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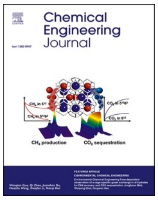

Polysiloxane as icephobic materials – The past, present and the future

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ABSTRACT

Polysiloxane is one of the most favorite polymeric materials used in the emerging field of passive surface icephobicity; This is due to its tailorable softness, hydrophobicity, and relatively high durability. Given the state-of-the-art ice adhesion strength of polysiloxane surfaces has reached a threshold below 1 kPa, a timely survey on the published polysiloxane icephobic surfaces can serve as a valuable reference concerning how far the research field has already progressed and how much potential remains to be exploited for the future development of polymeric icephobic materials. This review categorizes the use of polysiloxane materials for icephobic strategies into three classes according to their surface stiffness. The advantages and shortcomings of each polysiloxane material group are assessed. By scrutinizing the current ice adhesion strength theory, a reference coating thickness is identified, which can be used for optimizing icephobic coating design. A surface icephobicity diagram is also presented, where a lower bound of ice adhesion on a smooth surface is derived, depicting the needs of incorporating different mechanisms to break the theoretical low ice adhesion limit. Finally, the challenges in applying the polysiloxane icephobic materials are discussed, and the possible key research directions are highlighted.

1. Introduction

Ice accretion on exposed surfaces of various infrastructures and vehicles can result in critical safety issues and severe damage. Two extreme examples of unwanted icing catastrophes are the 2008 Chinese winter storms and the Northeastern United States blizzard of 1978, which caused enormous economic losses and casualties [1–3]. The traditional methods for combatting icing problems are by means of active processes, including heating, mechanical deicing, and chemical treatments e.g. anti-freeze liquids or salts. All of these strategies are either inefficient, time consuming, costly or environmentally hazardous [4–6]. For decades, substantial research efforts have been dedicated to the design and fabrication of passive icephobic surfaces and coatings. Such passive icephobic materials are expected to enable ice removal by natural forces, such as wind blowing and gravity, and possess the potential of solving the long-lasting unwanted ice accumulation problems [5,7].

As an analogy to hydrophobicity, the non-wetting surface property, the term “icephobicity” has been introduced for describing surface anti-icing properties in the recent literature [8–12]. Depending on different mechanisms for the anti-icing behaviour of a surface, three definitions of surface icephobicity have been formulated [9]. First, icephobicity can be defined as the ability of a surface to repel incoming water droplets in cold environments, meaning no water and thus no ice [13–16]. Second, icephobicity can be defined as the ability of a surface to delay or prevent ice nucleation and frost formation, maintaining the liquid phase of water on the surface as long as possible [17–31]. Third, icephobicity can also be defined as a surface having low adhesion strength (<100 kPa) to ice [32–36]. Because ice/frost formation is generally inevitable under cold and harsh environments for a given sufficient time, it is thus more practical to live with ice, but with low to super-low (<10 kPa) surface ice adhesion [37]. Icephobic materials design strategies aiming for low ice adhesion are highly promising, which is the driving motivation of this review.

To achieve low surface ice adhesion, it is crucial to understand the basis of interactions between ice and the substrate. Pioneering studies on this regard started in the 1950s [38]. Important factors including intermolecular forces, e.g. electrostatic forces, hydrogen bonding, van der Waals forces, and mechanical interlocking were identified to greatly influence ice adhesion [39,40]. Recent theoretical studies have explored the nanomechanics of ice adhesion and de-icing by employing atomistic modelling [41–44], which further revealed how the strength and the rupture modes of atomistic interactions impact ice adhesion. In parallel, investigations of ice adhesion on a continuum scale have established the key relationships between ice adhesion strength and surface energy,

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elastic modulus, crack size, and so on [10]. Unfortunately, a direct link between nanoscale and continuum scale ice adhesion knowledge is still missing. It is not yet possible to quantitatively predict macroscopic scale surface icephobicity from the atomistic interactions and forces at the ice-substrate interface. Currently, the majority of the icephobicity studies are experimental research, which desire theoretical guidance on icephobicity design. This is especially important for the not yet available multiscale prediction of surface anti-icing performances based on surface parameters of roughness, surface energy, and geometry.

Despite the slow progress in theoretical studies on surface icephobicity, a large variety of icephobic surfaces and coatings have been designed and fabricated in the last two decades. There is steady progress in experimental research on surface icephobicity. Taking the newly developed low ice adhesion surfaces as examples, the ice adhesion strength of the fabricated surfaces is reduced from hundreds of kPa to less than 1 kPa in the lab environment [45,46]. Among all of the published icephobic surfaces and coatings, polysiloxane was very often utilized as the base material, mainly owing to its chemical inertness, tunable mechanical property, hydrophobicity and mouldability [47]. A large diversity of polysiloxane icephobic surfaces have been reported, including those with eye-catching new functionalities of self-healing and lubricant dynamic secretion [45,48]. Despite several general reviews on icephobic materials focusing on introducing the above three definitions of icephobicity [5–7,49–53], an in-depth review solely focused on polysiloxane low-ice adhesion surfaces is not available. Such a review could be crucial to the anti-icing field, given that polysiloxane icephobic surfaces have been a workhouse in this area.

This review aims to establish a systematic classification of polysiloxane icephobic surfaces focused on realizing different strategies of low ice adhesion, covering materials of pure polydimethylsiloxane (PDMS), such as Sylgard 184, fluorinated grafting PDMS, functional polysiloxane, and others. This review organizes the polysiloxane surfaces by different low ice adhesion mechanisms and provides a summary of reported ice adhesion values. It discusses the relationship between the surface properties and the resulting anti-icing performance, as well as challenges and perspectives for the future development of anti-icing materials.

2. Ice adhesion research

The world-wide research outputs on ice adhesion have drastically increased in the last two decades, largely owing to the increasing cost and environmental impact associated with unwanted icing, As shown in Fig. 1. Research articles on surface ice adhesion show an exponential increase after the year 2000 (Fig. 1**a**). As shown in Fig. 1**b**, the first silicone materials used for anti-icing was in 1946 when Loughborough and Haas studied the adhesion of ice on the so-called de-icer surfaces [54]. They investigated the influence of the thickness of the rubber, temperature, and surface roughness on ice adhesion strength [54]. Another pioneering work of testing ice adhesion on various metal and polymeric surfaces was carried out by Raraty and Tabor in 1958 [55]. They found ice formed on a clean metal surface displayed cohesive failure, namely fracture occurring within ice itself rather than at the ice-substrate interface. They also showed for the first time that hydrophobic materials (low interfacial free energy), especially polytetrafluoroethylenes, exhibit favourable low ice adhesion. The importance of interfacial free energy for low ice adhesion was further confirmed by Jellinek in 1962 [56]. In addition, Jellinek also found that the ice adhesion to stainless steel and optically flat fused quartz was affected by the de-icing loading rate and surface roughness. Interestingly, Baker et al. reported adhesion of ice on lubricated steel in 1962 [57], which featured a forerunner of the modern slippery liquid-infused porous surfaces (SLIPS) [58]. There were unfortunately only a few anti-icing related papers published in the subsequent 50 years. Encouraged by the invention of superhydrophobic surfaces (SHS) and its impact on anti-icing in 1996 [59], the research on icephobic surfaces intensified [60,61]. Polysiloxane icephobic surfaces with lower ice adhesion strengths in the range of 100 kPa started to appear (Fig. 1).

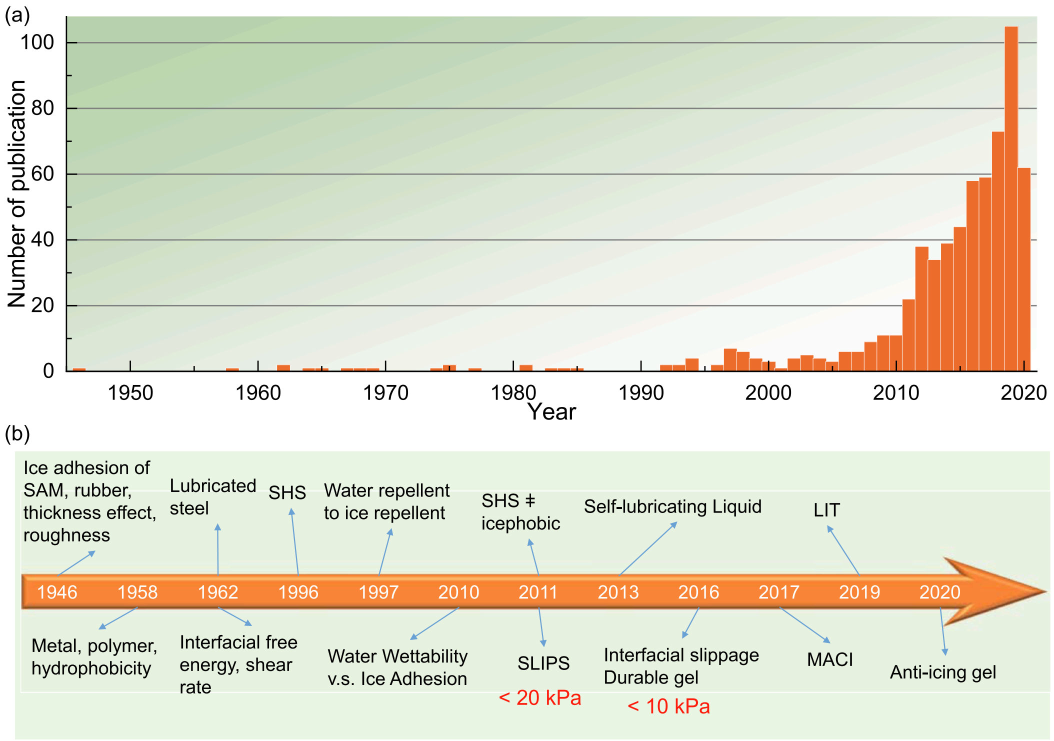

Fig. 1. The history of ice adhesion research. (a) The number of publications on ice adhesion from 1945 to August 2020. A total of 642 studies were found (Data from Web of Science). (b) Timeline (not to scale) of milestones achieved in lowering surface ice adhesion strength in the anti-icing research field. (SAM: self-assemble monolayers; SLIPS: slippery liquid-infused porous surfaces; SHS: superhydrophobic surfaces; MACI: macro-crack initiator; LIT: low-interfacial toughness materials).

A well-known relationship correlating ice adhesion strength (τ) and water receding contact angle (θ_{rec}) on a surface was proposed about a decade ago, namely $\tau \propto (1 + \cos\theta_{rec})$ [62]. Although this relationship is entirely empirical, the theoretical reasoning for its existence is somewhat known. This is so as contact angle relates the work of adhesion for water in the liquid state, whereas τ relates to adhesion on the solid state of water. Both quantities describe adhesion of the same matter, water. Nevertheless, the relationship has obvious limitations in reality. For example, the ice adhesion strength of inherently rough surfaces with very high water receding contact angle, e.g. SHS, can be extremely high due to mechanical interlocking between ice and the surface topography [63]. The later generation of icephobic surfaces, the SLIPS (Fig. 1**b**), were able to avoid ice interlocking and enable low ice adhesion strength by using lubricants, which greatly attracted interest in passive anti-icing researches and applications [58]. Many icephobic surfaces were fabricated following the concept of SLIPS, resulting in lowering the ice adhesion strength below 20 kPa [64]. The SLIPS icephobic surface family were unfortunately found to suffer from a critical issue of durability, namely the depletion of the lubricant film by water or ice [11,65,66]. A variant type of SLIPS using hydrophilic materials were developed in 2013 to address the lubricant depletion issue [67–69]. The new hydrophilic materials allow for accumulating an aqueous water layer for ice lubricating and ice adhesion strength reduction. Such coatings have a similar low ice adhesion effect with SLIPS, but can directly gain their interfacial liquid layer from the ice, and thus is believed to be more durable than SLIPS.

Ice adhesion strength has been reduced to a threshold below 10 kPa starting in 2016 (Fig. 1**b**). Such low ice adhesion strength was first seen by enabling interfacial slippage on icephobic surfaces without sacrificing durability [11]. There are many new notable updates in icephobic coatings recently, for instance, ultra-low modulus organogel for low ice adhesion, low-interfacial toughness materials for large scale deicing [70,71]. Super-low ice adhesion surface (SLIAS) by embedding macro-crack initiator (MACI) into PDMS coatings was also developed, providing an alternative icephobic surface design based on mechanical functions [37,46]. Besides, novel liquid layer generators (LLG), which dynamically melt the formed ice on the surface to generate an interfacial liquid layer, have expanded the possibility of anti-icing applications to extremely low-temperature environments [45]. The so-called low-interfacial toughness materials (LIT) were developed for effective large-scale deicing potential in the field [71]. Most recently, many anti-icing gels, including hydrogel and ionogel, were designed and fabricated [72–74]. These gel surfaces can generate a nonfrozen interfacial liquid, which grants the surface ultralow ice adhesion strength as well as inhibiting ice nucleation and growth simultaneously.

3. Ice adhesion fundamentals

3.1. Intrinsic ice adhesion versus macroscopic ice adhesion

Intrinsic ice adhesion is the atomistic attraction of water/ice molecules to a surface (Fig. 2**a**). Two most basic interactions, namely coulombic and van der Waals forces, are responsible for intrinsic ice adhesion [39,40]. Same as liquid water, ice can form hydrogen bonds with substrates with hydrogen bonding donors and/or acceptors [75], which enhances ice adhesion. The intrinsic ice adhesion is the key focus of atomistic modeling and simulations [41–44]. Due to the limited computation of power, investigation on intrinsic ice adhesion is generally carried out with nanoscale ice with perfect contact with a substrate. Because of the nano-meter scale sizes and the perfect ice-substrate contact interface, intrinsic ice adhesion value found from this scale of investigation are in the order of MPa, *i.e.* many orders of magnitude higher than experimental values. The intrinsic ice adhesion quantified in atomistic modeling can be regarded as the ice adhesion strength at good contact loci of a macroscopic ice-substrate interface.

Ice adhesion quantified in experiments is normally the macroscopic

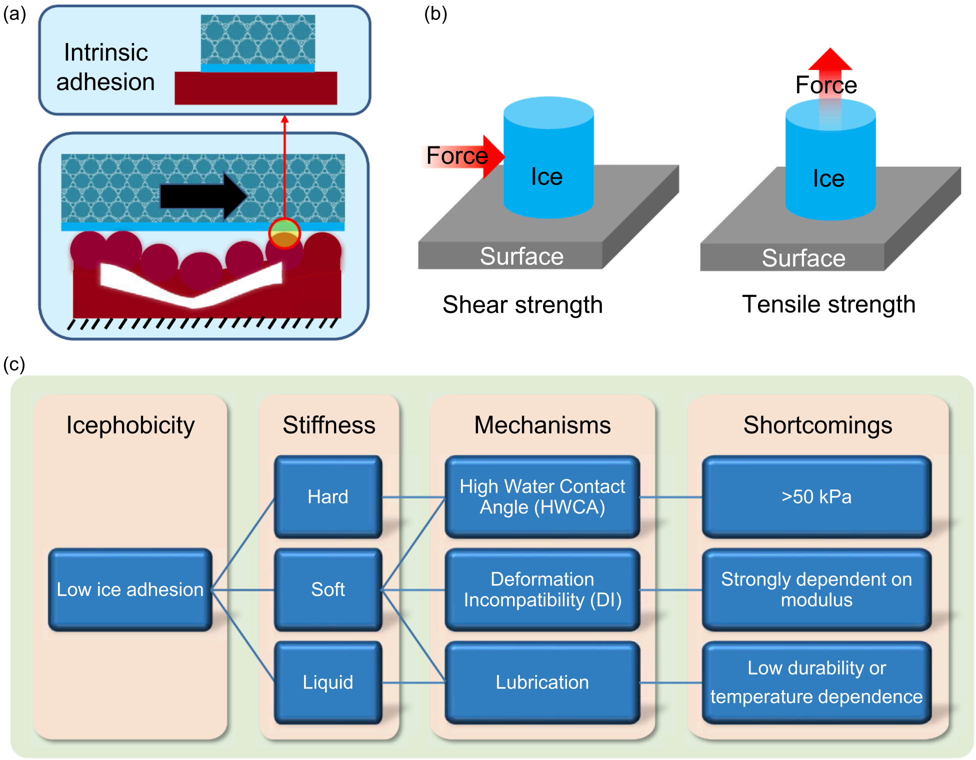

Fig. 2. Ice adhesion and mechanism for low ice adhesion strength. (a) Intrinsic ice adhesion and macroscopic ice adhesion. Possible pore inside the substrate is shown as a white area. (b) Ice adhesion strength: shear strength and tensile strength. (c) Categories of icephobic surfaces by stiffness and low ice adhesion mechanisms. The corresponding shortcomings associated with each category are also presented.

ice adhesion (Fig. 2**a**). Given that a real surface is not atomistic smooth, but rough on different length scales to water/ice molecules, there are voids and imperfect contacts between the ice and the substrate. The macroscopic ice adhesion is thus the average of intrinsic ice adhesion strength at the nanoscale perfect ice-substrate contacting loci and weak ice-substrate interactions at other locations. In addition, many other factors influence the macroscopic adhesion, for instance, mechanical interlocking resulting from surface roughness, the apparent contact area between the ice and solid surface, defects, cracks, and stress concentration.

In experiments, the macroscopic ice adhesion strength is defined as the peak force (F) to remove ice during loading, divided by the apparent contacting area (A), *i.e.* [34]:

$$\tau = \frac{F}{A} \tag{1}$$

Such quantification ideally assumes that the loading force is evenly distributed on the apparent ice contact area and disregards the intrinsic inhomogeneity of any given surface. In reality, a seemingly smooth surface has roughness at different length scales and a variety of local hydrophobicity, which leads to an inhomogeneous landscape of ice-substrate interface upon ice formation [76]. For a given surface, the ice-substrate interface also deviates depending on the ice formation process, meaning ice formed by impacting cold water is different from static water droplets sitting on a surface [77]. It is thus crucial to distinguish ice adhesion on different scales.

It should be further emphasized that cautions are needed in comparing macroscopic to intrinsic ice adhesion strength, given the ice-substrate interfaces of focus are on a different length scale (Fig. 2**a**).

3.2. *Ice adhesion testing method*

There are currently multiple experimental approaches for evaluating ice adhesion strength [34]. Although the idea of applying external force for ice removal is the same, ice adhesion test apparatus reported are normally home-made by diverse research groups, which include centrifugal test [78–81], tensile test [82–84], horizontal shear test [85–89], and vertical shear test [90–95]. The ice adhesion strengths reported in the literature can thus be either shear strength or tensile strength (Fig. 2**b**). Because of the different testing methods, ice adhesion results are generally difficult to compare [96]. Furthermore, ice adhesion testing conditions, for instance, temperature and load rate, in different reports are normally different, which make the ice adhesion results even more puzzling when comparing the data. There is an urgent need in the anti-icing research and application field for developing a standard for the ice adhesion testing standard.

Ice removal is a process of adhesive failure and/or cohesive failure. The adhesive failure of ice occurs at the ice-substrate interface, while the cohesive failure occurs in the ice itself or surface materials. There are ubiquitous imperfect contacts at the ice-substrate interface (Fig. 2**a**), i.e. defects, which play a crucial role in crack initiation and propagation in the fracture process. In this review, the discussions are focused on ice adhesion strength resulting from ice-substrate interface fracture and not cohesive failures.

4. Mechanisms for lowering ice adhesion

By scrutinizing the reported icephobic surfaces, the ice adhesion strength monitored on the individual surface is determined by the condition of the ice-substrate interface. The interface can be hard, soft, and liquid-like (liquid-like can also be considered as ultra-soft, Fig. 2**c**). Hard surfaces and coatings, including self-assemble monolayer [97] and SHS [98], possess low ice adhesion mainly due to their low surface energy (weak intermolecular forces) and possibly low actual contact area with ice. The way to obtain low ice adhesion on hard surfaces is to create the surface with a high water contact angle (HWCA) by either lowering the surface energy via chemical modification or introducing hierarchical nano/micro-structures [99–102]. The ice adhesion strength observed on these hard surfaces are often above 50 kPa [62], and can greatly increase if ice interlocks with the surface nano/micro-structures [103]. Soft icephobic surfaces rely on a different mechanism called deformation incompatibility (DI) for low ice adhesion [37]. DI functions in the ice removal processes and leads to the formation of voids at the ice-substrate interface, which can serve as crack initiators for promoting the crack propagation at the interface and easy detachment of ice [48,70]. In addition to the DI mechanism, ice adhesion strength on soft surfaces can be further reduced by enabling interfacial slippage [11] and MACI [37]. All these soft surfaces for low ice adhesion depend on low elastic modulus, and are suffering from the insufficient mechanical durability [104]. Surfaces with a liquid-like interfacial lubricant layer normally possess ultralow ice adhesion due to the lubrication [64,68,73]. Based on the source of the interfacial lubricants, such surfaces can be classified into extrinsic and intrinsic lubrication types. The SLIPS family is representative of the extrinsic lubrication because an external liquid is trapped at the troughs of a rough or porous polymeric surface [64,105,106]. As briefly notified above, these surfaces with extrinsic lubrication possess very low ice adhesion, but normally suffer from insufficient durability owing to depletion of lubricants. In contrast, intrinsic lubricated icephobic surfaces have polar groups in the hydrophilic polymer that interact strongly with individual water molecules to disrupt the ordering of crystalized ice structure, leading to creation of a lubricating layer by directly melting the ice [67–69]. The low ice adhesion on such surfaces depends on the interfacial non-frozen aqueous layer [107], which diminishes at low temperatures [68]. The details of different low ice adhesion mechanisms will be discussed below.

Polysiloxanes are commonly selected as the base material in the reported icephobic surfaces, due to two attractive properties: (1) hydrophobic (low surface energy) and (2) viscoelastic (softness) [108–113]. One of the typical polysiloxanes is PDMS, dimethylpolysiloxane, or dimethicone (see Fig. 3**a, b** for chemical formula). The hydrophobicity of polysiloxane is critical for creating SHS for low ice adhesion strength [24,35,114]. The high viscoelasticity of polysiloxane has led to the majority of soft icephobic coatings being polysiloxane [11,46,48,70,115]. Polysiloxane was also used in icephobic surfaces with a liquid-like ice-substrate interface. For example, linear and short-chain PDMS (Fig. 3**a**), namely silicone oil, was chosen to serve as a lubricant agent in fabricating SLIPS [116–118]. Besides, cross-linked PDMS (Fig. 3**b**) can be fabricated into a porous substrate to trap the slippery liquid in creating SLIPS [66]. Polysiloxane is chemically stable, non-toxic, non-flammable, and highly tunable in the polymer curing process, which enables it to be easily integrated into different mechanisms and functions for multiple purposes [109,119].

4.1. *High water contact angle (HWCA)*

The hydrophobic nature of polysiloxane is favourite for low ice adhesion. The hydrophobicity of a surface is widely quantified by water contact angle (Fig. 3**c**). For example, Metallic surfaces with polar groups expressing stronger electrostatic interactions with water molecules generally show low water contact angles. In contrast, fluorinated surfaces are highly hydrophobic and interact with water via weak van der Waals interactions, which results in high water contact angles. As such, it is expected that ice adhesion determined by molecular interactions (at least for the intrinsic ice adhesion) is low when a surface is hydrophobic. Indeed as early as in 1958, it was confirmed that hydrophobic polymers exhibited much lower ice adhesion strength than metallic surfaces [55].

Contact angle correlates with the work of adhesion (W_a) for a liquid in contact with a solid, given by:

$$W_a = \gamma_{lv}(1 + \cos\theta_e) \tag{2}$$

where γ_{lv} is liquid–vapor surface tension, and θ_e is the equilibrium

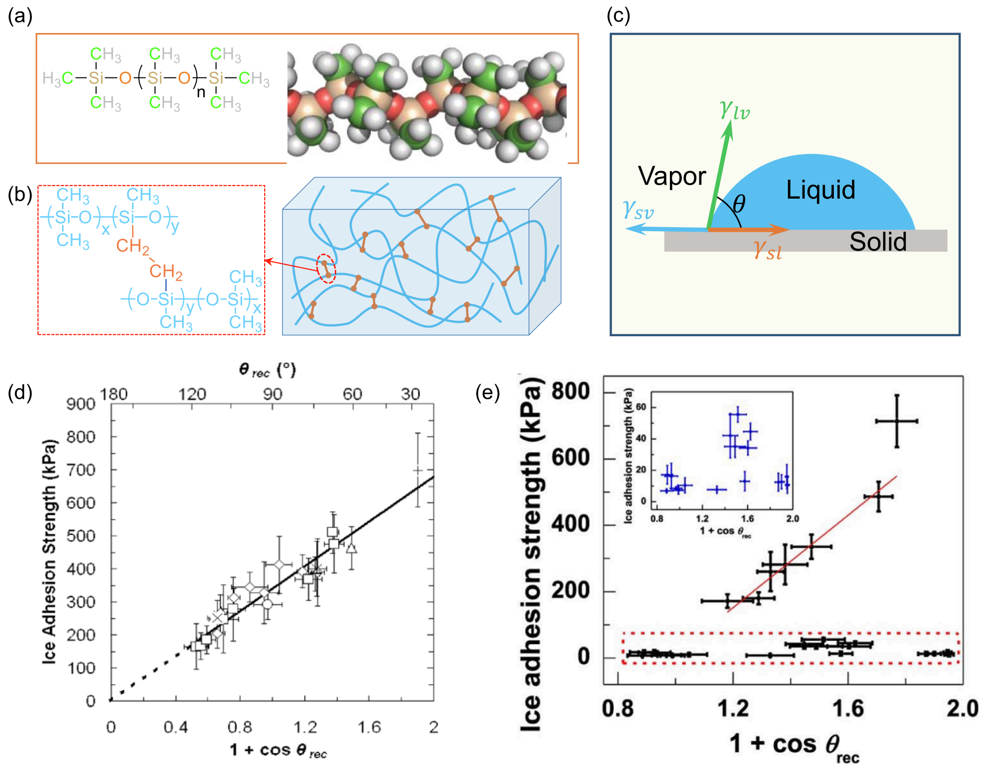

Fig. 3. PDMS-based icephobic materials, the wettability and icephobicity. (a) Chemical structure of pure PDMS and (b) Cross-linked PDMS. (c) Schematic of equilibrium surface free energies at the three-phase contact line and the contact angle, θ, of a three-phase system. (d) Ice adhesion strength against [1 + cos θ_{rec}] for difference surfaces (θ_{rec} is the water receding contact angle) [62]. (Copyright © 2010 American Chemical Society). (e) Ice adhesion strength against [1 + cos θ_{rec}] for difference surfaces [120]. (Copyright © 2017 Springer Nature).

contact angle. It is tempting to use the W_a of liquid water and make a direct connection to the adhesion strength of ice (a solid) to a surface (another solid), and to build or imply a foundation for a theoretical correlation between τ and θ_e. However, compared with water, ice has different molecular interactions with substrates owing to the confinement of the molecules in a crystalline state. As such, the correlation between water contact angle and ice adhesion reported in the literature [62,120] (Fig. 3**d, e**) should be viewed as purely empirical. There is an obvious limitation of the correlation in predicting ice adhesion strength because many other important factors affecting ice adhesion strength are neglected, such as ice-substrate contact area, surface hardness, and roughness, and the related possible interlocking [120].

Despite the shortcomings, the correlation between water contact angle and ice adhesion indicates the important role of atomistic interactions in ice adhesion. There are two ways to weaken the atomistic interactions between ice and a hard surface: (1) lowering the surface energy to decrease the intrinsic adhesion, and (2) reducing the real contact area for intrinsic adhesion by introducing hierarchical micro-nano structures on the surface. Both approaches can lead to HWCA. It should however be noted that HWCA can be the dominating mechanism for low ice adhesion on smooth surfaces, but can fail on rough surfaces with asperity topography. Damage of surface structures by deicing and interlocking between ice and the surfaces can lead to high ice adhesion. The ice adhesion performance of SHS should be taken cautiously.

Due to hydrophobicity of pure PDMS, e.g. Sylgard 184, has low ice adhesion strength [93]. To further reduce the surface energy and ice adhesion strength of the surfaces, various polysiloxane materials, for instance, fluorine-containing polysiloxane and those with surface textures, have been prepared. A series of works on the synthesis of the different polysiloxane-containing copolymers were performed, as shown in Fig. 4**a** [6,121–129], aiming for controlling surface morphology and wettability through tailoring its molecular structures. By using block and graft polyacrylate-polysiloxane copolymers, surfaces with microphase separation thanks to aggregation of polysiloxane segments on top of copolymers were prepared, as shown in Fig. 4**b** [128]. The observed low ice adhesion strength on these surfaces can be directly ascribed to the high water receding contact angle (θ_{rec}), which results from the low surface tension of the polysiloxane segments and also the roughness induced by microphase separation. Fluorine-containing polysiloxane block copolymers were also prepared to maximally reduce the surface energy [121,124]. Because polyhedral oligomeric silsesquioxane (POSS) as SHS had shown great promise [130], a series of POSS-containing coatings for anti-icing were fabricated. The results showed that POSS increased the surface roughness, and decreased the surface energy at the same time, leading to low values of ice adhesion strength (Fig. 4**c, d**) [123,125–127,129].

The water contact angle on smooth hydrophobic surfaces has a limiting value close to 120°. To further increase the water contact angle, hierarchical surface structures must be introduced. The nanostructures formed by copolymers do not result in hierarchical surfaces needed for SHS, owing to their small dimensions [125,128]. Other studies thus employed several methods to create hierarchical structures on polysiloxane surfaces. For instance, silicone rubber was doped with carbon-black, titania, or ceria nanopowder to generate surface textures [131]. The resulting surfaces were SHS and possessed low adhesion to ice. Similarly, superhydrophobic TiO_2-silicone rubber coatings were prepared for icephobicity. The contact angle on the coatings can reach 154.8°, with low contact angle hysteresis of 6.8°. The resulting ice adhesion strength was about 110 kPa [100].

Unfortunately, the low ice adhesion strength on superhydrophobic surfaces was generally not stable and showed a tendency of gradual increase largely due to the damage of the surface texture. The icephobicity of spin-coated and spray-coated superhydrophobic alumina/silicone rubber composites were compared, with ice adhesion being low on the spin-coated sample but high on the spray-coated counterpart [132,133]. The study indicated that the surface icephobicity was strongly affected by the icing conditions and could not directly be correlated with surface superhydrophobicity [132]. Such results were in accordance with earlier studies that condensation of water was observed in the troughs of the hierarchical structures in humid environments, leading to mechanical interlocking and high ice adhesion [63,134,135]. Despite these setbacks in achieving stable icephobicity, there are still

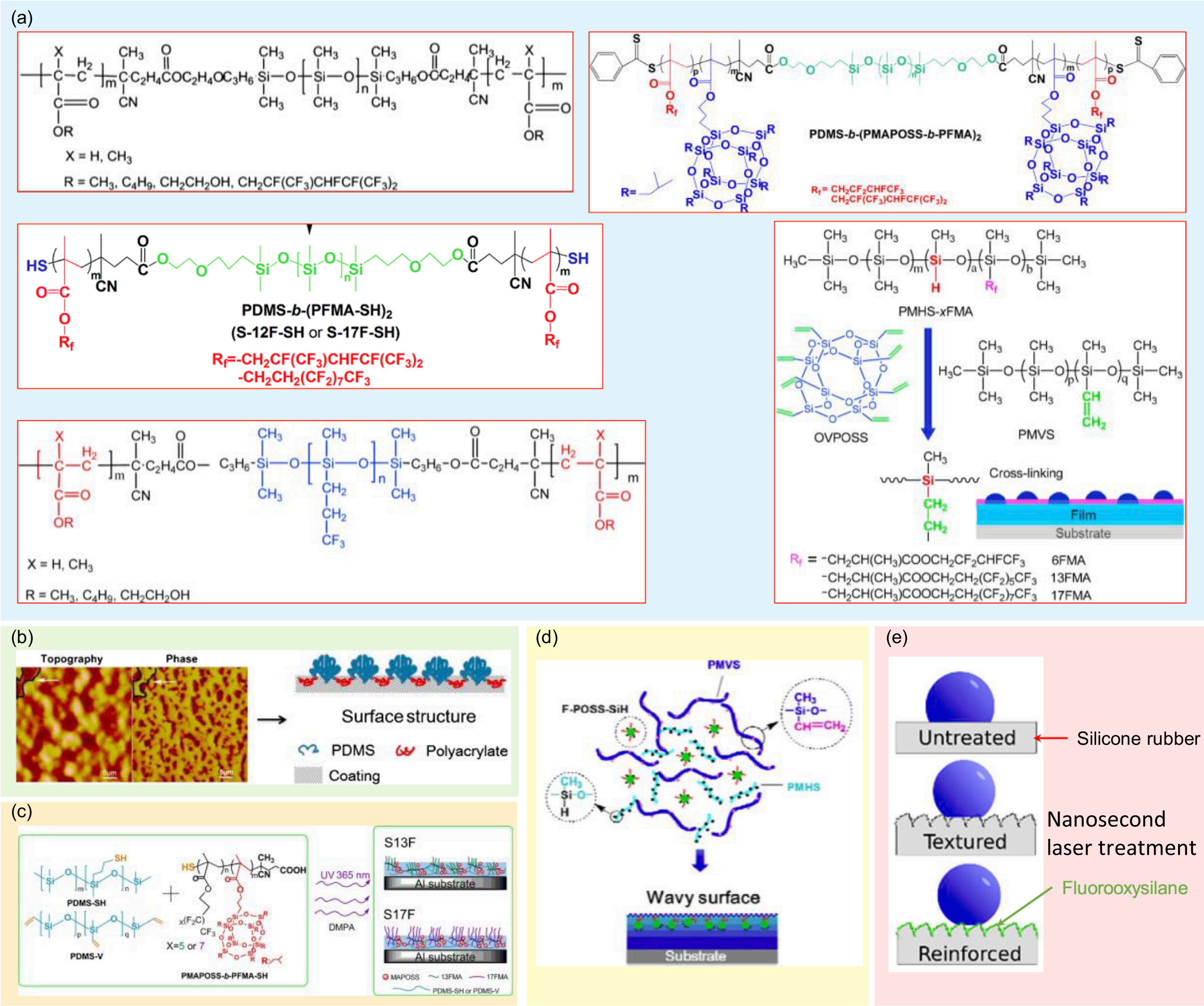

Fig. 4. Surfaces with high water contact angle for icephobicity. (a) Fluorine-containing polysiloxane coatings [121,123–126]. (b) Microphase separation of block and graft polyacrylate-polysiloxane copolymer. Polysiloxane segments prefer to aggregate on the top of the surface [128]. (c) POSS-containing coating prepared by UV crosslinking. Fluorine-containing chains tend to self-assemble on the surface [129]. (d) The introduction of Fluorine-containing POSS into polysiloxane enhances the surface roughness and reduces the surface energy of the resulting materials [127]. (e) Nanosecond laser treatment and grafting of fluorooxysilane on silicone rubber [99]. Figures reproduced from references [99,121,123–129] with permission from corresponding publishers Elsevier, RSC, and ACS.

efforts trying to enable SHS icephobic surfaces to survive in a highly humid atmosphere [98]. For example, SHS was prepared by coating polysiloxane on acid-etched aluminum plates, which was able to maintain ice adhesion of around 100 kPa even under highly humid environments [12]. The plausible icephobicity on these surfaces was attributed to the extremely high hydrophobicity (water contact angle of 166°) that survives even under severe environmental conditions. The so-called reinforced SHS was also prepared by a two-step treatment on polysiloxane, namely nanosecond laser treatment and grafting of fluorooxysilane, as shown in Fig. 4**e** [99]. The as-prepared samples showed contact angles as high as 170° even after outdoor tests, and ice adhesion strength of 64 kPa after exposed to outdoor testing for 7 months in the winter season.

It was speculated that if the texture size of the surface topography is small enough (reaching a critical size), water vapor could not penetrate or condense inside the surface hierarchical structures of fluorine-containing polysiloxane surfaces [103]. The possibility of overcoming the ice condensation inside the nanostructure of SHS in severe conditions via controlling the surface structure and surface energy indeed has been demonstrated [12,99]; however, the mechanism of which yet needs to be substantiated. The critical size of surface topography for stable icephobicity is thus worth further investigation, especially for polysiloxane surfaces. To avoid the degradation of the hierarchical structures of SHS during deicing, the following approaches have been tried with encouraging results [136,137]: including self-healing properties for repairing broken surface [48,95], a self-similar strategy for maintaining same surface texture even after abrasion [136], and tough, flexible and resilient surfaces for recovering deformed surfaces after deicing [137]. The potential of SHS still holds, if the issues of surface degradation and ice interlocking are resolved.

4.2. *Lubrication*

Lubrication for lowering ice adhesion relies mainly on a slippery ice contacting interface. Inspired by *Nepenthes* pitcher plants, slippery liquids were infused to porous substrate, resulting a stable, defect-free, and inert slippery surface [58]. When ice forms on such slippery surfaces, the presence of the liquid layer can serve as lubricant, thus weakening the interaction between ice and underlying substrate. Both the interfacial lubricating layer and the substrate can affect the anti-icing properties of the slippery surfaces. Polysiloxane can play different but equally important roles in fabricating slippery surface. For instance, short and linear PDMS can serve as excellent lubricants, while cross-linked PDMS networks can be the porous media for holding the lubricant components in SLIPS. Additionally, the low surface tension, viscoelasticity, mouldability, and large free volume of polysiloxane are also favourite for low ice adhesion. By using liquid polysiloxane, *i.e.* silicone oil, as a lubricant in polyethylene film with titania nanoparticles, ice adhesion strength as low as 12 kPa was observed (Fig. 5**b**) [105]. Embedding silicone oil microcapsules in latex paints for fabricating coatings led to similarly low ice adhesion [138]. To increase the durability of low ice adhesion, UV-cured polymer networks were incorporated into SLIPS to prevent the easy removal of the infused silicone oil, as shown in Fig. 5**c**, which had led to ice adhesion strength below 100 kPa for at least 13 deicing cycles [139].

PDMS-based materials as a porous or rough substrate for infusing lubricant is another important approach for fabricating SLIPS. Micro-nano silicone rubber was prepared by electrospray and infused with lubricant, which was further used for investigation of the lubricant retaining ability in the icing/deicing cycles [140]. Although ice adhesion on these SLIPS surfaces increased from ~60 kPa to 700 kPa after 20 icing/deicing cycles, the as-prepared heptadecafluorodecyl trimethoxysilane-fluorinated hierarchically micro-structured SLIPS were more durable than the ones without nano-structure and/or fluorination. However, there is significant space for improvement in the anti-icing performance of the prepared SLIPS for practical applications. Porous PDMS was also prepared by using polystyrene template method. After infusing lubricants, the resulting SLIPS showed a similar pattern of low ice adhesion but insufficient durability [141]. Most recently, a liquid layer generator (LLGs) by encapsulating ethanol into the PDMS matrix has been designed and fabricated [45]. The prepared LLGs can release ethanol to the ice-solid interface to form a non-frozen lubricating liquid layer for lowering ice adhesion. More importantly, the ice adhesion strength on the LLG maintained as low as 22.1 kPa, even at an ultralow temperature of −60 °C. Inspired by the skin of amphibians, a lubricant-regenerable PDMS-based SLIPS was developed to enhance the durability of SLIPS with PDMS substrate; The prepared surface maintained low ice adhesion strength below 70 kPa, after 15 icing/deicing cycles (Fig. 5**d**) [66].

To address the depletion of lubricant in SLIPS with porous PDMS, peanut oil have been infused into porous PDMS to develop phase transformable slippery liquid infused porous surfaces (PTSLIPS), as shown in Fig. 5**e**. Thanks to the solid phase of the lubricant at low temperature, the as-prepared PTSLIPS exhibited remarkably low ice adhesion strength of 4 kPa with improved durability (16 kPa after 30 icing/deicing cycles) [65]. Other SLIPS surfaces with solid lubricant rather than liquid oil also confirmed longer terms of low ice adhesion in ice/deicing cycle tests [142], which suggested an encouraging direction for enhancing the durability of the SLIPS family.

One additional advantage of crosslinked PDMS as the base substrate of SLIPS is the large free volume resulted from the swelling polymer chains. Such large free volume in the PDMS matrix can easily accommodate a large amount of infused lubricants. To utilize this feature, a large amount of silicone oil was blended to PDMS prepolymer before crosslinking. The resulting silicone oil-contained surfaces that showed low ice adhesion due to the migration of silicone oil to the surface (Fig. 5**f**) [143,144]. As shown in Fig. 5**g**, self-lubricating organogels were prepared by crosslinking reaction of PDMS in the presence of several organic liquids, aiming for the maximal intake of lubricant. The liquid layers generated on these samples featured an extremely low ice adhesion strength of 0.4 kPa [106]. Similarly, organogel was prepared by simply immersed crosslinked PDMS into a heated paraffin bath, yielding a durable SLIPS surface with ultralow ice adhesion even at −70 °C [145]. Despite the great efforts have been made to improve the SLIPS, insufficient durability remains a common issue. In the same way that interfacial lubrication enables low ice adhesion, it can also greatly weaken the adhesion of SLIPS to the substrate, given that the lubricants

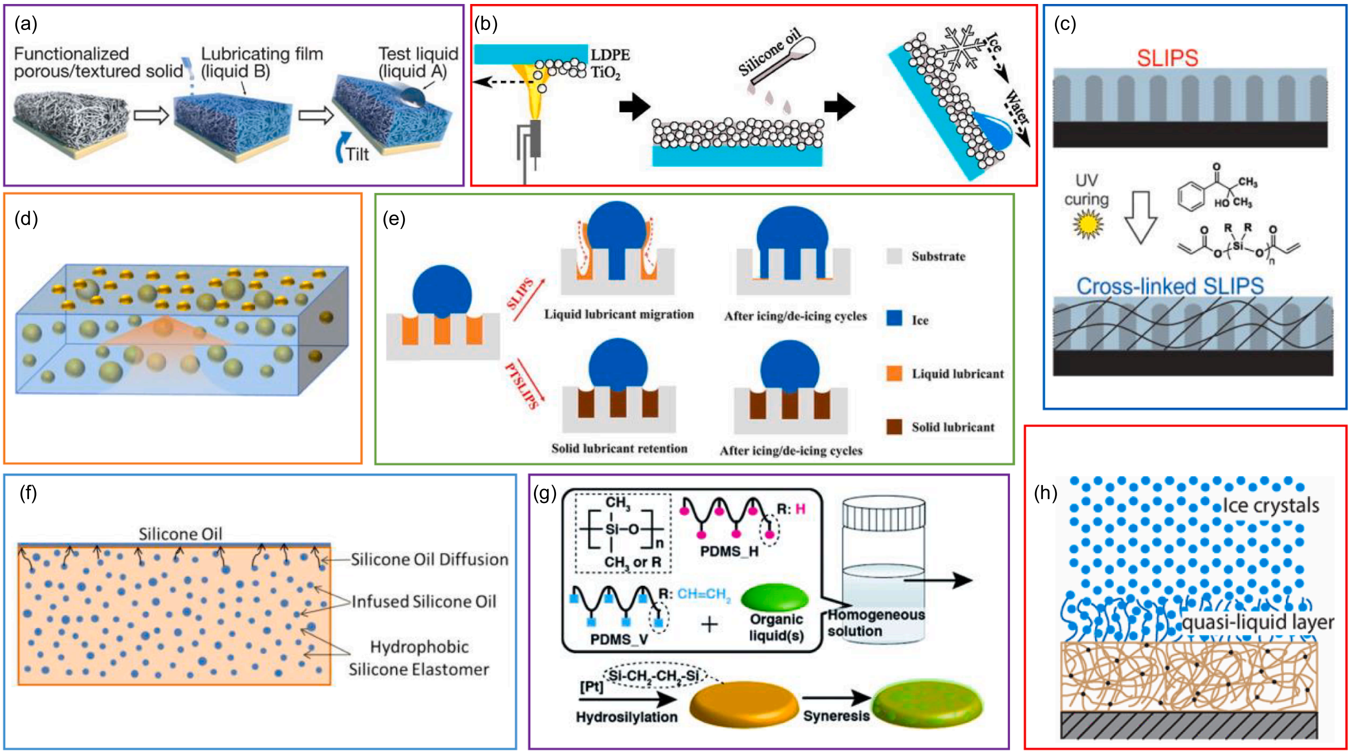

Fig. 5. Surfaces with low ice adhesion enabled by lubrication. (a) Fabrication and wettability of SLIPS [58]. (b) Infusing silicone oil into titania nanoparticles on polyethylene film [105]. (c) Incorporating UV-cured polymer networks into SLIPS to reduce the removal of the infused silicone oil [139]. (d) Amphibians-inspired SLIPS with regenerable lubricant [66]. (e) Phase transformable slippery liquid infused porous surfaces (PTSLIPS) [65]. (f) Blending silicone oil to PDMS prepolymer before crosslinking. The silicone oil can migrate to the surface [144]. (g) The syneresis effects of organogels during crosslinking generating a liquid layer [106]. (h) Bending PDMS-poly(ethylene glycol) (PEG) amphiphilic copolymers into a PDMS matrix. PEG brushes lead to a quasi-liquid layer at the interface to serve as a lubricant, resulting in efficient low ice adhesion [107]. Figures reproduced from references [58,65,66,105–107,139,144] with permission from corresponding publishers NPG, AIP, ACS, Elsevier, and RSC.

exist at both sides of SLIPS facing the ice and the solid substrate. Further work on SLIPS is needed to address such issues.

It should be noted that the lubrication mechanism for low ice adhesion can have a synergic effect with surface superhydrophobicity (HWCA). For instance, oil-infused microtextured silicone surfaces can have a high contact angle and lubrication simultaneously, both of which contribute to low ice adhesion [146]. It is also noteworthy that by incorporating hydrophilic polymer segments on icephobic surfaces, intrinsic lubrication can also be enabled by interfacial non-frozen water [107]. As shown in Fig. 5**h**, blending PDMS-poly(ethylene glycol) (PEG) amphiphilic copolymers into a PDMS matrix can lead to a self-assemble layer of hydrophilic PEG brush at the ice-substrate interface, which can efficiently promote the formation a quasi-liquid water layer for low ice adhesion [107].

4.3. Deformation incompatibility (DI)

The Young's modulus of ice is in the range of 0.3~3.6 GPa [147]. In contrast, soft coatings prepared by silicone usually have Young's modulus lower than 10 MPa [46,48,70,93,115]. The obvious mismatch in elastic modulus between ice and soft surfaces can result in a significant deformation incompatibility under stress during ice removal [93]. Such incompatibility in deformation can lead to the formation of cavities at the ice-substrate interface, thereby facilitating easy ice separation from soft materials. The adhesion of ice to soft silicone rubber surfaces had been studied as early as 1946 [54]. The influence of the thickness of the rubber substrates on ice adhesion strength was investigated by several individual studies in 1946, 1984, and 1994 [54,148,149]. In 2014, the semi-quantitative linear relationship between ice adhesion strength (τ) and $1/t^{1/2}$ (t being the coating thickness of crosslinked polysiloxane) was proposed [93].

The separation of ice from the soft coatings, in essence, is an interface fracture process, in which the shear ice adhesion strength (τ) is theoretically governed by

$$\tau = \sqrt{\frac{EG}{\pi a \Lambda}} \tag{3}$$

where E, G, a, and Λ are the substrate modulus, surface energy, total crack length, and non-dimensional constant related to interface geometric configuration, respectively.[150] Interestingly, for a smooth and homogeneous coating, the following empirical correlation has been used to estimate the ice adhesion [70,151]:

$$\tau \sqrt{\frac{EW_a}{t}} \tag{4}$$

where W_a is the work of adhesion.

As shown in Fig. 6**a**, the mismatch in the elastic modulus of ice and soft coating can lead to stress concentration points at the interface. Under the same stress, thicker coating facilitates larger vertical displacements, which leads to a higher possibility of promoting ice adhesion failure on the coating [93]. The DI mechanism thus paves a solid mechanical design path for achieving ultra-low ice adhesion strength.

Based on the DI mechanism, different surfaces with ultra-low ice adhesion using crosslinked PDMS have been developed. PDMS gels were prepared by hydrosilylation of vinyl-terminated PDMS with hydride-terminated PDMS in the presence of trimethyl-terminated PDMS, with the tuned concentration of trimethyl-PDMS [70]. The influences of shear modulus and thickness on ice adhesion strength of these coatings were then studied. Remarkably, the prepared gel samples with a thickness of

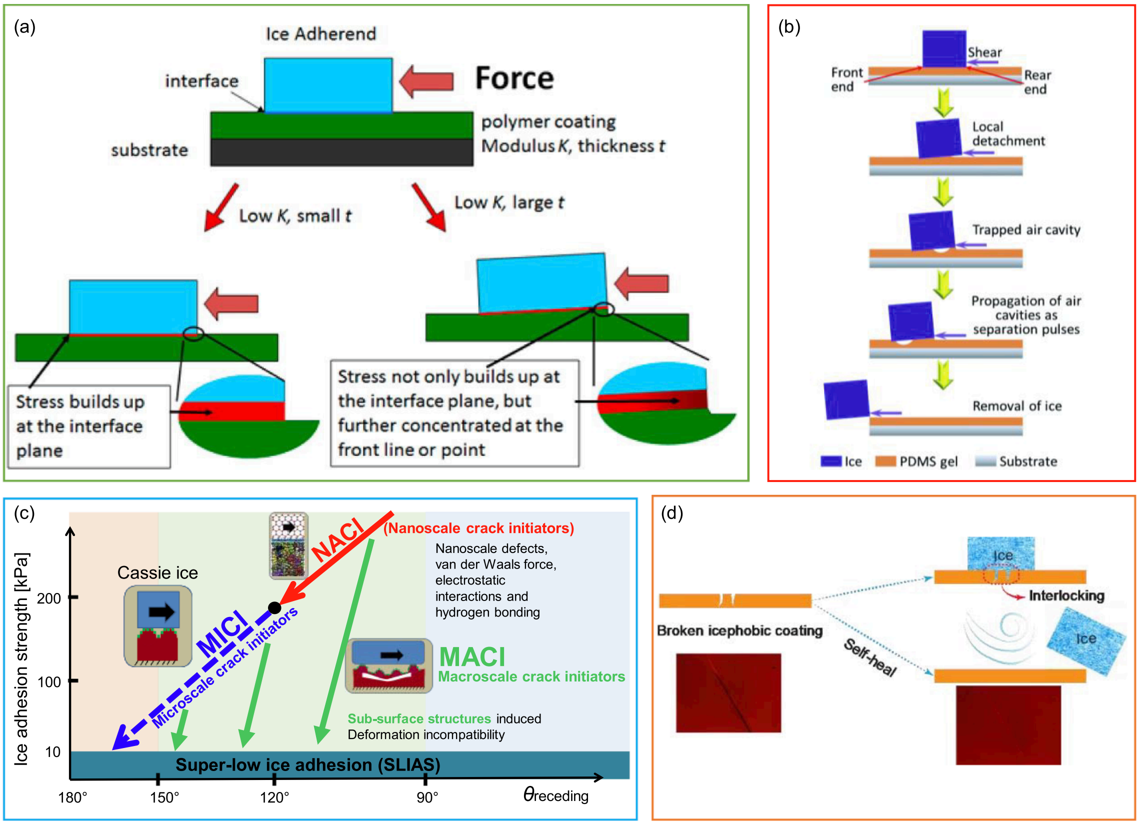

Fig. 6. Deformation incompatibility for low ice adhesion strength. (a) Schematic of a typical deicing process on soft coatings [93]. (Copyright © 2014 American Chemical Society). (b) Formation of cavities at ice adhesion interface on coatings with ultra-low modulus during in deicing [70]. (Copyright © 2016 Royal Society of Chemistry). (c) Design principle of the macro-crack initiator (MACI) for super-low ice adhesion [37]. Nanoscale-crack initiator (NACI) and microscale-crack initiator (MICI) for low ice adhesion are also present in the figure. (d) Schematic of PDMS-based self-healing icephobic materials [48]. (Copyright © 2018 American Chemical Society).

1400 μm and a low shear modulus (10.3 kPa) exhibited ultra-low ice adhesion of 5.2 ± 0.4 kPa and excellent mechanical durability. These coatings with ultra-low shear moduli indeed favored voids formation at the ice-substrate interface under applied forces (Fig. 6**b**), which served as fracture initiators facilitating ice adhesive failure [70,151].

Although soft elastomers possess appealing ice adhesion strength, as low as 10 kPa, their ultralow moduli limit the surface mechanical robustness. The improvement of surface icephobicity purely by using ultra-soft elastomers is thus not realistic [104]. To optimize surface elastic modulus, the crosslink density of PDMS was tailored to enable the so-called interfacial slippage (IS) for low ice adhesion [11]. In these new surfaces, the incorporated uncross-linked polymeric chains were able to enhance the mobility of the ice-substrate interface, altering the boundary condition from no-slip to “nonzero slip” with a certain velocity [152–155]. The high mobility of polymeric chains at the “nonzero slip” interface endowed the coating with ultralow ice adhesion strength [11,104]. For fine-tuned samples of such surfaces, not only DI, but also IS contribute to reducing ice adhesion strength [104].

DI mechanism can also originate from cracking. As indicated in Equation (3), ice adhesion strength is influenced by the total crack length,a. Increasing a is another important strategy for lowering ice adhesion. Based on this concept, a novel integrated macro-crack initiator (MACI) mechanism combining nano-crack and micro-crack initiators was presented recently, as schematically shown in Fig. 6**c**. By introducing MACI into smooth PDMS coatings, the ice adhesion strength can be drastically reduced to an ultra-low level of 5.7 kPa [37]. In addition, sandwich-like PDMS sponges with randomly distributed pores were also fabricated featuring the same MACI principles, which led to record-low ice adhesion strength of 0.9 kPa [46]. The stress-localized (SL) icephobic surfaces utilizing the MACI mechanism were subsequently developed, which can achieve super-low ice adhesion strength in the order of 1 kPa, and at the same time exhibited excellent mechanical, chemical and environmental durability [156].

All the polysiloxane icephobic surfaces that require DI for low ice adhesion are essentially soft. It is worthy to emphasize again that low elastic modulus is unfavorable to the mechanical robustness and durability of the surfaces. To enhance the mechanical durability of all soft icephobic surfaces and coatings, self-healing function was introduced into PDMS-based surfaces by our group, as shown in Fig. 6**d** [48]. The new self-healing icephobic materials have an ultralow ice adhesion strength of 6.0 ± 0.9 kPa, and extraordinary durability confirmed by a very low ice adhesion strength of ~12.2 kPa after 50 icing/deicing cycles. The material was able to self-heal from mechanical damages in a sufficiently short time, which shed light on the longevity of the icephobic surface in practical applications. To further meet complex environmental requirements in the field of anti-icing, a multifunctional coating with low ice adhesion, ultrafast self-healing rate, high transparency, and recyclability was designed and prepared [90].

The trade-off between low elastic modulus and mechanical durability in polysiloxane icephobic surfaces with DI mechanism needs to be optimized. Attempts like accelerating self-healing can be highly beneficial to this category of icephobic surfaces. Furthermore, the adhesion of these ultra-soft materials onto solid substrates is another challenge to be tackled in future studies.

4.3.1. *Reference thickness of coatings based on DI mechanism*

All the reported low ice adhesion utilizing the DI mechanism is based on fracture mechanics theory at the ice-substrate interface. As the ice adhesion strength (τ) on polysiloxane surfaces decrease with the increase in coating thickness (t), the Equation (4) can be simplified as:

$$\tau \quad \frac{\chi}{\sqrt{t}} \tag{5}$$

where the parameter $\chi = \sqrt{EW_{\mathrm{a}}}$ is related to modulus (E) and work of adhesion (W_a). According to Eq. (5), the reduced relationship between τ and t can be plotted, as shown in Fig. 7**a**. The running pattern of the $\tau \sim t$ relationship depicted in Fig. 8a applies to all the PDMS-based icephobic materials. Taking the previous report (Sylgard 184 (10:1) coating [93], 0.025 mm/s probe speed) as an example, the value of χ (green curve in Fig. 7**a**) is 1670 kPa mm$^{1/2}$. Other χ values, for instance, $\chi = 1000$ kPa mm$^{1/2}$, and $\chi = 2500$ kPa mm$^{1/2}$ in Fig. 7**a**, indicate the $\tau \sim t$ relationship in other modified PDMS-based materials with varied E and W_a. With the increased thickness t, ice adhesion strength $\boldsymbol{\tau}$ features a first decrease followed by a low plateau value.

It is feasible to define a reference thickness for icephobicity for polysiloxane surfaces using the DI mechanism, which can provide a good reference in materials fabrication. Although τ decreases with the increase of t, the reduction gradually becomes less obvious at high thicknesses. Due to the limitation of current methods for measuring ice adhesion strength, the system error is usually larger than 10% [62]. As shown in Fig. 7**a**, all the curves show less than 10% reduction in ice adhesion in every 100 μm increase of thickness starting at the highlighted thickness. This highlighted thickness value can be defined as the reference thickness for PDMS-based soft icephobic surfaces. From Eq. (4), the change of ice adhesion, τ, on thickness, t, is

$$\frac{d\tau}{\tau} \quad -\frac{dt}{2t} \tag{6}$$

Using the value of 10% ice adhesion reduction in every 100 μm increase in coating thickness, t, for solving the reference thickness, it is:

$$\left|\frac{d\tau}{\tau}\right| \quad \frac{1}{2t_{10\%}} \times 100\mu m = 10\% \tag{7}$$

which gives: $t_{10\%} = 500\mu m.$

The obtained reference thickness, 500 μm, indicates that it is difficult to achieve obvious improvement in icephobic performance when the coatings exceed this thickness threshold. Thus, caution is needed in polysiloxane icephobic surface fabrication when the thickness of the materials exceeds this reference value.

4.3.2. *Ice adhesion on soft and thick polysiloxane surfaces*

The surface icephobicity quantified by ice adhesion, τ, as a function of $(E/t)^{1/2}$, is plotted in Fig. 7**b, c** [37,46,48,70,91,92,156–158]. The six data points are given in Table 1 are labeled as black squares (among many other squared data points from other publications). Because the six data points in Table 1 can be fitted linearly according to Equation (4) (blue line, Fig. 7**b, c**), the slope of the fitted curve is proportional to $\sqrt{W_{\mathrm{a}}}$. In this case, the slope value of the example linear curve is defined by the work of adhesion of Sylgard 184 (10:1) with a water contact angle of 98.6° [46]. Assuming that all other parameters are kept the same, coatings with the same parameter set of thickness and elastic modulus but varied receding contact angle should also follow the linear relationship, but different slope (different $\sqrt{W_{\mathrm{a}}}$ value). Given that the work of adhesion can be described by contact angle as Equation (2), the relationship in Equations (4) can be further written as:

$$\tau \quad \sqrt{\frac{EW_a}{t}} = \sqrt{\frac{\gamma_{lv}(1+\cos\theta_e)E}{t}} \tag{8}$$

If the highest water contact angle of 120° on a smooth surface was used to fit a linear curve in Fig. 7**b, c**, the curve will represent the lowest bound of ice adhesion which can be achieved on a smooth surface by maximally reducing the surface energy. Then the ratio of the slope (ice adhesion) between Sylgard 184 (10:1, $\theta_e = 98.6^\circ$) and the maximum example ($\theta_e = 120^\circ$) is given,

$$\frac{\tau_{\theta_{rec}=98.6^\circ}}{\tau_{\theta_{rec}=120^\circ}} = \sqrt{\frac{(1+\cos 98.6^\circ)}{(1+\cos 120^\circ)}} = 1.304 \tag{9}$$

Using the data set in Table 1, and contact angle 120° instead of 84.6°, and Equation (9), the lowest bound of ice adhesion on smooth coating is

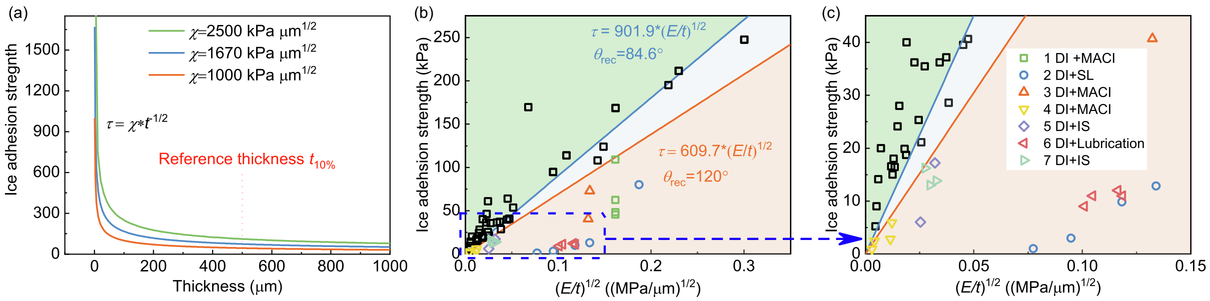

Fig. 7. The relationship between ice adhesion strength and coating parameters. (a) Ice adhesion strength as a function of coating thickness. Reduction in ice adhesion strength after the highlighted the reference thickness is not obvious. (b) Surface icephobicity diagram. Two sets of ice adhesion of smooth surfaces with water contact angles of 98.6° and 120° (blue and orange lines) indicate different slopes of linear fitting [37,46,48,70,91,92,156–158]. The orange linear line, fitted by the maximal contact angle of 120° on a smooth surface, represents the lowest bound of ice adhesion can be achieved by chemical modification for the lowest surface energy. The grey area between the green and the orange lines indicates the possible improvement space of surface icephobicity on the samples given in Table 1 by chemical modifications. All ice adhesions reported on pure smooth surfaces are shown in black squares. The coloured data points highlight the surfaces with combined icephobic mechanisms for lower ice adhesion. The legends indicate the coatings with different low ice adhesion mechanisms. 1 DI + MACI (macro-crack initiator) [37], 2 DI + SL (stress-localization) [156], 3 DI + MACI [91], 4 DI + MACI [46], 5 DI + IS (interfacial slippage) [48], 6 DI + Lubrication [158], 7 DI + IS [157]. (b) and (c) present in different scales. It should be noted that Young's modulus (E) is used here. The reduced modulus (E_r) and shear modulus (G_s) were converted to E by assuming the Poisson's ratio is 0.5. $E = 0.75E_r$, $E = 3G_s$ [48,92] (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

$\tau = 691.5 \times (E/t)^{1/2}$, as the orange line shown in Fig. 7**b, c**. This lowest bound approximately indicates that the maximal icephobicity of a soft coating with certain modulus and thickness can be improved by chemical modification. Interestingly, most of the ice adhesion results on smooth surfaces and coatings in the literature are indeed above this lowest bound (black squares in Fig. 7**b, c**), which reaffirms this theoretical prediction. To further improve the icephobicity beyond this lowest bound, solely chemical modification is insufficient. Other reported surfaces with even lower ice adhesion that can overcome the lowest bound of ice adhesion in the icephobicity diagram are highlighted in Fig. 7**c**, all of which combined mechanisms of MACI (macro-crack initiator) [37,46,91], SL (stress-localization) [156], IS (interfacial slippage) [48,157] and Lubrication [158]) with DI. This predicted lowest bound of ice adhesion can be used as guidance for research directions in fabricating new soft icephobic surfaces.

4.3.3. *Durability of soft polysiloxane*

It should be noted that the ultra-low ice adhesion strength of DI coatings highly relies on the extreme softness of the polysiloxane, which usually leads to poor mechanical properties [104]. In addition, the polysiloxane coatings were demonstrated to possess unsatisfied long-term erosion resistance [159,160]. Both of these deficiencies hamper the commercialization of the soft polysiloxane icephobic coatings in critical applications like leading edges of aircraft and wind blade.

Although a few strategies, such as self-healing [48,90,95,161], have been introduced to improve the mechanical durability of the DI coatings while maximizing icephobicity, new coating designs towards enhanced durability are still in urgent demand. One of the measures is to design icephobic coatings with high toughness against mechanical wear [72,73]. Elevated toughness can resist mechanical abrasion and thus avoid catastrophic damage of the coatings. Applying an anti-corrosion tie coat between the icephobic coating and substrate is also a wise option, which increases the bulk erosion resistance of the coating system, and at the same time improves the adhesion between different layers [162].

5. Ice adhesion strength versus surface wettability and roughness

The relationship between ice adhesion strength (τ) and $(1 + \cos\theta_{\text{rec}})$ is very often referenced in the literature, as shown in Fig. 8**a** [11,37,46,62,92,93,99,101,107,115,123,125–129,146,157,158,163]. Yet the empirical form $\tau = 340 \times (1 + \cos\theta_{rec})$ is only valid for some of hard and smooth surfaces with ice adhesion larger than 60 kPa [62,120]. For example, low-energy surfaces are normally prepared by either chemical modification or surface structural design. When surface asperities are introduced, some surfaces can demonstrate lower ice adhesion compared to smooth surfaces due to the reduction of the real contact area. However, the surface hierarchical texture may lead to mechanical interlocking with ice, which results in much higher ice adhesion strength than on smooth surfaces (the empirical line in Fig. 8**a**) [134].

In order to gain further insight into the reported polysiloxane icephobic surface utilizing three low ice adhesion mechanisms, all the available results in the literature are summarized and compared with consideration of the surface wettability in the Ashby plot (Fig. 8**a**). The ice adhesion strengths reported on polysiloxane surfaces with low surface energy are indicated in green in Fig. 8**a**. For the icephobic surfaces enabled by lubrication mechanism (indicated in blue, Fig. 8**a**), the observed θ_{rec} ranges from 60° to 120°, depending on the lubricant used. Most of the polysiloxane surfaces in this category showed ice adhesion lower than 200 kPa. Polysiloxane soft coatings generally show lower ice adhesion strength than smooth surfaces and can reach an ultra-low ice adhesion strength due to the DI mechanism. The θ_{rec} of smooth polysiloxane surface is commonly smaller than 120°. Hence, the icephobic surfaces and coatings based on the DI mechanism mostly feature the lowest values in the plot in Fig. 8**a** (shown in orange).

As shown in Fig. 8**b** (enlarged low ice adhesion values in Fig. 8**a**), only the surfaces utilizing DI or Lubrication icephobic mechanisms can reach ice adhesion value lower than 20 kPa. It indicates that the only effective strategy for achieving ultralow ice adhesion strength is by lowering the stiffness of surfaces. Specifically, DI requires ultralow modulus to achieve ultralow ice adhesion and the Lubrication requires the presence of liquids (ultralow stiffness) at the interface. As shown in Fig. 8**b**, most of the surfaces that possess ice adhesion strength lower than 20 kPa are located in the light blue region ($80° < \theta_{rec} < 110°$) since this is the θ_{rec}-range of smooth hydrophobic polysiloxane surfaces. One can see that there is a large and empty space in Fig. 8**b** for further exploitation of developing icephobic surfaces. Future studies could focus on the unexplored regions beyond $80° < \theta_{rec} < 110°$.

The introduction of surface texture may increase the water contact angle on the surfaces; however, it can also result in a high roughness, thus subsequently induce the mechanical interlocking between ice and surface. The relationship between ice adhesion strength and root mean squared roughness (R_q) of polysiloxane-based surfaces is plotted to study the effect of surface morphology (Fig. 8**c, d**) [46,48,93,121,123–129,131,145,163–165].

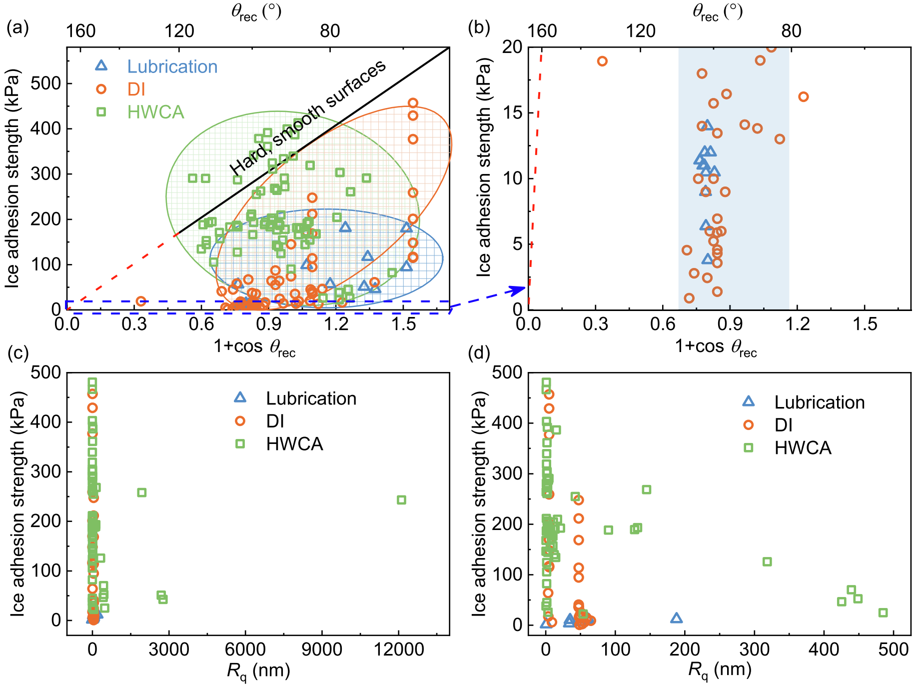

Fig. 8. Ice adhesion versus surface wettability and roughness. (a) Ashby plot of ice adhesion strength versus $(1 + cos\theta_{rec})$ [11,37,46,62,92,93,99,101,107,115,123,125–129,146,157,158,163]. (b) Ice adhesion lower than 20 kPa observed in the literature. All the reported super-low ice adhesion values are concentrated in the wettability range with $80^\circ < \theta_{rec} < 110^\circ$, leaving empty space for future exploitation of surface icephobicity. (c) The relationship between ice adhesion strength and root mean squared roughness (R_q) [46,48,93,121,123–129,131,145,163–165]. (d) Most prepared polysiloxane-based surfaces possess R_q lower than 500 nm.

Table 1
Parameters of smooth Sylgard 184 (10:1) coatings and their corresponding ice adhesion strength [46].

No.	Thickness t (μm)	Modulus E (MPa)	$(E/t)^{1/2}$ ((MPa/μm)$^{1/2}$)	θ_e (°)	τ (kPa)
1	1130.52	2.53	0.04732	98.57	40.64
2	282.63	2.53	0.09465	98.57	94.58
3	212.90	2.53	0.1091	98.57	113.79
4	96.64	2.53	0.1619	98.57	168.47
5	47.96	2.53	0.2298	98.57	211.33
6	28.06	2.53	0.3004	98.57	247.54

Overall, there is no obvious relationship between R_q and ice adhesion strength. Most of the study investigated the surfaces with R_q lower than 500 nm (Fig. 8**d**). All surfaces based on lubrication mechanism presented low ice adhesion regardless of the roughness, indicating that the lubrication mechanism works well in different patterned surfaces. The current surfaces based on DI mechanism all showed R_q lower than 100 nm. Whether the DI mechanism can only function in such roughness range requires further studies. If the DI mechanism can function properly on rougher surface, DI and HWCA can be combined to prepare new generation of icephobic materials.

6. Challenges and perspective

Polysiloxane is a multipotent polymer material with many appealing properties for anti-icing, although there are deficiencies in the current polysiloxane surfaces. Its low surface energy, controllable elasticity as well as high mouldability entitle it to be one of the most favourite polymer materials for fabricating icephobic surfaces. The analysis on the works on polysiloxane surfaces here shows that polysiloxane materials can be a good base candidate for realizing all the state-of-the-art icephobic strategies. Notably, the ultra-soft polysiloxane icephobic coatings enabled by the DI mechanism can yield record ice adhesion strength below 1 kPa, which demonstrates a promising future of polysiloxane surfaces for passive de-icing [46]. By comparing the different icephobic mechanisms, DI appears to outperform its counterparts in terms of ice adhesion and durability. Better surface icephobicity based on the DI mechanism can be anticipated in the near future, if the current active anti-icing research proceeds. It is also important to note, there is a reference coating thickness for low ice adhesion strength governed by an empirical law. This reference thickness can serve as key guidance in fabricating new PDMS-based icephobic coatings. Because the empirical equation used for deducing the reference thickness is not material specific, there could exist a similar reference thickness in icephobic coatings using other non-polysiloxane materials, which yet needs further verification. There is also large space in term of surface wettability (beyond $80^\circ < \theta_{rec} < 110^\circ$) to be explored in fabricating polysiloxane icephobic materials in further work. Key perspectives of polysiloxane low-ice adhesion surfaces are highlighted in Fig. 9, which are further detailed by the following items to shed light on the on-going and future surface icephobicity research:

1. *New mechanisms to lower ice adhesion*. The mechanisms known for low ice adhesion are compatible. It is important to explore the synergy of different icephobic mechanisms in the same coatings for super-low ice adhesion strength [166]. For instance, low ice adhesion can be achieved by decreasing the elastic modulus of icephobic coatings and enabling interfacial slippage at the same time. Consistent works on probing the synergic effects of combined icephobic mechanisms might provide a bright anti-icing future. Moreover, other low ice adhesion mechanisms may await to be discovered.
2. *Lowering ice adhesion strength*. The ultimate and basic question that needs to be addressed is how low ice adhesion can a polysiloxane passive icephobic surface achieves. The newest polysiloxane materials are able to achieve ice adhesion strength lower than 1 kPa [46,106]. However, these values are still 2–3 orders of magnitude higher than 1 Pa range observed on active low ice adhesion surfaces [167]. Basic research focusing on probing the limits of PDMS-based icephobic materials is needed.
3. *Balancing the trade-off between low ice adhesion and coating durability*. To achieve ultra-low ice adhesion strength, compromises may need to be done for the stiffness of the current polysiloxane surfaces. Ultralow stiffness comes with low durabilities, such as SLIPS. As a result, despite a bulk of studies focusing on fabricating icephobic coating and devoting to improve the durability, the progress is still not satisfactory. It is encouraging to see emerging efforts are devoted to addressing the durability issues of icephobic surfaces, for instance,

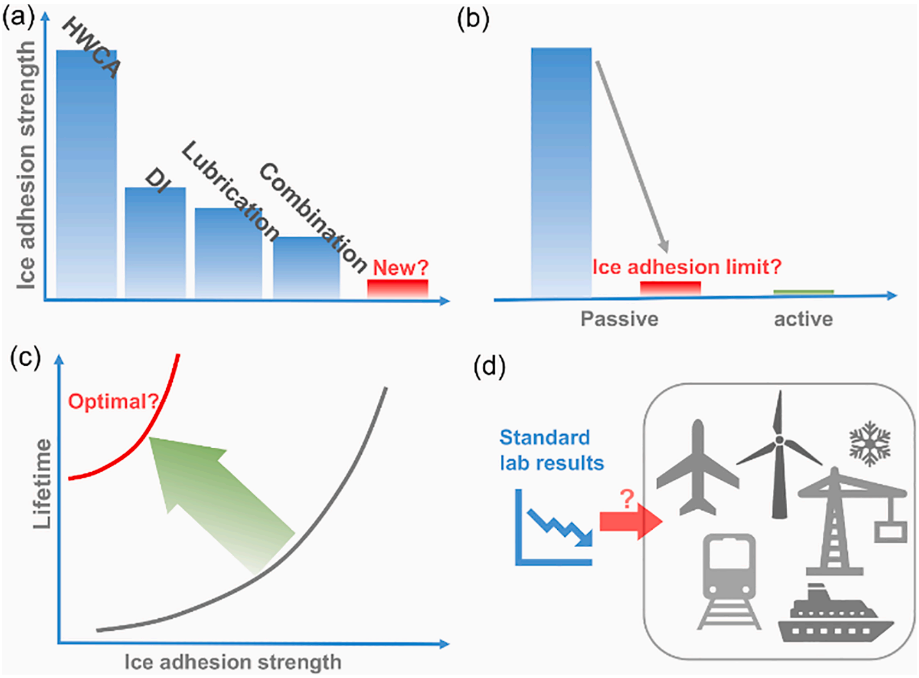

Fig. 9. Perspectives of polysiloxane low-ice adhesion materials research. (a) Anticipated novel low-ice adhesion mechanism in the future, which realizes lower ice adhesion strength than state-of-the-art icephobic surfaces by mechanisms of high-water contact angle (HMCA), deformation incompatibility (DI), interfacial lubrication and/or their combination. (b) Probing the possible low-ice adhesion limit (red) on polysiloxane icephobic surfaces, with the current low ice adhesion strength observed on active low-ice adhesion surface (green) for comparison. It should be noted that the ice adhesion on active low ice adhesion is on 1 Pa scale, which is 2 to 3 orders of magnitude lower than the newest results monitored on PMDS-based surfaces [167]. (c) Improving the lifetime of polysiloxane surfaces with optimally low ice adhesion. (d) Global-wide standard ice testing methods for quantifying ice adhesion strength and application of standardized lab results in the field under environmental conditions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

designing double networks or energy dissipating networks to enhance the toughness of soft coatings against mechanical damage [168–171]. Consistent efforts are still needed for enhancing the durability of polysiloxane icephobic surfaces.

4. *The testing standard for ice adhesion.* Currently, there are varied ice adhesion testing standards in the laboratory, which make results difficult to compare [77,96]. A united test standard should be proposed for both academia and industry [34]. To establish such a standard, research groups from different countries should work closely together to draft initiatives and to present to the related industrial partners internationally. More international conferences on icephobic materials are needed for this purpose.
5. *Lab versus environmental anti-icing conditions?* Nowadays, achieving super-low ice adhesion strength (<10 kPa) is not a significant challenge in laboratory. The true challenge is to bring the laboratory technology into industry. It is not certain that which level of low ice adhesion is enough for practical applications. To meet the requirement of practical applications, passive icephobic surfaces and coatings must possess properties including not only low-ice adhesion, but also mechanical durability, weather resistance, and many others at the same time, which needs yet to perform large-scale tests in the field. Given that there are different types of ice in different situations, some icephobic coatings might have to be developed based on specific applications. For practical anti-icing applications, coating bulk durability as well as the adhesion between icephobic coating and substrate are two of the important problems to be further addressed.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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