Repurposing superhydrophobic surfaces into icephobic surfaces

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ABSTRACT

Ice accretion on surfaces is a major economic concern in the energy and transportation industries, particularly due to the need of frequent maintenance. In contrast to the traditional de-icing approaches involving the use of electrical, mechanical, chemical or a combinatorial approach, the use of icephobic surfaces inspired by nature is a more environmentally friendlier solution. Understanding the fundamentals of the interaction of liquid with solid surfaces at low temperatures is at the core of this topic which motivated the need of this review. While shedding light on the ice nucleation kinetics and ice accretion on solid surfaces, this review provides an overview of icephobicity and provide a pathway to repurpose superhydrophobic surfaces for their use as icephobic surfaces.

1. Introduction

Icing is defined as the formation, build-up and adherence of ice on a surface because of supercooled liquid impingement and freezing. Ice accretion and ice adhesion on surfaces of aircrafts and wind turbines can result in severe operational and safety hazards [1]. Not only that but icing also causes delays, incidents (2.1% of total incidents), and severe accidents (4% of fatal accidents). In-flight ice accumulation causes airflow disruption, decreasing control and performance. Large pieces of ice separating during flight cause problems to engines and hit propellers. Frozen contaminant can jam control surfaces. A clean and smooth wing surface is essential for natural laminar flow (NLF), envisaged in next aircraft generation to reduce drag by 20%, fuel consumption by 4%, and CO_2 emissions. This follows manufacturers' and operators' current direction towards UK and international commitments (CCC, ETS, IATA): CO2 emission reduction to 2005 levels by 2050 and improved fuel efficiency of 1.5%/year.

De-icing systems can add up to 5% of aircraft weight, which increases an aircraft's carbon footprint by 4%. De-icing fluids are also used on the ground to remove ice from aircraft prior to take-off (ICAO regulations), with expensive operations.

Just like aircrafts, wind turbines are susceptible to ice accumulation while operating in cold areas. Ice accumulation on wind turbines reduces energy output, causes mechanical and electrical problems, interferes with monitoring and control, and creates safety issue [2]. The uneven formation of ice on wind turbine blades can cause imbalances, leading the parts of a turbine to wear out faster. It may also cause turbines to shut down as a result of the vibrations. Turbines may not be able to be restarted for hours or even days due to excessive built up and accumulation of ice. To combat this problem, measures have been developed for de-icing and anti-icing measures [3]. De-icing is the process of removing ice from aircraft/wind turbine surfaces using heating, vibration (contact or non-contact), mechanical means, and sprayed-icing fluids. Anti-icing, on the other hand, is a preventive intervention that delays or lowers ice build-up on surfaces such that de-icing is not necessary or takes less time/energy. Anti-icing can be accomplished by spraying anti-freezing fluids on a regular basis or by applying icephobic coatings, which are permanent anti-icing coatings [3]. Thus, icing represents a complex and expensive problem in different industrial and energy applications - aircraft, wind turbines, power lines, causing incidents and severe accidents. Use of passive ice-repellent surfaces is expected to be a significant opportunity for preventing ice from sticking on aircraft, ensuring an ideal clean surface for NLF with potential global savings of £3BN and 30 M tonnes of CO_2 emission/year. This review gives a brief overview on ice nucleation and ice accretion of solid surfaces, icephobic

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surfaces and application of superhydrophobic surfaces as icephobic surfaces.

2. Ice nucleation and ice accretion

Understanding the ice formation process and the accompanying means of limiting ice formation and development is critical for designing novel ice repellent strategies. When water is chilled to 0 °C (dew point) under atmospheric pressure, it undergoes a phase transition as shown in Fig. 1 leading to the formation of ice.

The transformation of water to ice can be explained as heterogeneous ice nucleation. Fig. 2 shows a schematic illustration of the homogeneous and heterogeneous nucleation of ice.

Pure water may normally supercool to around −40 °C before homogenous nucleation develops in the supercooled condition (under ambient pressure) [5]. If there is no large-scale disruption in the system, supercooled water can persist as a liquid mass. The first crucial stage in homogeneous nucleation in undisturbed pure water is the spontaneous creation of many long-lasting hydrogen bonds between water molecules in an area [6]. These water molecules produce ice nuclei that develop slowly and eventually change shape, until more stable hydrogen bonds proliferate quickly across the system, enabling water to transition from a liquid to a solid state [6]. The most typical route for ice to develop is by heterogeneous ice nucleation, or ice crystallisation on a surface. It may occur at temperatures greater than those required for homogenous nucleation due to the presence of contaminants and the interfacial topography and chemical composition. The complexity to develop anti-icing materials stems from the underdeveloped science that a combination of these factors plays which motivated the need of this review.

When a supercooled liquid droplet comes in contact with a sub-freezing item, the object serves as the point of nucleation for formation of ice, and any supercooled liquid that comes into touch with it freezes instantly. The ice will develop more rapidly if the item is colder, or the droplet is smaller. The average droplet size, liquid water content, and air temperature are used to quantify
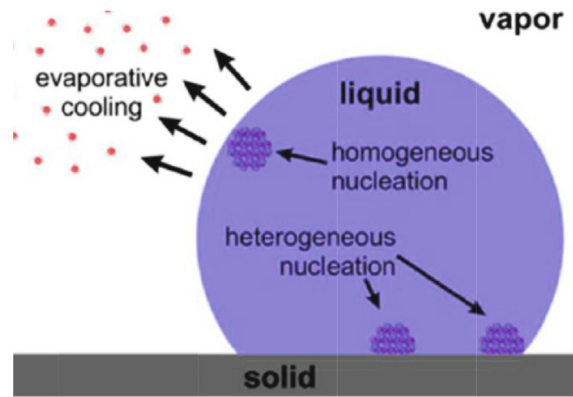
Fig. 2. Homogeneous and heterogenous ice nucleation [4].

Table 1
Basic properties of different forms of ice.

Ice type	Description
Rime ice	−10° to −20 °C freezing of small, supercooled water droplets (5–70 μm) white brittle and feather like
Glaze ice	0° to −10 °C freezing of large, supercooled water droplets (70 μm to few millimeters) clear, dense, and hard ice
Mixed ice	Mixture of glaze and rime ice

icing conditions. Temperature and altitude affects the quantity and distribution of supercooled water droplets and ice crystals inside the cloud as shown in Fig. 2 [3].

The basic conditions in which different type of ice, namely, Rime ice and Glaze ice or a mix of these forms are summarized in Table 1.

When supercooled water droplet with a mean diameter of less than 50 μm impinge the skin of an aircraft that is already at sub-freezing temperature, it can result in the formation of rime ice
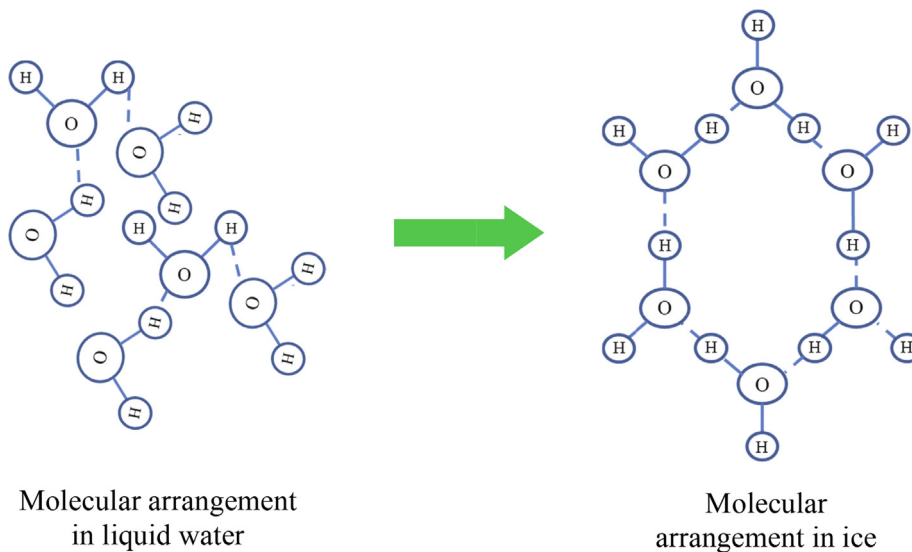
Molecular arrangement in liquid water   Molecular arrangement in ice

Fig. 1. Phase transition of water to ice.

which is brittle and has a feather like structure. Rime ice tends to retain air inside its structure due to its rapid freezing, giving it a milky look and making it more malleable than other types of ice.

Glaze or clear ice forms between 0 °C to −10 °C when super-cooled water droplets of mean diameter greater than 70 μm impinge on an aircraft skin that is already at subfreezing temperature. On contact, just a small portion of the supercooled water droplet freezes [7,8]. The heat generated during the first freezing by impact with a portion of the droplet raises the temperature of the aircraft skin to 0 °C. A considerable part of the droplet is allowed to spread out and mix with other droplets before freezing slowly and completely [3]. As a result, a solid sheet of pure ice develops, with no air bubbles to impair its structure. As additional ice forms, it forms a single or double horn shape that protrudes from the wing, tail surface, antenna, or other surface on which it forms. The airflow is substantially disrupted by this unusual ice formation, which causes an increase in drag. Mixed ice occurs in the freezing zone between 0° and −20 °C (-40 °C in extreme conditions). Mixed ice occurs in the freezing zone between 0° and −20 °C (-40 °C in extreme conditions) where both small (mean diameter of less than 50 μm) and large (mean diameter of greater than 70 μm) supercooled water droplets coexist as shown in Fig. 3. Mixed ice has features of both glaze ice and rime ice [3].

3. Icephobicity

As shown by Fig. 4, researchers have defined icephobicity in various ways for instance (i) as the ability of a surface to rebound or roll-off supercooled water droplets [9], as (ii) decrease of ice adhesion strength [10–12], (iii) delayed heterogeneous ice nucleation [13–16], (iv) delayed freezing time and (v) impeded frost formation. Here an overview of these aspects has been provided in light of a superhydrophobic surface.

3.1. Surface wettability

It has been suggested in the literature that superhydrophobic surfaces possess icephobic behaviour too [17,18]. Superhydrophobicity was initially discovered in nature to be a characteristic of the surface of lotus leaf and other non-wetting plant leaves [2]. A superhydrophobic surface has low surface energy and a static water contact angle of more than 150° and a contact angle hysteresis (CAH) of less than 5° [19] (See Fig. 5). Droplets on the substrate surface may offer distinct wetting states due to micro and nanos-
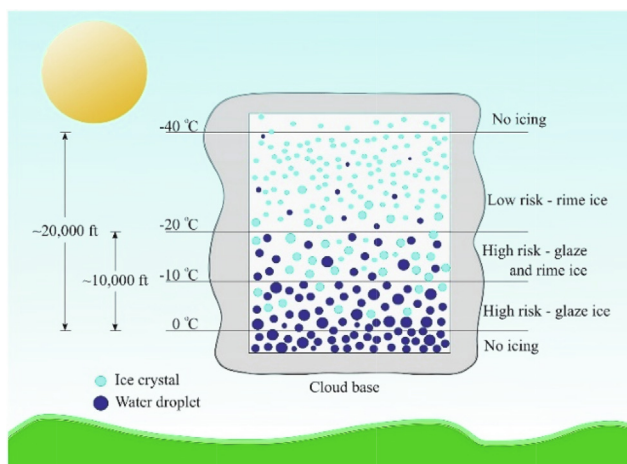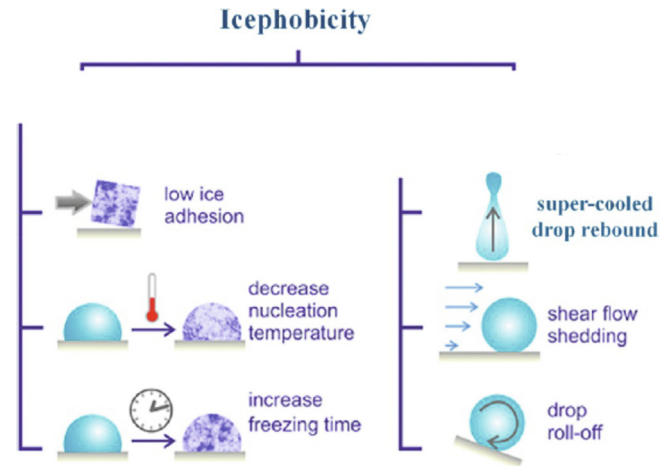
Fig. 4. Present icephobic surface approaches [2].

cale geometrical features coated with diverse low-energy chemical components on top of which topography of the surface can also play its role. Water wettability on superhydrophobic surfaces may be explained using classic Cassie–Baxter (CB) [20] and Wenzel models [21] as shown in Fig. 6. According to Wenzel state [21], water droplets may create a continuous wetting interface along the topography of a solid surface. Water droplets in the Cassie–Baxter state will not entirely attach to hydrophobic surfaces and will remain on a composite interface. Micro and nanoscale topography of superhydrophobic surfaces allows air to be trapped between the surface roughness and the impinging supercooled liquid i.e., Cassie–Baxter state. The entrapped air may bounce off the striking supercooled water droplets before freezing. On a rough substrate, liquid droplets may also exist in the Wenzel state, where the liquid droplet displaces the air and completely wets the substrate. Some studies have shown a shift from a Cassie–Baxter to a Wenzel state during the icing stage while other have argued that a reduced ice adhesion strength on superhydrophobic surfaces indicates water freezing in the Cassie–Baxter state [22]. The ice repellence or icephobic property of a solid surface is directly linked to the interfacial energies of water and the solid phase, as well as the topography (roughness) and chemistry (surface tension) of the surface.

Shen et al. [9] have shown that the single nanostructured surface exhibit very low wettability and outstanding icephobic ability as compared to the hierarchical micro nanostructure surface. This is because a single nanostructure entraps more air pockets below the droplets creating superior water repellency. However, when exposed to high relative humidity, superhydrophobic or hydrophobic surfaces cannot always retain their icephobic properties. To an extent, this aspect can be overcome especially when a lubricant-infused slippery surface is applied for instance, Yin et al. [26] reported lowest ice accretion on slippery surfaces when compared to hydrophobic and superhydrophobic surfaces.

3.2. Ice adhesion

It is vital to reduce the adhesion of ice with the surface, so that it can be blown off the surface by wind, gravity, or vibration. Icephobic surfaces have an ice adhesion strength of less than 100 kPa [27]. According to several studies, chemical bonding and electrostatic forces at the ice-solid interface are the two key factors that influence ice adhesion. Short-range interaction forces, such as covalent, electrostatic, and/or metallic forces (e.g., van der Waals forces), exist at the atomic or molecular level [28]. Mechanical
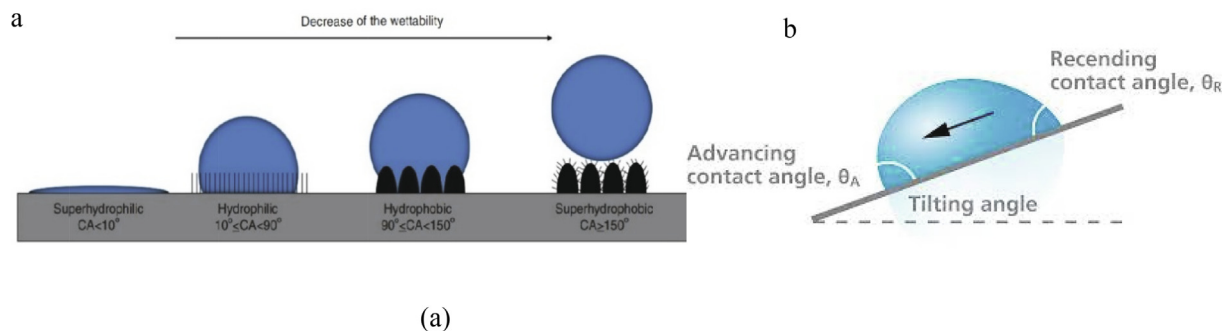
Fig. 3. Ice development inside clouds and the risk of aircraft freezing consequently [3].

Fig. 5. (a) Types of surfaces categorized by surface wettability [23]; (b) Contact angle hysteresis [24].
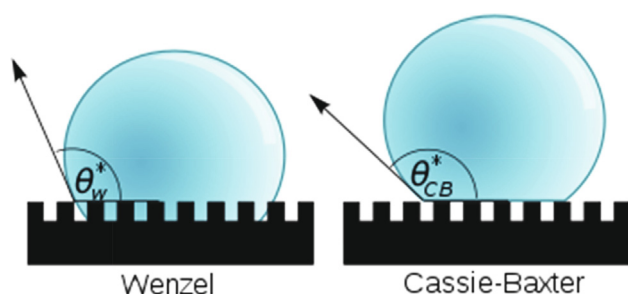

Fig. 6. Wenzel and Cassie-Baxter wetting models [25].

adhesion may also happen when tiny asperities at the interface between two materials form a mechanical interconnect. Varanasi et al. [29] investigated the influence of chemical composition and surface topography on ice adhesion strength in a systematic way. They reported maximum adhesion strength on micro-textured superhydrophobic surfaces, with values exceeding those on smooth surfaces. On the other hand, hierarchically textured surfaces had lower ice adhesion, although having a greater adhesion strength than smooth hydrophobic surfaces and nano-textured superhydrophobic surfaces. The development of Wenzel ice, which happens when micro-droplets solidify inside an air layer, is responsible for the high ice adhesion on such surfaces. Therefore, enhanced anti-icing qualities and reduced economic losses are anticipated to come from an efficient and appropriate design of a solid surface structure. Studies [30,31] reported that surfaces with nanoscale roughness prevented water droplets to condense in the air layer and thus decreased ice-solid contact interface, resulting in low water wettability and ice adhesion to solid surfaces. Differences in techniques (shear, tension, or mixed mode fracture) for assessing ice adhesion strength might be a cause of dispute in judging icephobicity based on ice adhesion strength.

Oil infused surfaces offer a new bioinspired soft material technique for reducing ice adhesion. The infused oil or lubricating liquid introduces a rough structure to generate a continuous liquid layer on solid surfaces, preserving strong ice resistance while also allowing droplets on an aqueous layer to roll off without being obstructed [32–34]. Nature inspired materials can be a route to investigate this concept in more details [35].

Wang et al. [36] introduced liquid layer generators, which dynamically secreted a lubricating ethanol layer at the ice–solid interface after ice formation, yielding low ice adhesion strength. They performed atomistic modeling and simulations to depict the ice adhesion reduction effect of ethanol layers with different thicknesses at the ice–solid interface at various temperatures.

The molecular genesis of icephobicity was investigated by Zhao et al. [37] using molecular dynamics simulations at the interface between distinct water phases (ice, liquid, and vapour) and a solid substrate. The connection between inherent contact angles of a substrate revealed that both hydrophobic and hydrophilic materials have icephobic qualities and may be textured to reduce ice adhesion strength.

3.3. Delay ice nucleation and freezing time

According to researchers who have investigated ice nucleation process, icephobicity is defined as the capacity to inhibit or delay ice nucleation and deposition on surfaces. Schutzius et al. [4] reported that the nucleation temperature remains unaffected by surface roughness when just a portion of the surface area is filled by nanoscale pits below a critical stable radius. They also reported that, long delay of heterogeneous nucleation is possible theoretically by designing a surface with an array of nanoscale pits with small asperities using the existence of a quasi-liquid layer and lowering freezing point of water. It is to be noted here that it is now possible to fabricate such nanoscale surfaces using atomic force microscope with predictive deterministic accuracy to study these nanotechnological aspects in greater details [38].

On the other hand, Wilson et al. [39] suggested that smooth and chemically homogenous lubricant-based surfaces may decrease or eliminate ice by minimis[17]ing the number of possible nucleation sites. Alizadeh et al. [40] reported that the ice nucleation delay caused by decreased water–solid contact area is only noticeable at modest supercooling temperatures. However, ice nucleation in the bulk and at the air–water interface become equally relevant as the supercooling temperature approaches – 40 °C.

The heat transfer rate between a water droplet and a substrate determines how long it takes for a water droplet to freeze when it meets a surface [1]. In case of superhydrophobic surfaces, small interfacial area and low heat transfer rate between water droplet and surface delay the ice formation. Rougher superhydrophobic surfaces have limited number of sites at the solid-liquid interface for heterogeneous ice nucleation.

Zhang et al. [41] used molecular dynamics simulations to study icephobicity of the graphene surfaces functionalized by sodium ions, chloride ions, or methane molecules. The simulation results showed that, depending on the quantity of functional groups and the type of functional group, the functionalized surfaces have a reduced ice nucleation ability than the smoother graphene surfaces. These studies showed that functionalizing surfaces with specific ions or molecules can improve their icephobicity.

3.4. Impede frost formation

Condensation of the vapour phase followed by freezing causes formation of ice crystals which becomes denser with time and are known as Frost [42]. Water vapour in the air turns into liquid and condensation happens as the temperature falls below the

dew point. Furuta et al. [43] reported that water contact angle decreases with decreasing surface temperature in a humid environment which implies a shift from Cassie–Baxter to Wenzel on a cold rough superhydrophobic surface. They have also reported that water adsorption caused a shift in the interfacial free energy of the solid–gas contact. The change in wetting behaviour of the superhydrophobic surfaces is due to the capillary condensation of liquid water in the crevices present in the textured surfaces. To avoid the formation of so-called "Wenzel ice" in icephobic applications in a humid climate, it is critical to avoid this transition. Farhadi et al. [44] found that as water vapour condenses on surfaces under high humidity, ice adhesion may increase three times compared to the reference compound. Wang et al. [45] produced superhydrophobic surfaces which maintained water contact angles of more than 150° across a broad temperature range of −10 °C to 17.5 °C which can be applied in icephobic applications. Frost development may be delayed in the same way that water droplets freeze slowly. Liu et al. [46] observed a 55-minute delay in frost development on superhydrophobic surfaces. They also discovered that the frost that forms on superhydrophobic surfaces has low ice adhesion strength and is easily removable.

4. Authors perspective

Many attempts have been made to provide a cost-effective, energy-efficient, and environmentally friendly solution to the icing issue of aeroplanes and wind turbines. Passive anti icing solutions, such as icephobic coatings may mitigate the detrimental effect of using de-icing fluids for aircraft and wind turbine de-icing and anti-icing purposes [3]. Superhydrophobic surfaces can be promising alternatives of de-icing fluids, and other expensive mechanical and thermal de-icing/anti-icing methods [14,47,48]. When the roughness (topography) and chemistry (surface tension) of a surface meet the criteria of air to be trapped in the surface and the surface energy to be much lower than that of water, the surface is said to be superhydrophobic. While these characteristics are generally established, the exact circumstances and design attributes that lead to superhydrophobic behaviour must be explored further. Although subtractive manufacturing techniques like micro milling and ion beam etching [49] will give greater control and precision in developing superhydrophobic surfaces, they will be restricted by production scalability. Moreover, it was reported that the hierarchical structures of the superhydrophobic coated test surface were progressively degraded because of the constant impingement of supercooled water droplets [50]. Water droplets on the degraded superhydrophobic surface easily undergo transition from the partly wetted Cassie-Baxter state to the fully wetted Wenzel state as the hierarchical textures/ roughness vanishes. Due to the interlocking between ice and the textures, it would be quite difficult to remove the ice once it freezes inside the surface textures in the Wenzel condition. As a consequence, the ice adhesion strength to the superhydrophobic coated test surface increases exponentially as the rain erosion experiment lasts longer [50]. A way of producing permanent superhydrophobic coatings is extremely desired. In general, a composite structure composed of micro- and/or nano-scaled hard particles embedded in a low sur-
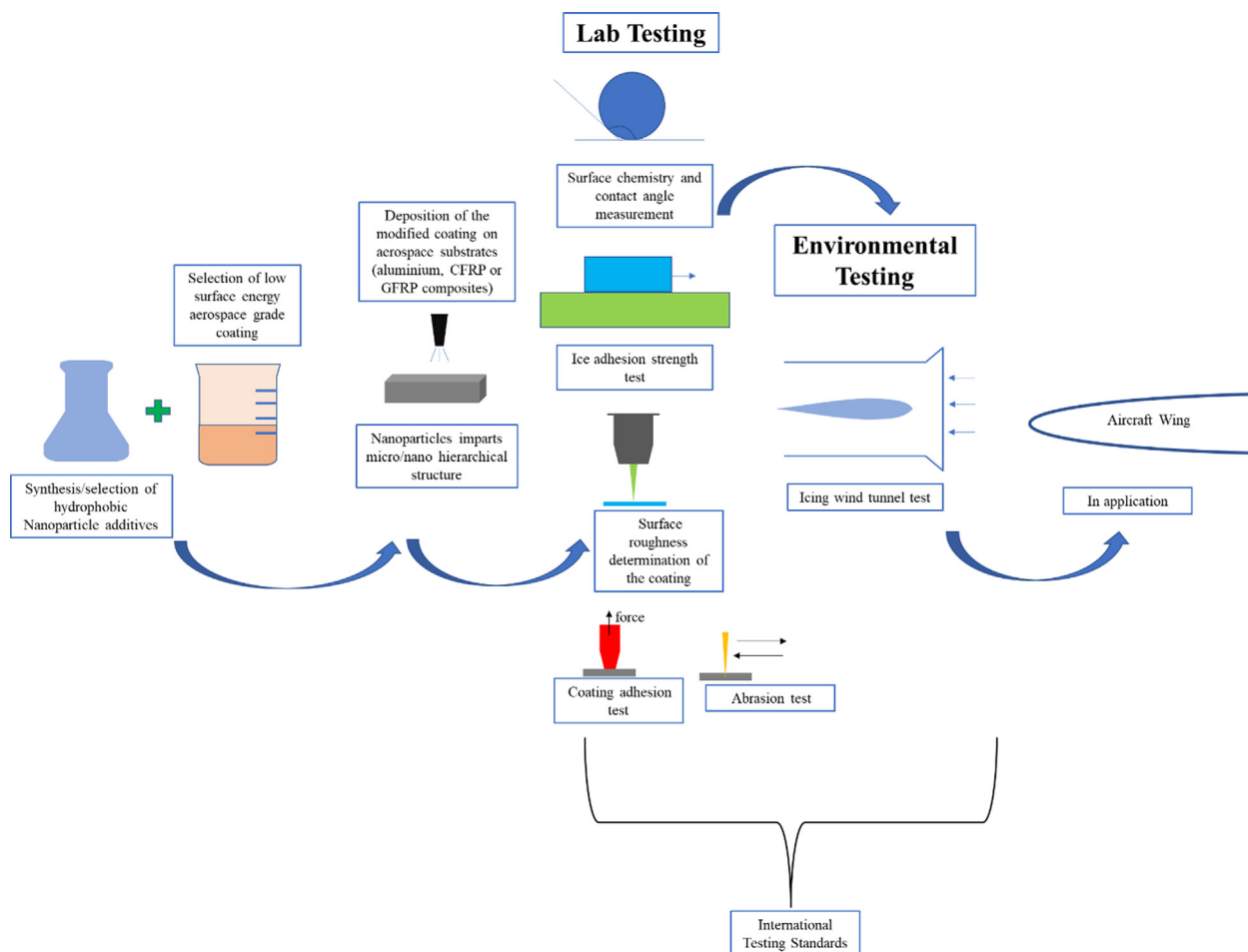

Fig. 7. Development of superhydrophobic coating for aerospace and energy sector.

face energy polymer matrix may be the answer. Particles may offer the necessary mechanical and physical qualities, as well as a means of imparting texture to the surface. Researchers have studied low surface energy epoxy coatings modified with nanosilica particles to achieve superhydrophobic characteristics [51,52]. Nanoparticles were incorporated to create micro/nano hierarchical structure and to make the coating mechanically durable. Despite several ongoing research investigations, no nanosilica modified ice-repellent coating can provide durability. To ensure NLF over the aeroplane wing superhydrophobic anti-icing coatings should be applied on wings which are typically manufactured using aluminium. However, glass fibre reinforced polymer (GFRP) composites, carbon fibre reinforced polymer (CFRP) composites and metal-composite laminates are increasingly being used in the construction of aeroplane bodies by commercial airlines as they are stronger than steel, lighter than aluminium, and can be moulded into almost any form. The adhesion of the coating to various aerospace substrates should be considered during the coating development process, as the coating's adhesion to the surface is just as critical as its icephobic properties. There is also an urgent need for worldwide standards to standardise the sample size, coating thickness, coating and ice adhesion testing, and icing test circumstances before completing comprehensive icephobic coating development and certification tests. There are numerous icing variables that must be specified, including substrate and coating temperature and inclination, the size, velocity, and temperature of supercooled water droplets, wind speed (turbulent or laminar), wind tunnel humidity, and chamber temperature. All coatings may be ranked in relation to one another, which is helpful for aligning the development of new coatings for aerospace applications. In terms of practicality, icephobic coatings must withstand erosion, abrasion, and UV radiation while retaining structural and chemical integrity. From a commercial standpoint, the proposed surfaces must be cost-effective, environmentally friendly, and simple to scale-up production. The authors' perspective for the development of an superhydrophobic anti-icing coating for aerospace application is summarised in Fig. 7.

5. Concluding remarks

In this review, the current state of anti-icing and icephobic studies are discussed. The first step in the development of passive anti-icing (icephobic) surfaces is to understand the kinetics of ice nucleation and heat transfer during the ice formation and accretion on solid surfaces. This review also presents an overview of the five icephobic aspects of a superhydrophobic surface namely (i) the ability of a surface to rebound or roll-off supercooled water droplets, (ii) decrease ice adhesion strength, (iii) delaying heterogeneous ice nucleation, (iv) delaying freezing time and (v) Impeding frost formation. Ice formation may be controlled to a significant degree by effectively employing superhydrophobic surfaces with hierarchical structure and low surface energy chemical composition by reducing ice nucleation and thermal conductivity. Research in this area still needs to answer many questions for example, what influence does topography play and if it does, whether it is the peak-to-valley measure or the average surface roughness which provides a better indicator to quantify this aspect. Likewise what type of fabrication methods and manufacturing approaches are more suitable. If one would like to use these concepts at a commercial scale, we need to have the ability to deposit coatings on surfaces of aluminum and CFRP panels which are more pronounced in aviation and energy sector so the issue of adhesion of such coatings with these materials would also need addressal. We hope that this review will inspire the young scholars to take these thoughts forward and will allow development of icephobic coatings addressing the above issues in near time.

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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