydrodynamics, which broke the lower bound on the impact time of ordinary SHS to 7.8 ms (Fig.1(g)). Shen et al.[48] revealed the lower bound on the impact time was 5.5 ms on SHSs with multi ridged structures (Fig.1(h)).

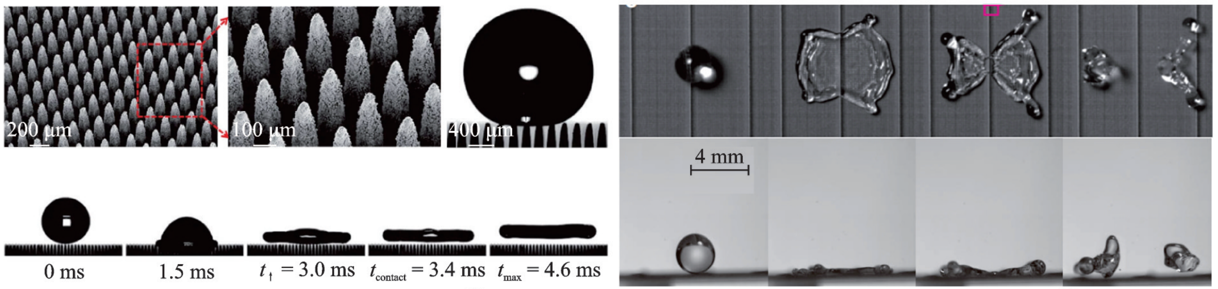
Droplets exhibit lesser pinning when impacting on SLIPS compared with SHS. Yeganehdoust et al.[49] compared the behaviors of droplets when impacting on SHS and SLIPS by simulation, and proposed that the microdroplets penetrated the microstructure on SLIPS was much weaker than that on SHS at high *We* ($We \approx 160$). Biroun et al.[50] offered an effective way of applying surface acoustic wave technology along with SLIPS to reduce the contact time and alter the droplet rebound angles. These phenomena provide an idea for the subsequent suppression of heat transfer and phase change of the supercooled droplet impacting on surfaces



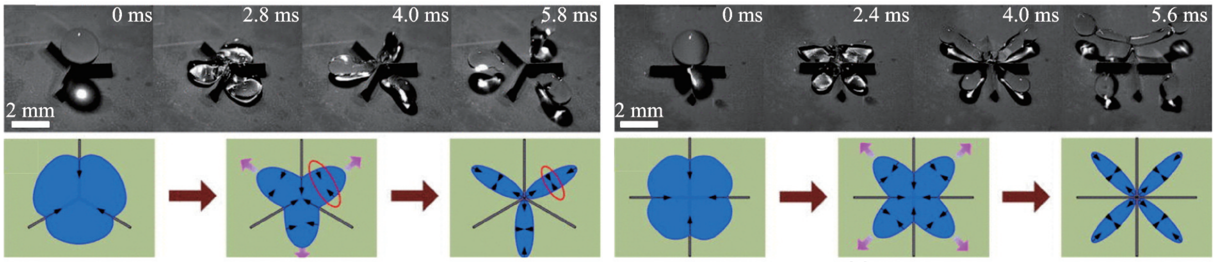
(a) Contact angle (b) Wenzel model[23] (c) Cassie-Baxter model[24] (d) Droplet on SLIPS[26]



(e) Droplet impact and freeze processes



(f) Droplets bounce in pancake shape[44] (g) Microstructure to split droplet[47]



(h) Impacting droplet on SHS with complex macrotextures[48]

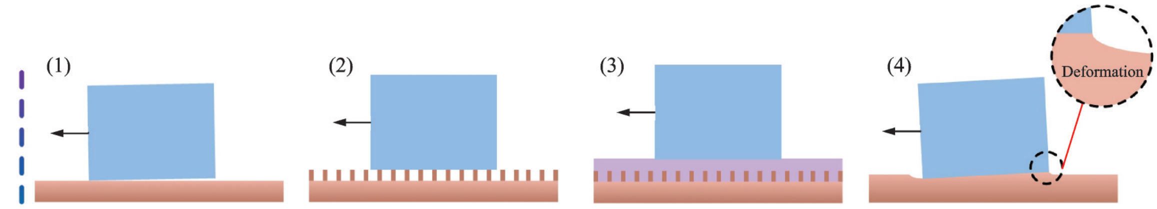
Fig.1 Schematic of droplet morphology and freezing processes and mechanism of shortening the contact time of droplets on SHS

with a high speed.

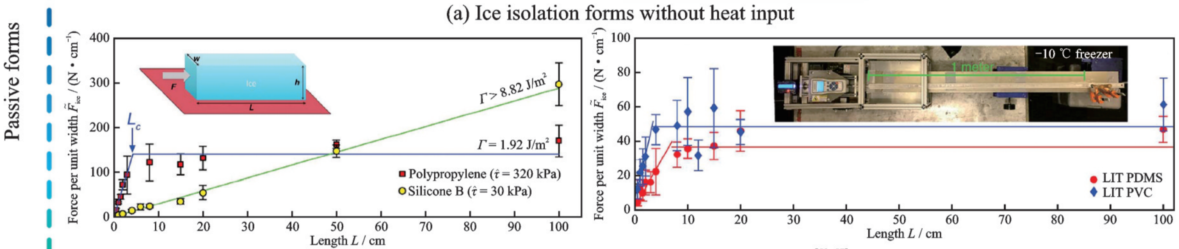
1.2 Build ice-phobic interfaces

Bio-inspired anti-icing surface has been proved to effectively delay the occurrence of icing in static environments, however, in harsh flight environment, the function will gradually fail and generate massy ice accretion with time. Generating isolation between the ice and the surface has been deemed as an effective and promising strategy to solve this problem[25, 51]. Thus, ice adhesion strength $\tau_{ice} = F/A$ is the key parameter for measuring the ability of ice-phobic of these surfaces, which represents the required force (F) for removing ice with unit area (A) on the surface[52-53]. The surface with ice adhesion strength lower than 100 kPa are recognized as ice-phobic surface[54]. Ice-surface isolation forms can be divided into two categories according to the presence or absence of heat input. The passive forms without heat input include hard-hard interface crack (on ordinary surfaces), Cassie ice falling off (on air film), ice slipping away (on oil film), and hard-soft interface crack (on soft substrate), as shown in Fig.2(a). Additionally, combining bio-inspired surface properties with the soft substrate is promising to promote crack initiation, further reducing the force required[55-56]. Golovin et al.[52, 57] reported an anti-icing material with low interfacial toughness (LIT), which significantly reduced the force required to remove a large area of ice, and confirmed that the force was independent of the ice area (Fig.2(b)). In Fig.2(b), L_c is the critical bonded length at which a transition between the interfacial cohesive strength mode and bonding energy mode of failure occurs and Γ the interfacial toughness. Under the action of external force (e. g. gravity, wind, etc.), local deformation stress concentration occurred on the surface of the low interface toughness material and interface micro-cracks are generated, after which the force required for the expansion of micro-cracks is no longer affected by the ice area. Compared with the super-wetting interface, the elastomer surface can promote the occurrence of interfacial cracks and accelerate the debonding of the ice by means of the obvious mismatch of the elastic moduli of the substrate and the ice[57-58]. Thus, on the soft substrate, $\tau_{\text{ice-soft}}=\sqrt{\dfrac{EG}{\pi a\Lambda}}$, where E is the substrate modulus, G the surface energy, a the total crack length, and Λ the non-dimensional constant related to interface geometric configuration[59]. According to different test forms, ice adhesion experimental devices can be divided into centrifugal type, parallel push type, etc. Ice adhesion experiments need to be standardized, such as the environment of the icing process, the geometric parameters of the ice, the kinematic parameters of the testing process, etc. This standardization helps to compare the ice adhesion results from different laboratories, and establish an industry standard that can measure the anti-icing ability of the surface[60]. However, the research of ice behavior on the soft substrate is still limited to the macroscopic measurement of icing adhesion[61-62]. To further research the ice desorption behavior and crack formation mechanism on various interfaces is necessary and urgent for anti-icing field.

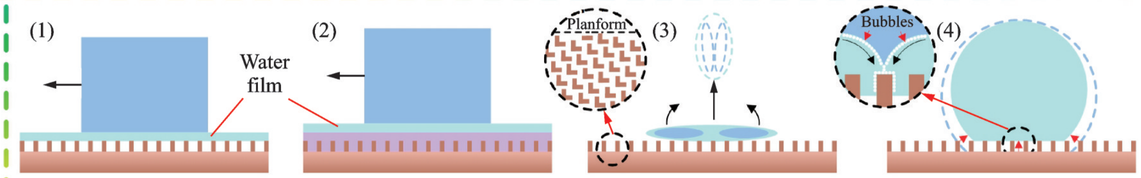
The active forms accompanying by ice melting process include slipping away (isolated by heat-induced water film), melting frost self-jump[36] and spontaneous Wenzel to Cassie-Baxter transition[63] (Fig.2(c)). Ref.[51] revealed the bio-inspired anti-



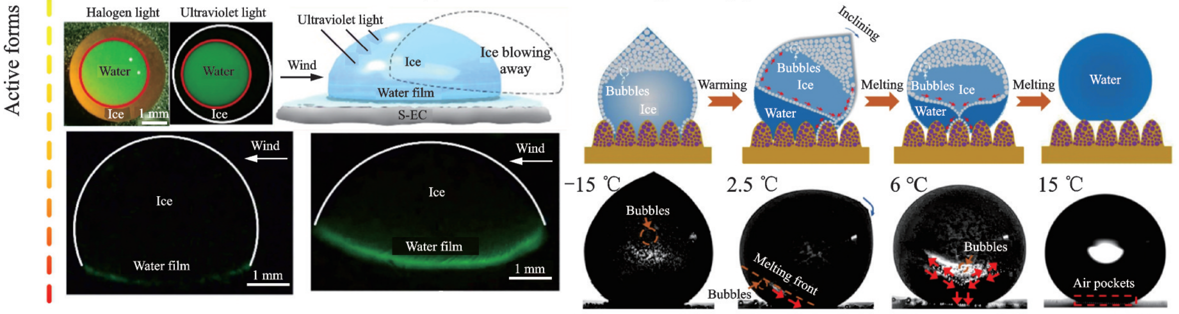
(a) Ice isolation forms without heat input



(b) Low interfacial toughness anti-icing surface[52, 57]



(c) Ice isolation forms during melting process[36, 63]



(d) Superhydrophobic electrothermal coating anti-icing mechanism[51] (e) Ice Wenzel-Cassie transition on superhydrophobic surface[63]

Fig.2 Schematic of ice isolation forms

icing synergistic mechanism of "heat-induced water film isolation". The contact interface switched from "solid-solid contact" into "solid-liquid-solid contact" when heating, which greatly weakened the ice adhesion force (Fig.2(d)). In addition, the spontaneous motion phenomenon of frost on SHS during the melting process has also attracted attention[64-65]. Refs.[36, 66] revealed the mechanism of melting frost self-jumping phenomenon from the energy point of view, and proposed a frost directional self-jumping control method based on anisotropic macro-micro structure. Generally, droplets turn from Cassie-Baxter model to Wenzel model during icing and the process is hard to recover to Cassie-Baxter model after melting. Zhong et al. realized the spontaneous transformation from Wenzel state to Cassie-Baxter state at the interface between ice and the surface during the ice-melting cycle[63]. The reversible switching was attributed to the bubbles in the ice drop which rapidly hit the micro-nano structure under the action of Marangoni force (Fig.2(e)). Thus, the combination of active and passive anti-icing strategies is significant and meaningful for anti-icing in aerospace.

2 Anti-icing Strategies

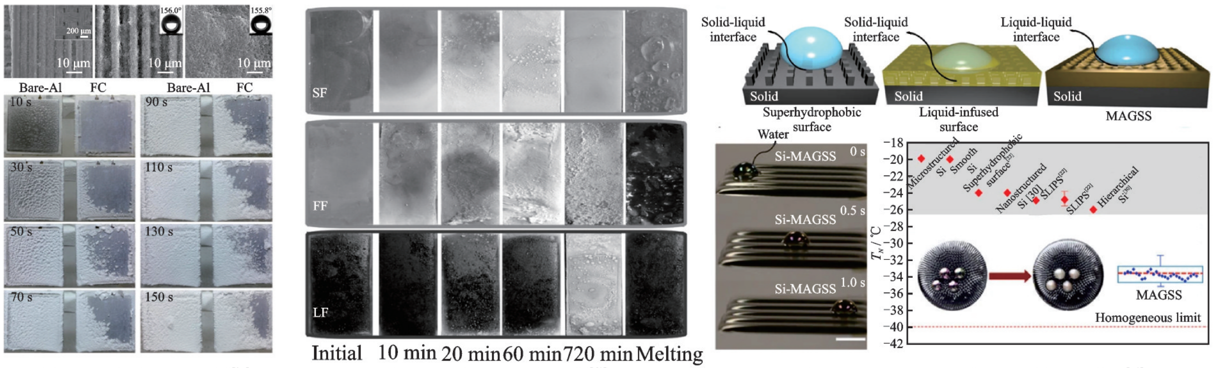
2.1 Lessening ice accumulation

The strategies of lessening ice accumulation can be divided into solid-solid (SHS), solid-liquid (SLIPS) or solid-biology (Antifreeze protein, AFP) contact form based on the substrate.

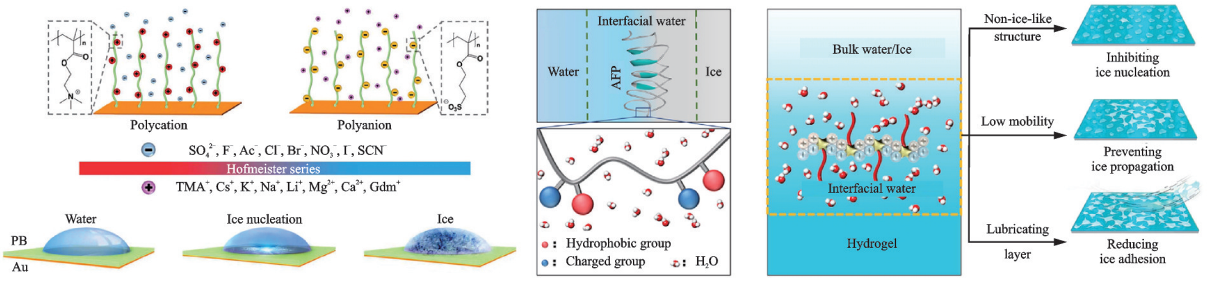
Compared with ordinary surfaces, SHS minimizes the effective contact area between supercooled droplets and cold solid substrates, inhibiting the formation of ice[67-68]. Ref.[25] constructed a robust SHS based on fiberglass cloth (FC) with micro-texture and adhesive. The surface exhibited significantly icing delay and greatly reduced ice accretion in dynamic ice test compared with origin substrate (Fig.3 (a)). In addition, high mechanical strength substrate and strong adhesive strengthen the abrasion resistance of the surface.

SLIPS is covered by a low surface energy lubricant film, delaying the heterogeneous nucleation of ice. Yin et al.[69] demonstrated a scalable and reproducible coating method to create a slippery surface on aluminum (Fig.3(b)). Compared with pure and smooth PDMS film without pores fluorination (SF) and lubricant fluorinated porous film (FF), droplets on fluorinated porous film infiltrated with perfluoro polyether lubricant (LF) could easily slide off from the surface before freezing, significantly reducing ice accretion. The ice on LF also could be easily removed from the surface by gravity at low tilt angles after partially melting. Qian et al.[70] constructed SLIPS with pressure responsive property by a facile template strategy. The surface regulated the porous structure under external pressure, which effectively reduced the loss of lubricant and droplets can easily shed on the as-prepared surface results in a short contact time, avoiding ice nucleation. Irajizad et al.[71] reported a new magnetic slippery surface (MAGSS) with anti-icing at -34 ℃ and excellent icing delay ability (Fig.3(c)). In addition, the surface also exhibited extremely low ice adhesion strength (about 2 Pa) and stability in shear flows up to Reynolds number of 10^5. However, loss of lubricant will cause the function failure of SLIPS under dynamic environments, especially at high speed and shear force situations. To improve this shortcoming, researchers adopted non-flowing media instead of flowing to enhance the stability of the surface, including polyelectrolyte brushes and AFP grafted surface, both of which exhibited excellent ice delay properties[72-75]. The combination of polymer brush and ion endows the surface with unique capabilities of inhibiting ice nucleation and propagation. Wang et al.[75] revealed the inner mechanism of ions at polyelectrolyte brush (PB) for controlling the ice nucleation and propagation processes, further reinforcing the significance of ion in inhibiting ice formation (Fig.3(d)).

AFP in polar animals and plants has two characteristics of thermal hysteresis activity: Reduction of recrystallization, ice crystal growth inhibition[72-73]. He et al.[74] prepared a multifunctional anti-icing hydrogel by combining antifreeze proteins with polydimethylsiloxane-grafted polyelectrolyte hydrogels (Fig.3(e)), which can simultaneously achieve



(a) Icing delay on SHS[25]
(b) Icing delay on SLIPS[69]
(c) Magnetic slippery ice-phobic surface[71]



(d) Ice nucleation polyelectrolyte brush surfacesp[75]
(e) Antifreeze protein grafted surface[74]

Fig.3 Strategies to reduce ice accumulation

inhibition of ice nucleation (ice nucleation temperature below -30 ℃), prevention of ice growth (ice propagation rate below 0.002 cm^2/s), and reduction of ice adhesion (below 20 kPa). Wang et al.[76] designed a material with cold adaptation by combining hydrogel and AFP. The surface exhibited excellent anti-icing performance at low-temperature conditions.

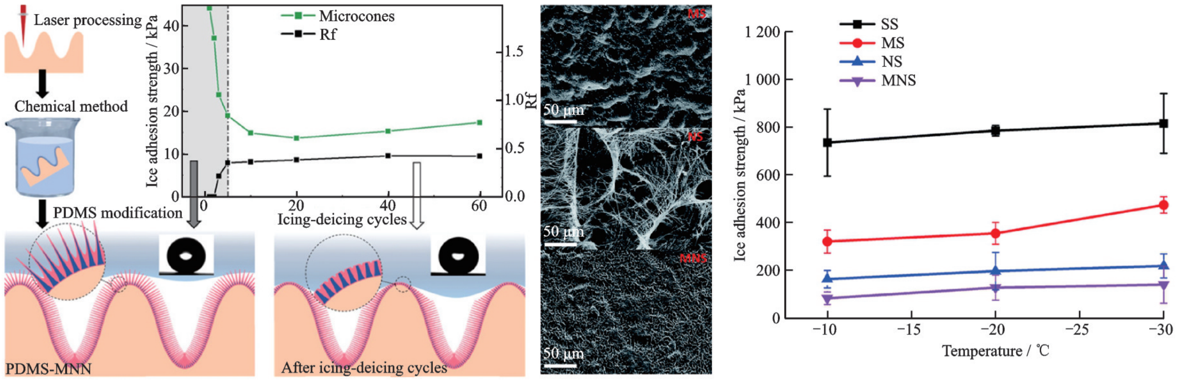
2.2 Weakening ice adhesion

The existing anti-icing strategies for weakening the ice adhesion can be classified into passive and active approaches[14, 77-78]. Passive strategies rely on special micro-nano structures, such as SHS and SLIPS. Active anti-icing methods mainly prevent ice accretion with an external energy source[79-80]. Both surfaces have been deemed outstanding candidates for anti-icing due to their low ice adhesion[80-81].

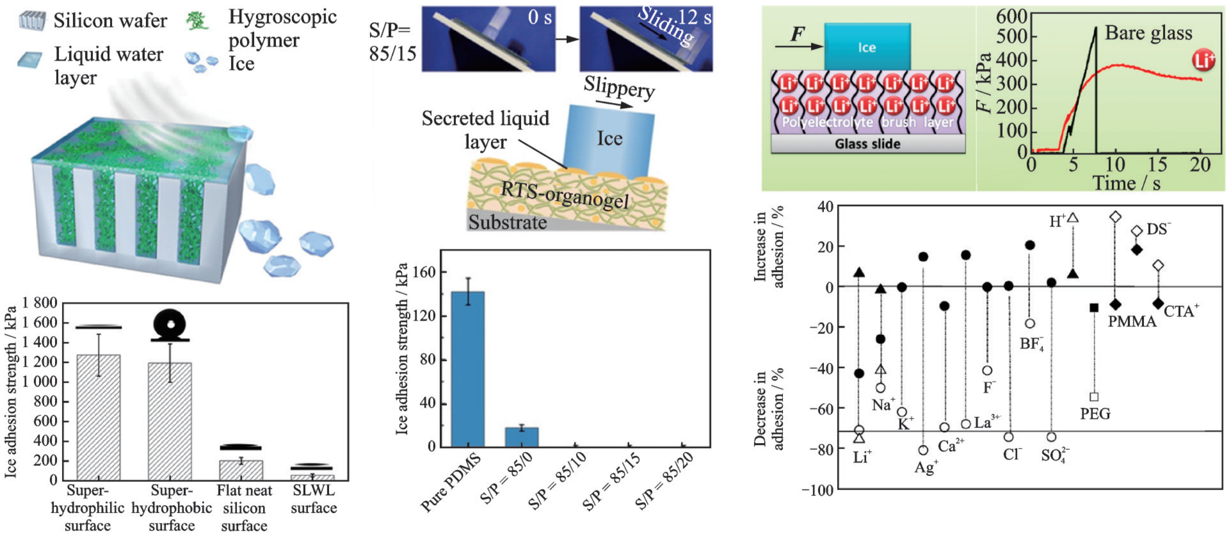
2.2.1 Passive strategies for weakening ice adhesion

As shown in Figs.1(b, c), air film traps into the micro-/nano-structure when the droplet contacts with SHS. In the process of icing, a nano-air film also will be trapped and generate “air film isolation”, leading to low ice adhesion on SHS, which has been observed by cryo-focused ion beam-assisted scanning electron microscopy[39]. The existence of a large amount of air pockets confirmed that the frost at this time was in the Cassie state (so-called “Cassie ice”), which intuitively explained the reason for low adhesion of ice on the nanotextured SHS. Chen et al.[82] fabricated a robust micro-nano-nanowire triple structure-held PDMS superhydrophobic surface (PDMS-MNN), which showed a long-term anti-icing performance with high deicing robustness and low ice adhesion strength (Fig. 4(a)). In Fig.4(a), Rf means the product of surface roughness and substrate area fraction. Shen et al.[83] proposed that the icing-delay time of a droplet on SHS was many times longer, and the ice adhesion strength on SHS was greatly reduced, which was attributed to the Cassie wetting state of a droplet on the surface (Fig.4(b)). In Fig.4(b), SS, MS, NS, and MNS refer to Ti6Al4V substrate surfaces without modification, micrometer-scale pit surface with FAS-17 modification, nanowire structured surface with FAS-17 modification, and micropit-nanowire structured surface with FAS-17 modification, respectively. Based on this theory, Zhou et al.[84] constructed an anti-icing concrete material which realized good anti-icing and easy deicing ability (deicing stress: 0.06 MPa). However, it is still controversial in some papers whether SHS can reduce ice adhesion, which can be explained by the gradual damage of the surface structure, the condensation of water in the microstructures[85], and the mechanical interlocking between the ice and the surface texture[86].

Except from SHS, novel slippery surfaces, such as SLIPS and liquid-like surface, exhibit excellent ability for reducing ice adhesion. Chen et al.[87] fabricated a robust anti-icing coating with a self-lubricating liquid water layer (SLWL) where the ice adhesion was one order of magnitude lower than that on SHS, exhibiting excellent capability of self-healing and abrasion resistance (Fig.4(c)). Wang et al.[88-89] proposed an anti-icing coating with self-lubricating water layer, which formed a water lubricating layer between the ice and the solid surface. The as-prepared surface could remain liquid at −42 ℃ and greatly reduces the adhesion of the ice. Liu et al.[90] incorporated a binary liquid mixture with an upper critical melting temperature into reversibly thermosecreting organogel (RTS-organogel) to obtain a SLIPS with the temperature-controlled phase isolation of solution (Fig.4(d)). The surface exhibited extreme low ice adhesion strength, and the ice adhesion strength decreased with the decrease of the mass ratio of silicone oil to liquid paraffin (S/P). Zhao et al.[91] observed the behavior of ice adhesion on liquid-like surface and demonstrated the macroscopic evidence of the liquid-like of surface-tethered poly (dimethylsiloxane) brushes. Whereas ice permanently detaches from solid surfaces when subjected to sufficient shear, commonly referred to as the material's ice adhesion strength, adhered ice instead slides over PDMS brushes indefinitely. The liquid-like surface exhibited extreme low ice adhesion strength of 0.3 kPa. Chernyy et al.[92] constructed various polyelectrolyte brush surfaces on glass surfaces, and found that Li$^+$ polyelectrolyte brush



(a) Ultra-long-term ice-phobic SHS[82] (b) Ice-phobic potential of hierarchical SHS[84]



(c) Self-lubricating anti-icing coating[87] (d) Low ice adhesion on the organogel surface[90] (e) Polymer brush anti-icing surface[92]

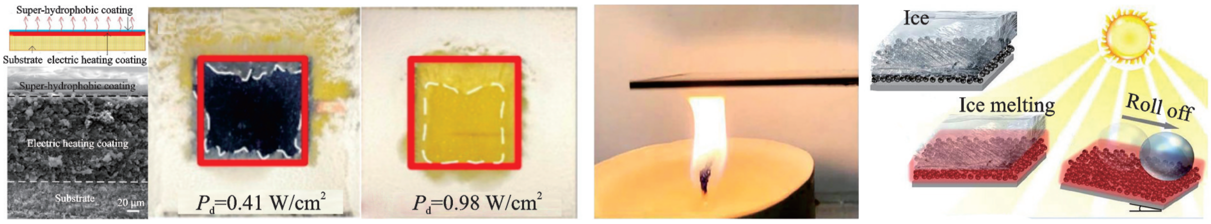
Fig.4 Strategies to reduce ice adhesion without additional energy input surfaces reduced the ice adhesion by 40% at −18 ℃ and by 70% at −10 ℃, while Ag^+ ions reduced ice adhesion by 80% at −10 ℃ (Fig.4(e)).

2.2.2 Active and integrated strategies for weakening ice adhesion

Passive strategies have shown excellent water repellency and low ice adhesion in laboratory environment. However, these strategies exhibit serious limitations in dynamic environments, such as severe flight icing condition with high speeds, low temperature[86, 93]. The contact form between the ice and SHS will become mechanical interlocking/solid-solid contact. Meanwhile, the low ice adhesion effect of SLIPS is greatly dependent on the amount of oil in the liquid film which is extremely easy to decrease gradually with the icing-deicing cycles[94]. The active and integrated strategies generate a water lubricating layer on the contact interface to form a "heat-induced water film isolation", assist the passive anti-icing surface to play a role, and ensure a low adhesion effect. The existing active strategies rely on external energy input to maintain a liquid layer for completely anti-icing, which also have been adapted in commercial.

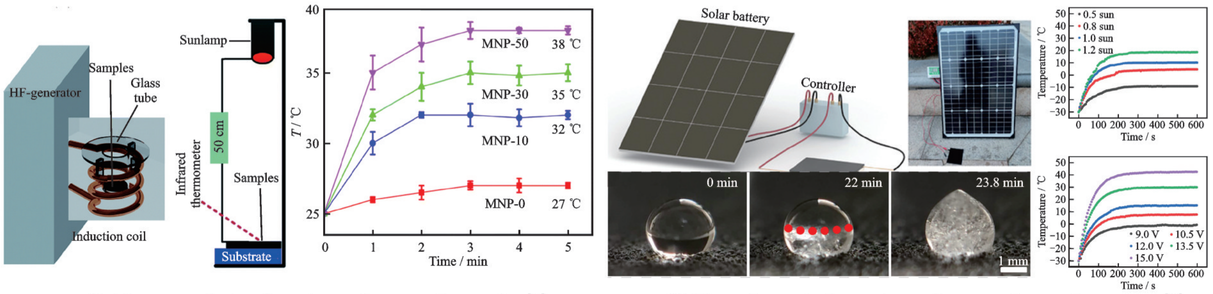
The novel active anti-icing methods mainly include electrothermal, photothermal, magneto-thermal and several synthetical anti-icing methods[51, 61-62, 95-100]. Compared with the traditional built-in heating sheet, the novel conductive heating coating can directly act on the icing interface with high heat transfer efficiency and rapid response time, which mainly relays on conductive polymer or mixed metal nanoparticles, carbon nanotubes, graphene and other conductive nanoparticles[61-62]. Ref. [51] obtained an electric heating anti-icing SHS by combining carbon nanotube and hydrophobic substances. The surface realized excellent anti-icing ability with low energy consumption (Fig.5(a)), which reduced the energy consumption by 58% compared with the traditional electric heating method. The coating has been widely adapted by various aircraft wings, engine inlets, wind turbine blades, and other surfaces for anti-icing. In addition, Cheng et al.[95] designed a new nano-integrated coating with superhydrophobicity, magnetocaloric effects and wetting stability by combining magnetic nanoparticles (MNP) with fluorine-containing copolymers. Under the alternating magnetic field of 7.8 kW, the surface raised from 24 ℃ to 44 ℃ at 25 s. Meanwhile, the temperature could rise more than 10 ℃ at 1 min under 75 W infrared light (Fig.5(b)).

The above-mentioned ways need energy from external input by the equipment. However, sunlight is a natural source of heat, which can be fully used for surface heating for active anti-icing with energy-saving, thus, photothermal surface has attracted researchers' attention gradually[99]. Wu et



(a) Superhydrophobic electrothermal coating anti-icing mechanism[51]

(c) Ice-phobic surface based on candle soot[96, 100]



(b) Magnetocaloric-photothermal composite coating[95]

(d) Photothermal-electrothermal composite coating system[97]

Fig.5 Novel active and integrated strategies for weakening ice adhesion by "heat-induced water film isolation" al.[96, 100] obtained a low-cost, high-efficiency superhydrophobic photothermal surface based on candle soot, which exhibited fast heating characteristics under sunlight, inhibiting icing and rapidly melting frost on the surface (Fig.5(c)). Liu et al.[97] prepared a carbon nanotube-modified fluoropolyacrylate-based SHS, which had both photothermal and electrothermal effects to ensure energy-saving and reliable deicing under different sun irradiations (Fig.5(d)). Zhang et al.[98] used an ultrafast pulsed laser deposition method to prepare a photothermal superhydrophobic coating, which could maintain excellent photothermal conversion, droplet self-clearing, anti-icing and frost-resistant properties in extreme environments of low temperature and high humidity.

Above all, we can see that various active anti-icing methods can effectively achieve surface anti-icing. The integration of active and passive strategies can realize long-term, high-efficient and energy-saving anti-icing, which have been deemed as next-generation anti-icing technology[12-16].

3 Challenges and Trends of Bio-inspired Anti-icing/De-icing Strategies

Biomimetic passive and active anti-icing strategies have shown excellent results, but their large-scale applications on aircraft are still subject to challenges, such as poor durability, large energy consumption requirements, and multi-function compatibility problems. The researches on overcoming these challenges will be the development trend of the bio-inspired anti-icing technology.

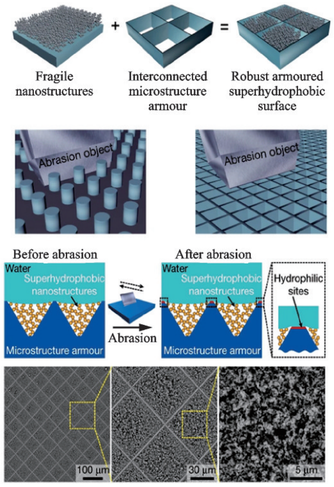
As above mentioned, active anti-icing methods mainly prevent ice accretion with an external energy source[79-80], but require complex regulatory equipment to prevent overheating or excess energy consumption under various cold loads[77]. Passive strategies rely on special micro-nano structures but their poor durability and fragility cause function failure in dynamic environments[19, 74, 101]. Poor tolerance, large energy consumption, and multi-function compatibility have become urgent problems for next generation functional surface for anti-icing.

3.1 Robustness enhancement of passive anti-icing surfaces

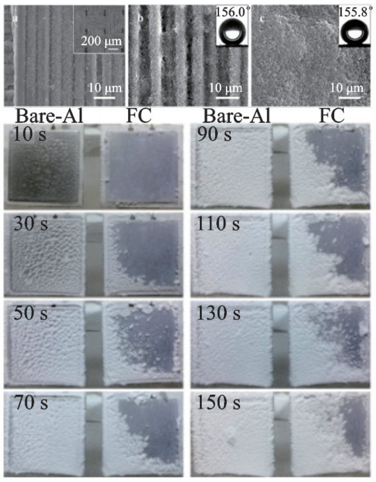
The passive bio-inspired surfaces realize excellent anti-icing performance by gas film/liquid film within micro-nano structures. However, surfaces will fail during icing cycles due to nanostructures with poor mechanical durability and lubricants that easily to evaporate or lose[85, 102-103]. On SLIPS, the droplets would pull up and can even become completely cloaked by lubricant, which can accelerate lubricant depletion from the textured surface and cause pinning[104-105]. Even more fatal for SHS and SLIPS, once the droplet enters the micro-nano structure and nucleates into ice, the newly formed ice will interlock with the structure, making the ice more difficult to move[37, 53]. Moreover, the ice-wind tunnel tests also revealed that the SHS cannot withstand high-speed, low-temperature and high-humidity conditions during flight[105-106].

To promote the application of bio-inspired anti-icing performance in flight environments, it is necessary to ensure the reliability of micro-nano structure and interface film. The general robustness enhancement strategies for SHS can be divided into microstructures protection[107-109], reinforcement by adhesive[25, 110], and homogeneous structure[111-115]. Deng et al.[107] combined the superhydrophobic property of nanostructures with the durability of microstructures (Fig.6(a)). A SHS with strong wear resistance was prepared on the glass substrate on which the microstructures acted as “armor” was used to protect the fragile nanostructure inside the frame. Zhong et al.[116] designed a three-scale integrated micro/nano-structured SHS on the metal surface by ultrafast laser ablation, chemical oxidation and other methods, which exhibited excellent Cassie state stability, high critical Laplace and low ice adhesion strength. Lu et al.[110] used commercial adhesives to bond the superhydrophobic paint composed of perfluorosilane-coated titanium dioxide nanoparticles to various substrates and promote robustness. These surfaces maintained their water repellency after finger-wipe, knife-scratch, and even 40 abrasion cycles with sandpaper. Combined the micro-texture of the fiber and the strong adhesion of the adhesive, our group[25] prepared a glass fiber cloth-adhesive-based SHS, which immensely enhanced the sandpaper abrasion resistance. Without additional heating, the obtained surface showed significantly icing delay under dynamic conditions (Fig.6(b)). Peng et al.[115] designed an all-organic, flexible superhydrophobic nanocomposite coatings by combining polytetrafluoroethylene, perfluoropolyether (Krytox oil) and fluorinated epoxy (denoted as PKFE coating) with strong mechanical robustness under cyclic tape peels and Taber abrasion (Fig.6(c)). Our group[111] used fluorinated epoxy resin and carbon/PTFE particles to prepare the multifunctional anti-icing/de-icing coating, which exhibited excellent robust superhydrophobic property by repeated sandpaper abrasion and tape peeling tests.

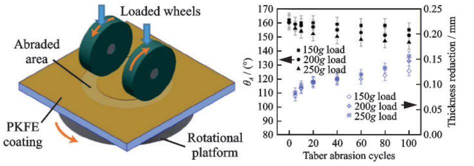
On the other hand, the general robustness enhancement strategies for SLIPS can be divided into lubricant holding structures[117-120], phase-changeable or solid lubricants[121-123], and slow release or continuous delivery structure of lubricant/antifreeze[124]. Wang et al.[125] constructed a uniform WO_3 nanofiber network layers on the stainless steel which possessed highly lubricant holding ability and realized long-term stability of SLIPS (Fig.6(d)). Liu et al.[90] incorporated a binary liquid mixture with an upper critical melting temperature into an organogel (Fig.4(d)) to achieve a temperature-controlled phase isolation of solution infiltration gels. As a result, the surface friction coefficient changed from 0.4 to 0.03 and the ice adhesion strength fell down below 1 kPa. Our group[124] added phase-change lubricant microcrystalline wax into the elastomer network to construct a temperature responsive slippery surface whose slippery ability could gradually increase with temperature rising (Fig.6(e)). The combination of elastomer and phase change lubricant enhances the



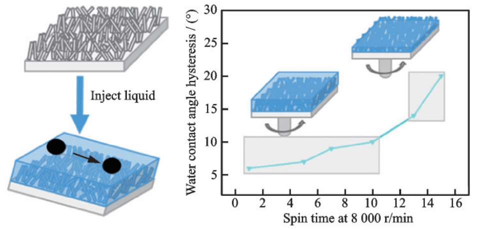
(a) "Armor" type wear-resistant SHS[107]



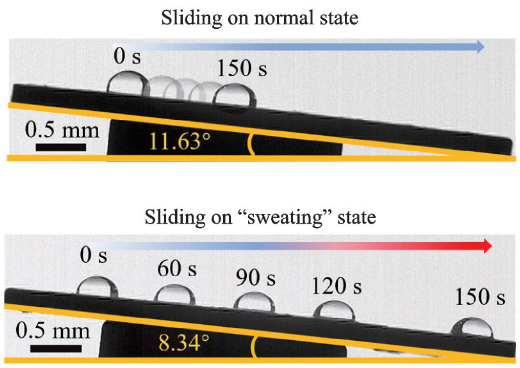
(b) Wear-resistant SHS with adhesive[25]



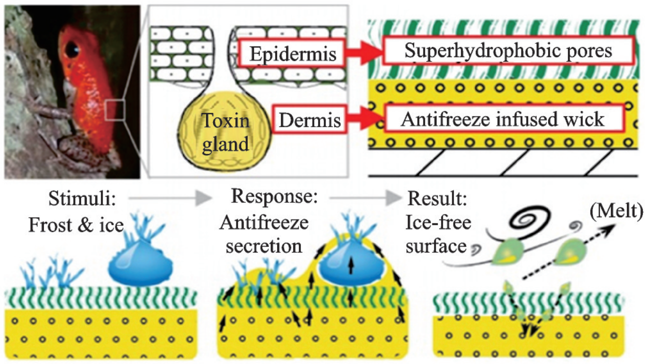
(c) Robust Superhydrophobic coatings with homogeneous structure[115]



(d) Oil-holding nanofiber networks on SLIPS[125]



(e) Slippery surface with phase-changeable lubricants[125]



(f) Antifreeze sustained release surface[124]

Fig.6 Robustness enhancement strategies for passive anti-icing surfaces lubricant stability of SLIPS, and now the mass loss is extremely tiny under the erosion of the water vapor two-phase flow. Sun et al.[124] deeply studied the way in which the poison dart frog stores and secretes toxins, then proposed and constructed a double-layer with different infiltration properties as shown in Fig.6(f).

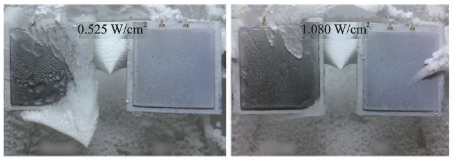
However, the robustness of passive anti-icing surface is still not enough for aerospace requirements, it is necessary for developing anti-icing surface with higher mechanical strength.

3.2 Active-passive integrated anti-icing/de-icing strategies for energy saving under flight conditions

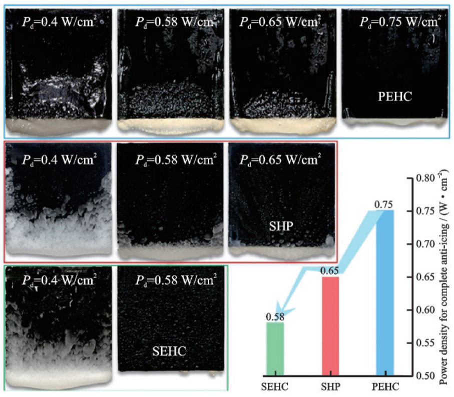
In aerospace, the power consumption of the anti-icing system is required to be as small as possible due to the limited power supply and cost on the aircraft. Excessive anti-icing energy consumption will not only bring unnecessary weight burden to the aircraft, but also be detrimental to energy saving and environmental protection.

To solve the problem of high energy consumption, we designed a integration surface with superhydrophobicity and electro-thermal ability[25]. The surface realized reducing over 50% energy consumption for dynamic anti-icing, as shown in Fig.7(a). Meanwhile, we fabricated a slippery liquid-infused porous electric heating coating (SEHC) which exhibited lower ice adhesion compared with superhydrophobic porous electric heating coating (SHP) and porous electric heating coating (PEHC). And we also realized the "heat-induced water film isolation" anti-icing and energy-saving synergies with the "oil film isolation" state (Fig.7(b))[51, 126]. In addition, researchers have devoted themselves into designing the surface with bio-inspired interface characteristics and strong photothermal effect for anti-icing with none-consumption[127-130]. Ref.[111] prepared a multifunctional anti-icing/de-icing coating with superhydrophobic passive anti-icing and electrothermal/ photothermal active de-icing properties and the synergistic ice-phobic mechanism was validated by characterizing the different freezing states of water droplets on surfaces (Fig.7(c)). In Fig.7(c), EPD means electric heating power density.

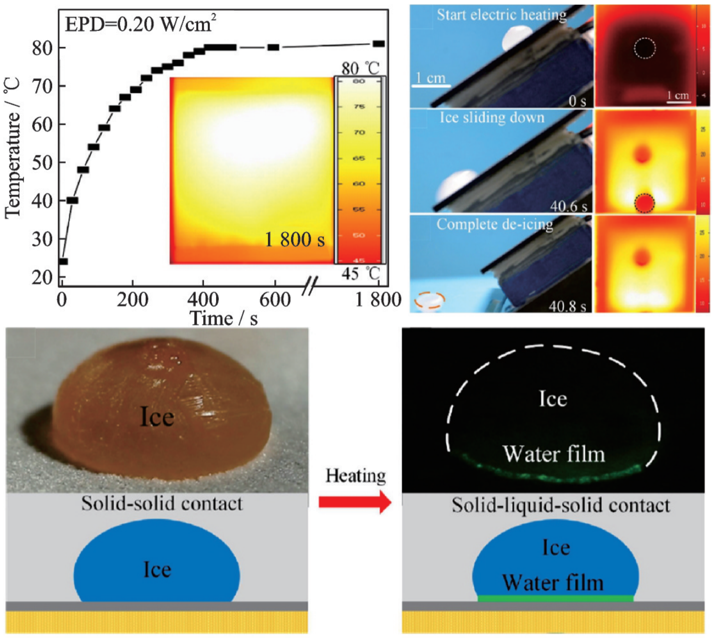
However, for energy saving, the synergy of active and passive anti-icing strategies requires the surface with fast heating rate and durable interface characteristic. Thus, the integration of interfacial and thermodynamic properties needs to be further promoted and improved to fit various requirements in aerospace.



(a) SHS composite electrothermal anti-icing[25]



(b) Oil film composite electrothermal anti-icing[51, 126]



(c) Anti-icing/de-icing composite coating with electrothermal and auxiliary photothermal performances[111]

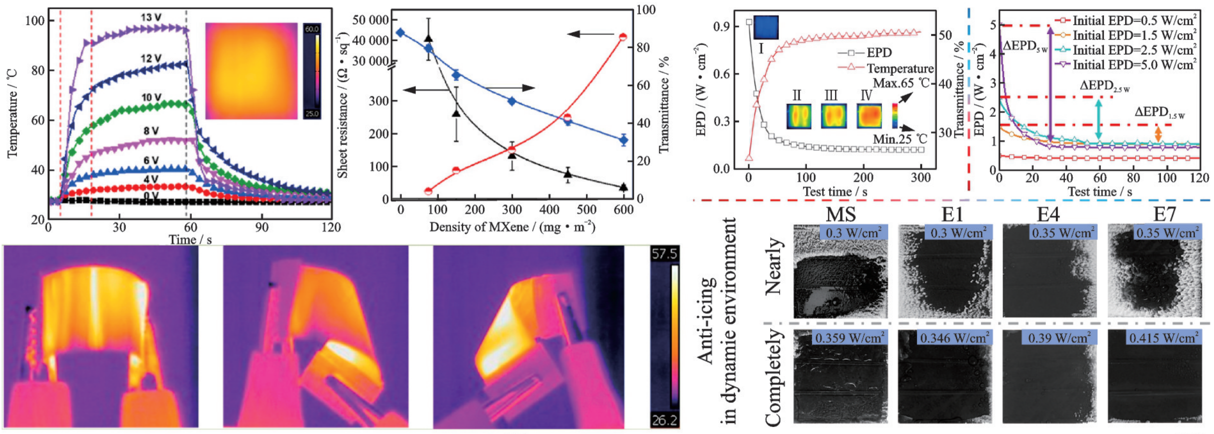
Fig.7 Active and passive integrated anti-icing strategies

3.3 Multi-function compatibility of anti-icing surfaces on aircraft

Considering the multi-function requirements of the aircraft surfaces, such as transparency of windshield, flexibility of moving parts, temperature/flow field/ice sensing ability of airfoils, etc., anti-icing systems on aircraft should coordinate with other functions. The multi-function compatibility of anti-icing surface becomes a new development trend.

Zhou et al.[131] fabricated a kind of flexible hydrophobic MXene-based transparent conducting films based on the excellent electrical conductivity of two-dimensional $Ti_3C_2T_x$ (Fig.8(a)). The obtained film exhibited the balanced optical and electrical properties with a low electrical resistance of 35.1 Ω/sq and corresponding transmittance of 33.4%, showing the rapid steady Joule heating performance at safe voltages (about 100 ℃ at 13 V). We prepared an electric heating anti-icing coating with intelligent self-controlling heating effect and solid-liquid interface conversion effect by combining elastomeric polymer network and phase change lubricant (Fig.8(b))[125]. Compared with the matched sample (MS), the lubricant induced a temperature-controlled transition from a solid state to a slippery surface, reducing the icing adhesion. Furthermore, the positive temperature coefficient effect endowed the coating with an intelligent self-controlling heating effect. Under the conditions of different cold loads and initial heat input, it showed excellent energy saving and temperature self-regulation performance. In addition, anti-icing for irregular surfaces is also significant for aerospace, especially for engine intake. We designed a novel sandwich structural electric heating coating, which realized electric heating properties and high efficiency of anti-icing/de-icing for miniature complex components[132].

With the expansion of application environments, the requirements for function surface have become stricter. Anti/de-icing surface is demanded not only to accomplish its original function but also to realize intelligent self-regulation, environment sensing and integrated design based on multidisciplinary intersection.



(a) Flexible transparent conductive film[131]

(b) Intelligent self-controlling temperature-slip coating[125]

Fig.8 Novel multi-functional active anti-icing/de-icing coatings

4 Conclusions

Aircraft anti-icing technology has become a necessary requirement to ensure all-weather flight safety. The development of large-scale UAVs, stealth aircraft, and integrated material skins makes the need for weight reduction and efficiency enhancement of anti-icing technology more urgent. The peculiar evolution of nature has captured endless imagination and constantly enlightens our scientific work. The development of bio-inspired anti-icing coating technology has made great progress, and the design and development methods for micro-nano multi-level structure and low surface energy materials have become more mature. "Quasi-liquid film/heat-induced water film isolation effect" can achieve the goal of significantly reducing ice adhesion. The integrated design method provides a feasible solution for the integrated anti-icing and energy-saving and efficiency enhancement of aircraft and becomes a new way for high-efficiency anti-icing applications, such as large UAVs, stealth aircraft, and integrated skins.

The bio-inspired anti-icing coating technology is still facing challenges to realize their actual application on aircraft. The research direction to overcome these bottlenecks has become the development trend of the bio-inspired anti-icing coating technology. Mechanical durability enhancement, active/passive integrated energy-saving strategy, multifunction compatibility as well as the large scale/low cost manufacture are the main topics of bio-inspired anti-icing surfaces development, in terms of application on aircraft.

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生物仿生表面防/除冰机理及在飞机上的应用进展

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摘要：飞机表面结冰不仅会增加飞机质量，影响飞行控制，甚至会造成安全事故，是制约全天候飞行的重要因素之一。仿生防冰表面因其低滞后性、不粘接性、成核速度慢、粘冰强度低等优点，近年来受到广泛关注。这些仿生防冰表面如超疏水表面、注入液体的光滑多孔表面和准液膜表面在结冰的各个阶段都实现了优异的防冰性能。然而，对于恶劣的环境，仍然存在许多问题和挑战。本文从结冰过程出发，从仿生的角度综述了结冰成核、液体反弹和结冰粘附的机理，防结冰的应用进展和瓶颈问题。随后，分别对主动式，被动式，主动、被动式一体化防冰技术的可靠性和发展前景进行了探讨。

关键词：机械制造及自动化；飞机防冰；超疏水表面；注入液体的光滑多孔表面；电热涂料