

# Theoretical Studies and Quantification of Ice Adhesion Mechanisms

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**Abstract** - Several countries subjected to ice storms can face potential disastrous economic and social issues when excessive ice accumulation triggers failures on aluminum ground wires and phase conductors of overhead power lines. Power supply companies and researchers are looking to develop new strategies to fight ice adhesion by developing icephobic coatings with homogeneous or heterogeneous chemical composition and/or specific surface roughness leading to superhydrophobicity. To engineer such coatings, it is crucial to comprehend the different forces acting at the ice-solid interface. In this paper, theoretical evaluation of these forces and the correlation between them as well as the influence of solid surface roughness on ice adhesion will be discussed.

Keywords: Ice adhesion, superhydrophobicity, icephobicity, interfacial interaction, ice microstructure, surface roughness, coatings.

## I. Introduction

Ice or wet snow accumulating on power network equipment can lead to serious problems in cold climate regions, as was the case with the 1998 and 2007 ice storms in Eastern Canada and Oklahoma [1, 2]. The costs associated with such events can be substantial as they can interrupt economic activity, affect public safety, and require active de-icing operations using chemical, mechanical and/or thermal techniques.

Industries and researchers around the world are realizing the need for developing alternative new strategies like the prevention of ice accumulation by covering exposed surfaces with icephobic coatings (passive methods). To produce such coatings, it is crucial to fully understand the ice-solid interactions in different conditions, such as temperature or the type of accumulation for instance. While many authors have concentrated on one particular aspect of the ice-solid interface, none have attempted to bring all the factors together to

develop a realistic picture of all the interactions. Therefore, the aim of this paper is to study the forces acting at the ice-solid interface, as well as the influences of both surface chemical composition and morphology. By optimizing these last two parameters, researchers will be able to develop efficient icephobic surfaces.

## II. Ice-solid interactions

If one puts aside ice accumulation parameters such as temperature and water droplet speed, ice adhesion strength is dependent on surface chemical composition (hydrophilic or hydrophobic materials) and surface morphology.

As far as chemical composition is concerned, several authors have studied the relationship between the surface tension of liquid water (contact angle of water drops) with ice shear strength, see Figs. 1 and 2 [3, 4]. In both figures, an increase of surface hydrophobicity ( $CA < 120^\circ$ ) leads to a reduction in ice shear strength. These results are logical since ice is composed of water molecules. At the ice-metal interface, broad ice crystals are formed within a few hours and are accompanied by smaller polygonal grains produced by recrystallization. This process, occurring in any polycrystalline material subjected to steady icing conditions, generally leads to ice dislocations and cracks [5]. The latter occurs due to the difference in thermal expansion coefficients of the ice and to substrate contraction. In addition, since ice volume changes during its solidification, one can expect that the interface is still subjected to a constraint. Crutch and Hartley also claimed that water molecules move through hydrogen bonding at a solid surface and, depending on the structure of the substrate surface, supercooled layers of water can occur at low temperatures [5]. They added that the orientation of water molecules in the first layer is less important for hydrophobic surfaces than for

hydrophilic ones. On hydrophobic surfaces, these dislocations are concentrated near the interface, where they generate interfacial defects and cracks. Interfacial forces at the ice-hydrophilic interface are strong and suppress the formation of ice, which results in a low concentration of defects occurring in the interfacial ice cracks of constraint blocks [5].

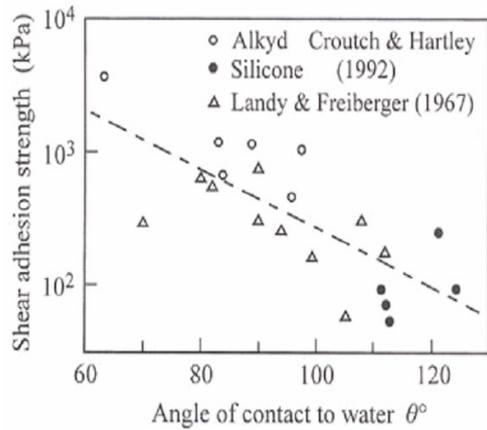


Fig. 1: Ice shear adhesion strength as a function of contact angle for different materials [3].

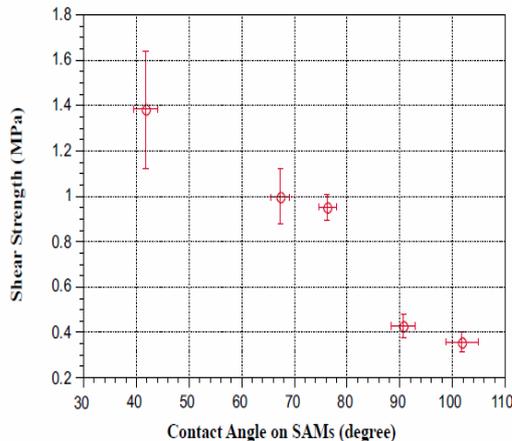


Fig. 2: Ice shear adhesion strength as a function of water contact angle on self-assembled monolayers [4].

To better understand the behaviour of the water molecules at the ice-solid interface, the theories of Petrenko [6] and Wilen [7], as well as intermolecular modelling, were used to define the role of electrostatic forces, van der Waals forces and hydrogen bonding. A self-consistent field-molecular orbital (SCF-MO) method was also used to determine the type and value of the interactive energy, and the distance between the molecules on either side of an interface [8]. In Table 1, hydrogen bonding interaction energies values were very different when comparing a water molecule interacting with a  $-\text{CH}_n$  type of group (see columns 1 and 2) or with a  $-\text{CF}_n$  type of group (see column

3). In fact, these simulations were carried out to prove that a heterogeneous chemical surface composition (e.g  $-\text{CH}_n$  and  $-\text{CF}_n$  groups) led to ice dislocations and very low ice shear strengths.

Table 1: Interaction energies between hydrophobic polymers and water [8].

Interaction Forms	LHC	DMS	FHC
Bond length (nm) (OH, FH)	0.252	0.329	0.187
$E_1^1$	-14.95	-15.64	-50.89
$E_1^2$	-5.70	-12.30	-48.51
$E_2^2$	-4.07	-4.64	-0.40
$E_3^3$	-0.81	+1.75	+39.89
$E_4^4$	-1.40	+1.79	+35.81

$E_1^1: \text{O}_{\text{H}_2\text{O}}^{\text{H}} \cdots \text{H}_{\text{HC}} \cdots \text{DMS} \cdots \text{F}_{\text{HC}} \cdots \text{H}_{\text{H}_2\text{O}}$  LHC: Hydrocarbon: ethane  $\text{C}_2\text{H}_6$   
 $E_2^2: \text{O}_{\text{H}_2\text{O}}^{\text{H}} \cdots \text{H}_{\text{HC}} \cdots \text{DMS}$  DMS: Dimethylsiloxane:  $\text{CH}_3\text{-(CH}_2\text{)}_2\text{-Si(CH}_3\text{)}_2\text{-Si(CH}_3\text{)}_3$   
 $E_3^3, E_4^4: \text{H}_{\text{H}_2\text{O}}^{\text{H}} \cdots \text{H}_{\text{HC}} \cdots \text{DMS, FHC}$  FHC: Hexafluoroethane:  $\text{C}_2\text{F}_6$

As far as surface morphology is concerned, some authors have very recently examined the behaviour of superhydrophobic surfaces ( $\text{CA} > 150^\circ$ ) [9-16]. These studies were performed at the CIGELE laboratory of the University of Quebec at Chicoutimi and they showed that a certain surface roughness may help to reach icephobicity. In this case, and contrary to mechanical anchoring, water does not penetrate the superhydrophobic 3D structure of the coatings and therefore freezes upon air and a small fraction of the coating, leading to very low ice adhesion shear strengths.

This short literature review reveals that there is a shortage of studies aimed at determining ice adhesion mechanisms and which type of surface can be considered icephobic. So far, all the factors implied in the ice adhesion were never taken into account in the works. It is therefore important to study the influence of all the forces present at the ice-solid interface, as well as the influence of roughness and the type of precipitation.

### III. Forces and parameters influencing ice adhesion

Adhesion is a physicochemical process in which two surfaces are bound by interfacial interactions [17, 18]. Three different types of adhesion forces are present (see Fig. 3): Firstly, "chemical adhesion" involves covalent, electrostatic and/or metallic atomic bindings, which are short-range interactions. Secondly, "thermodynamic adhesion" corresponds to molecular interaction forces across the interface involving van der Waals and hydrogen bonding type of forces (long-range interactions). Thirdly, "mechanical adhesion" results from the mechanical

interlocking of two materials due to microscopic surface asperities [19].

The total force of adhesion in the case of an ice-solid interface is composed of four entities: electrostatic interactions, van der Waals forces, hydrogen bonding type of force, and mechanical adhesion due to solid surface roughness [6]. Other phenomena also should be taken into account, such as the presence of a quasi-like layer (QLL) at the ice surface that can influence the adhesion strength. All these aspects will be developed in the next sub-sections.

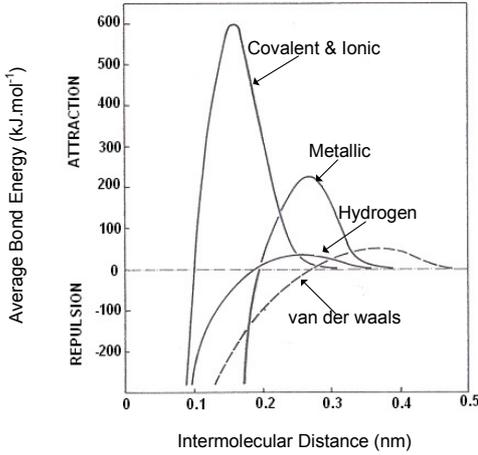


Fig. 3: Strength and range of the typical intermolecular and interatomic forces [18].

#### A. Electrostatic forces

Electrostatic interaction occurs between two different substrates under the influence of their electronic band structures leading to the transfer of electrons between the atoms and/or the molecules at the interface. This concept of attraction is based on Coulomb's law and the acceptor-donor interaction [3]. Petrenko and Ryzhkin claim that the electrostatic interaction is always present at the ice-metal or ice-dielectric interfaces and that it plays a significant role in the adherence of the ice [6]. In fact, electrostatic attractions result from the difference of permittivity between the two materials, by which a double electrical layer is created. The works of Petrenko and Ryzhkin are based on the Jaccard theory, which stated the existence of electric charge at the ice surface [20]. These authors supposed that the surface charge comes from the capture of protonic charge or defects at the ice surface. Thus (see Fig. 4), the passage of a proton from one water molecule to another allows for the creation of a pair of ionic defects ( $\text{OH}^-$  and  $\text{H}_3\text{O}^+$ ). Additionally, a water molecule, rotated at a  $2\pi/3$  angle around one of the O-O bonds, leads to the creation of a rotational Bjerrum type **L** defect (Leer, suppression of a

hydrogen bond between two oxygen) or **D** (Doppelt, connection occupied by two protons) [3, 6]. For example, at the ice-metal interface the charge distribution on the ice side induces another charge on the metal surface, giving rise to an electric field between the two solids. Depending on the type of charge carriers, the adhesion energy has been found to vary between 0.08 and  $1.3 \text{ J.m}^{-2}$  [6]. The mechanism of the electrostatic bonding remains significant up to distances equal to  $10 r_{\text{OO}}$ , where  $r_{\text{OO}} = 0.276 \text{ nm}$  is the distance between two oxygen atoms.

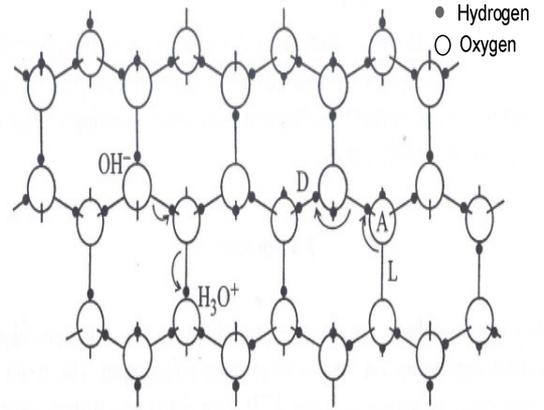


Fig. 4: Ionic defects and Bjerrum on the ice structure [3].

Similarly, on the metal or dielectric surface side, a surface charge is also created, leading to the formation of an electrical double-layer structure. The corresponding force, or image force, is described by Equation (1), where the solid has an effective dielectric constant ( $\epsilon$ ) greater than that of the ice ( $\epsilon_{\text{ice}}$ ) [6]. This equation shows that the force  $F$  (or image force) is inversely proportional to the square of the distance between the interfacial charges  $r$ , where  $q$  is the charge and  $\epsilon_0$  the absolute permittivity in vacuum.

$$F = \frac{q^2}{16\pi\epsilon_0\epsilon r^2} \frac{\epsilon_{\text{ice}} - \epsilon}{\epsilon_{\text{ice}} + \epsilon} \quad (1)$$

A charge  $q$  on the ice surface induces another charge known as "image charge" on the metal surface. However, the same charge  $q$  induces a weaker image charge  $q^*$  on the dielectric surface, as evidenced by Equation (2), where  $q^*$  is the charge image on the dielectric solid surface,  $q$  is the charge on the surface of the ice and  $\epsilon$  is the permittivity of the dielectric surface [6].

$$q^* = q \left( \frac{\epsilon - 1}{\epsilon + 1} \right) \quad (2)$$

In most dielectrics,  $\epsilon$  is higher than one and the induced charges are comparable with those induced

in metals. It is also obvious from Equation (2) that the lower is  $\epsilon$ , the lower will be the electrostatic-related adhesion. For example, Teflon<sup>®</sup> has a very low dielectric constant ( $\sim 2.1$ ) resulting in weaker image charges and therefore leading to potential icephobic properties [6].

### B. Lifshitz – van der Waals forces

Van der Waals forces, also called secondary gravitational attractive or repulsive, are always present [18, 19]. These interactions are common interfacial forces and are regarded as universal, resulting from dipole-dipole interaction. Wilen *et al.* [7] calculated the Lifshitz van der Waals (LW) interaction between ice and a number of materials, including several metals. They concluded that LW interactions were not the main contributors to the interfacial energy, and that electrostatic interaction may play a more important role in ice adhesion. For their part, Van Oss *et al.* [21] measured the contact angles of a flat polycrystalline ice surface with a number of liquids. They concluded that the contribution of the LW non-polar component to the ice surface tension was smaller ( $26.9 \text{ mJ.m}^{-2}$ ) than that of the polar Lewis acid-base component ( $39.6 \text{ mJ.m}^{-2}$ ).

### C. Hydrogen bonding

Hydrogen bonding, which can be regarded as an electrostatic interaction, results from the distribution of a proton (hydrogen atom) between two electronegative atoms such as oxygen, nitrogen or fluorine. In fact, these attractions are responsible for the cohesion of solid ice and are also present in liquid water. To the best of our knowledge, little has been done to determine the true role of hydrogen bonding in the adherence of ice to a given surface. This type of bonding should play a major role not only due to the presence of water molecules, but also that of oxides on the solid side,  $-\text{CH}_n$  or  $-\text{CF}_n$  ( $n = 1$  to  $3$ ) type of chemical composition. Therefore, chemical bonding between ice and other solids is largely hydrogen bonding but has not been studied in detail yet [4].

### D. Mechanical bonding

Surfaces of industrial structures are actually never perfectly plane and may have a certain degree of roughness and even porosity. Asperities and/or pores can influence the mechanical bonding strength at the ice-solid interface since liquid water may penetrate this 3D structure resulting, after solidification, in mechanical anchoring [22].

Mechanical bonds occur on scales larger than atomic or molecular, often in the micrometer range, and are therefore observable with optical microscopes [22]. The thermal coefficient of water around  $0^\circ \text{ C}$  is larger than those of metals and oxides. As well, water dilates during freezing,

while metals and oxides tend to contract. During the freezing process, interfacial bonds are formed between the ice and the substrate, thus leading to the re-arrangement of  $\text{H}_2\text{O}$  molecules, resulting in a phase conversion by the generation of additional hydrogen bonds. Consequently, small changes in the positions of protons in motion and in the reorientation of hydrogen bonds with a higher stability are produced [17]. Alternatively, additional hydrogen bonds between molecules are formed. Ice volume expansion is hence expressed based on the standard formation of hydrogen bonding [23].

### E. Quasi Liquid Layer (QLL)

In our everyday lives, ice is frequently present at temperatures very close to its melting point. Consequently, a quasi liquid layer is present at its surface as well as at the ice/solid interface, and constitutes an influential parameter regarding ice adhesion strength. Studies have been carried out to understand the behaviour of ice surface and its interaction with solid surfaces, accounting for the existence of a quasi liquid layer relative to temperature. Jellinek estimated that the thickness of the QLL can extend from  $100 \text{ \AA}$  to  $1000 \text{ \AA}$  at  $-4.5^\circ \text{ C}$ . This corresponds to a 30 to 300 water molecules depth layer [24]. Dash's review quoted that many laboratory studies have explored the interfacial fusion of ice with various substrates such as quartz, metals, graphite, glass, silica, polystyrene and PVC [25]. A variety of techniques have been used for that purpose, namely, atomic force microscopy, mercury porosimetry, quartz micro-balance and X-ray scattering. Many authors noted that interfacial fusion starts at temperatures much lower than that of surface fusion. For example, in the case of graphite, interfacial fusion is detected as low as  $-30^\circ \text{ C}$ .

The role of the QLL is a factor to take into account in this study, since it wets the surface of the solid and therefore increases the contact surface area between the ice and the solid, which affects ice adherence.

## IV. Experimental and theoretical studies

The experimental and theoretical studies of the different factors influencing ice adhesion strength are described in this section. Ice adhesion force will be measured using centrifuge adhesion tests (CAT) [26]. Since surface roughness can change the ice shear strength, control and knowledge of this parameter is crucial for these calculations and measurements.

### A. Solid permittivity

As described above, electrostatic force plays a significant role in the ice adhesion process. This force depends on the dielectric permittivity of the solid and that of the ice. Permittivity can be calculated from the value of the complex

impedance  $Z = 1/Y$ , measured in a frequency band of 40 Hz to 110 MHz, using an impedance analyzer. The value of capacity (in F) can be obtained from Equation (3), where  $\text{Im}(Y)$  is the imaginary part of admittance  $Y$  and  $f$ , the frequency (Hz).

$$C = \text{Im}(Y)/2\pi f \quad (3)$$

The dielectric constant  $\epsilon$  (permittivity) of the metal or film is directly calculated from the parallel plate capacitor formula, see Equation (4), where  $A$  is the electrode area ( $\text{m}^2$ ) and  $d$  the thickness of film (m).

$$C = \epsilon\epsilon_0 A/d \quad (4)$$

#### B. Surface roughness

The influence of surface roughness on ice adhesion is a major point in this research since the adhesive bond between the surface of the solid could be very high (mechanical interlocking) or very low (presence of air pockets). In order to study this phenomenon, samples with different roughness will be analyzed. Techniques like plasma etching, chemical etching, or anodizing could be used to obtain different degrees of roughness. The RMS roughness can be measured by atomic force microscopy (AFM), or using profilometry techniques such as laser ones. Where force measurements are concerned, a refrigerated wind tunnel at controlled temperatures and wind velocities is used to accumulate ice on a sample fixed at one end of an aluminium beam. Thereafter, the beam is allowed to spin at increasing rotational speeds and the resulting centrifugal force is used to detach ice. The shear stress and the adhesion reduction factors can be calculated from the centrifugal force, ice mass, beam radius, rotation speed and surface area. The adhesion strength is assumed to be equal to the centrifugal force (in N), see Equation (5), where  $m$  is the ice mass (kg),  $r$ , the beam radius (m) and  $\omega$ , the rotation speed ( $\text{rad.s}^{-1}$ ).

$$F = mr\omega^2 \quad (5)$$

#### C. Study of the QLL

The quasi liquid layer must also be taken into consideration. Therefore, conductivity measurements will be performed to predict the presence of water in its liquid or solid state. This technique is based on the impedance measurement of a medium disturbed by a sinusoidal signal. The set-up comprises two interdigital transducers (IDT), made by photolithography on a piezoelectric substrate. Sinusoidal tension is applied to the boundaries of a transmitting transducer and is converted into an acoustic wave surface (SAW). A

receiving transducer, IDT, reconverts the acoustic wave into electric voltage. Conductivity is deduced from these data. This method is regarded as a quick, non-destructive and precise.

The results will be compared to experimental observation of the ice granulometry in contact with the solid surface. Several parameters may influence the formation of the QLL and the granulometry: chemical composition and morphology of the solid as well as temperature, wind speed and water droplet size. Based on these parameters and on the ice and solid thermal coefficients, the freezing process will be experimentally studied using an optical microscope in a cold room, as well as a high speed video camera to study ice grain size and form, the possible presence of air pockets and the impact of supercooled water droplets.

#### D. Molecular modelling

Molecular modelling has become an essential tool to determine molecular structures. In the current work, determination of interaction types and energies as well as bonding lengths between sample surface and the first water molecule layer, are regarded as essential data to understand the molecular structure at the ice-solid interface. Theoretical results obtained by modelling will be analyzed and compared with experimental results such as measurements of ice adhesion strengths.

For instance, some researchers used Grand Canonical Monte Carlo (GCMC) to simulate molecular structures of carbonaceous materials and silica [27]. This method has allowed for determining isothermal adsorption of these molecules on ice surfaces, by keeping the temperature constant and controlling the chemical potential of the adsorbed molecules. This method was employed successfully to simulate the adsorption of methanol, formaldehyde and acetone molecules at 200 K [28]. The orientation of the formaldehyde molecules and acetone on the ice surface was computed and the formation of the hydrogen bonding between formaldehyde and ice (water molecule) was used.

#### V. Conclusions

Ice adhesion on a solid surface involves many physical and chemical phenomena. While some very interesting studies can be found in the literature, these only focus on one specific aspect. Therefore, it is very difficult to predict which factor will have the most significant impact on the ice adhesion force. To improve understanding of the various physical mechanisms responsible for ice adhesion on a surface, a general theoretical and experimental study is crucial. The aim of this research is therefore to quantify the mechanisms of ice adhesion. From an industrial point of view, future icephobic coatings will probably be

superhydrophobic or chemically heterogeneous. Since these coatings will be exposed to variable atmospheric conditions and precipitations (temperature, wind, snow, rain and frost), it is very important to integrate all these parameters to find out which icephobic materials can resist extreme ice accumulations.

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