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Study on the Preparation and Performance of Anti-Ice Hydrogels for Low-Temperature Environments

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Abstract

Icing presents significant risks to transportation, infrastructure, and the safety of equipment, while surfaces designed to prevent ice formation, such as superhydrophobic coatings, often fail in low-temperature environments. In this study, three-dimensional crosslinked κ-carrageenan hydrogels were separately impregnated with ethylene glycol, propylene glycol, and glycerol, resulting in the formation of ethylene glycol–impregnated κ-carrageenan hydrogel (EGIκCH), propylene glycol–impregnated κ-carrageenan hydrogel (PGIκCH), and glycerol-impregnated κ-carrageenan hydrogel (GLIκCH). Among these, the EGIκCH surface exhibited outstanding anti-icing performance. The freezing delay time of water droplets on the surface of each hydrogel were measured using a contact angle goniometer. The results revealed that all three hydrogels demonstrated significantly longer freezing delay time than glass and aluminum surfaces. At -30°C, the freezing delay time of the EGIκCH surface reached up to 1269 s, markedly exceeding that of the other two types. This study provides a new idea for the preparation of anti-icing materials for low-temperature applications.

Keywords

Hydrogel, Anti-icing, Low-temperature.

1. Introduction

Ice is a solid crystalline phase of water that forms when the temperature drops to or below its freezing point (0℃ at standard atmospheric pressure), as water transitions from the liquid state. This process originates from the ordered arrangement of water molecules. In subzero environments, the surfaces of numerous objects, such as roads, water pipelines, ships, and wind turbine blades, are highly susceptible to icing [1][3]. Once ice forms on these surfaces, it can lead to severe hazards [4]-[6]. It is well established that road icing causes a substantial reduction in the friction between tires and the road surface, resulting in tire skidding and a cascade of loss-of-control risks, thereby posing a serious threat to vehicular safety. Likewise, icing on airport runways can cause flight delays or cancellations. Furthermore, because water undergoes an approximate 9% volumetric expansion upon freezing, the freezing of water within supply pipelines in low-temperature environments may result in pipeline cracking or rupture. As a result, icing poses significant threats to transportation systems, infrastructure, agricultural production, and everyday life, underscoring the critical importance of adopting scientific and effective measures to address icing-related challenges [7]-[9].

To address icing issues encountered by various equipment and facilities in low-temperature environments, a wide range of surface-engineering-based anti-icing strategies have been developed. Among these, the design of specialized surface coatings to inhibit or delay ice crystal formation and adhesion has become an important research direction. Such coatings mainly include interfacial slippage surfaces [10],[11], lubricating surfaces [12]-[15], and superhydrophobic coatings [16]-[20]. These approaches rely on distinct physical or chemical

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mechanisms, including reducing the solid–liquid contact area, forming a lubricating liquid layer, and increasing the water contact angle, to delay ice formation and reduce ice adhesion strength, thereby alleviating efficiency losses and safety risks associated with icing to a certain extent. However, despite their promising anti-icing potential, these surface technologies still face significant challenges related to long-term durability and environmental adaptability in practical applications. For example, superhydrophobic surfaces depend on high water contact angles and low rolling angles to enable water droplets to roll off prior to freezing, thus delaying ice nucleation [21]. In low-temperature and high-humidity environments or prolonged condensation, however, surface microstructures are prone to being filled with condensed water, leading to the failure of hydrophobicity and a limited extension of icing delay.

In this study, we propose a simple and cost-effective surface anti-icing strategy based on organic hydrogels. The hydrogels consists of a three-dimensional crosslinked polymer network impregnated with organic alcohols possessing anti-icing properties, enabling the material to maintain flexibility in low-temperature environments. while suppressing ice formation. Experimental results demonstrate that surfaces constructed from this organic hydrogel can effectively delay ice formation. While preserving fabrication simplicity and economic feasibility, this strategy provides a new perspective for the preparation of anti-icing materials.

2. Materials and Methods

2.1. Materials

κ-Carrageenan, deionized water (ACS, for trace analysis), ethylene glycol (AR, ≥98%), propylene glycol (AR, ≥99%), glycerol (AR, ≥99%), glacial acetic acid (reagent grade, ≥99.5%), and glutaraldehyde solution (AR, 50% aqueous solution) were obtained from Shanghai Aladdin Technology Co., Ltd. All chemical reagents were used without further purification.

2.2. Dilution of Glutaraldehyde Solution

1 mL of glutaraldehyde solution (50% aqueous solution) was measured and placed into a 50 mL beaker, to which 49 mL of deionized water was added. After thorough mixing, the final 50 mL of glutaraldehyde solution (1% aqueous solution) was obtained.

2.3. Preparation of Hydrogels

1 g of κ-carrageenan was added to 50 mL of water and stirred continuously at 80°C for 30 minutes to ensure complete dissolution. Then, 2 mL of glutaraldehyde solution (1% aqueous solution) and 1 mL of glacial acetic acid were added to the solution, mixed thoroughly, and poured into molds to cure and form the κ-carrageenan hydrogel. The hydrogels were subsequently placed in Petri dishes containing ethylene glycol, propylene glycol, and glycerol, respectively, for 12 hours. Afterward, the hydrogels were removed and classified into ethylene glycol-impregnated, propylene glycol-impregnated, and glycerol-impregnated κ-carrageenan hydrogels.

2.4. Measurement of Freezing Delay Time of Water Droplets

The freezing delay times of water droplets on the surfaces of the three hydrogels, glass, and aluminum were measured using a contact angle goniometer. During the experiment, the cooling stage temperature was set to -5°C, -10°C, -15°C, -20°C, -25°C, and -30℃. Once the temperature stabilized, the organic hydrogel samples were placed on the cooling stage to reach thermal equilibrium. Then, 8 μL water droplets were placed on the sample surface, and the timing began simultaneously. The appearance of the droplet was continuously captured at a rate of once per second until sharp ice crystals formed at the droplet's top due to the expansion of water during freezing. The freezing delay time was defined as the time interval from the start of the timer to

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the stop, representing the time required for the water droplet to freeze under the given surface conditions.

3. Results and Discussion

Under various temperature conditions, we measured the freezing delay time of water droplets on the surfaces of EGIκCH, PGIκCH, GLIκCH, glass, and aluminum. Our observations revealed that the freezing process of the droplets initiated at the three-phase contact line where the droplet met the low-temperature substrate, subsequently expanding along the liquid-solid interface toward the interior of the droplet. Notably, a significant expansion of the droplet's volume occurred during freezing, and upon completion of the phase transition, sharp ice crystal protrusions were formed at the droplet's apex. Figure 1 presents a detailed quantitative comparison, highlighting the significant impact that different surfaces have on the freezing delay time of water droplets.

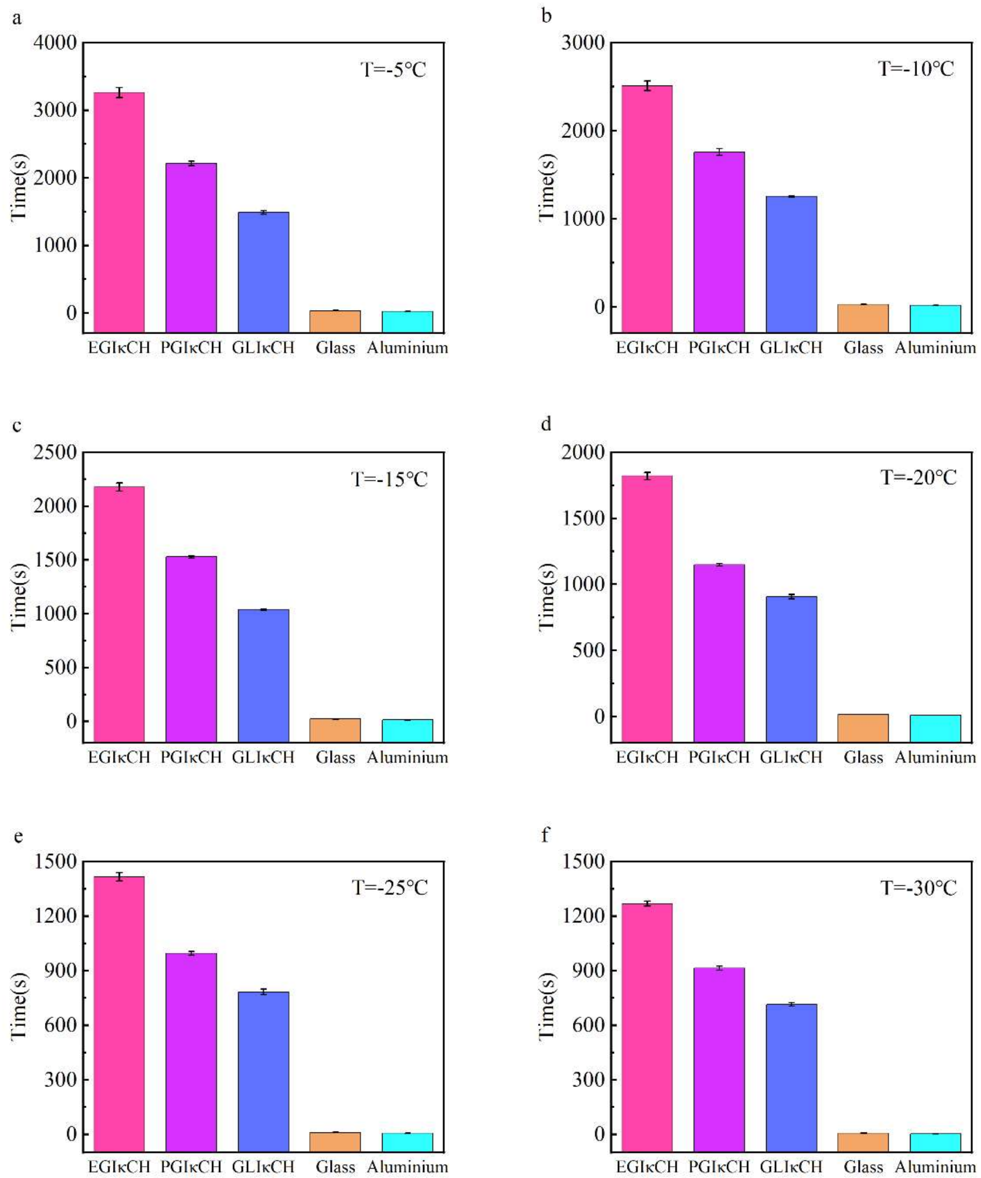

Figure 1. The freezing delay time of water droplets on different surfaces at various temperatures

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As seen in Figure 1a, at -5°C, the freezing delay time of water droplets on the surfaces of EGIκCH, PGIκCH, and GLIκCH was notably longer than that on glass and aluminum. Specifically, the freezing delay time on the surfaces of EGIκCH, PGIκCH, and GLIκCH reached 3261 s, 2214 s, and 1488 s, respectively, far exceeding the freezing delay time on glass and aluminum, which reached only 34 s and 22 s, respectively. In a direct surface comparison, the freezing delay time on the EGIκCH surface was approximately 96 times that of glass, on the PGIκCH surface it was about 65 times that of glass, and on the GLIκCH surface it was about 44 times that of glass. Compared to aluminum, the freezing delay time on the EGIκCH surface was approximately 148 times that of aluminum, on the PGIκCH surface it was about 101 times, and on the GLIκCH surface it was about 68 times that of aluminum.

Figure 1b shows that at -10°C, the freezing delay time on EGIκCH, PGIκCH, and GLIκCH surfaces reached 2510 s, 1756 s, and 1256 s, respectively, while the freezing delay time on glass and aluminum reached only 27 s and 17 s, respectively. In comparison with glass and aluminum, the freezing delay time on the EGIκCH, PGIκCH, and GLIκCH surfaces was significantly longer at -10°C. Specifically, the freezing delay time on the EGIκCH surface was about 93 times that of glass and about 148 times that of aluminum, the PGIκCH surface was about 65 times that of glass and about 103 times that of aluminum, and the GLIκCH surface was about 47 times that of glass and about 74 times that of aluminum.

As shown in Figure 1c, at -15°C, the freezing delay time on the surfaces of EGIκCH, PGIκCH, and GLIκCH reached 2178 s, 1528 s, and 1039 s, respectively, compared to 23 s on glass and 14 s on aluminum. In comparison, the freezing delay times on EGIκCH, PGIκCH, and GLIκCH surfaces were significantly longer at -15°C. Specifically, the freezing delay time on the EGIκCH surface was approximately 95 times that of glass and about 156 times that of aluminum. The PGIκCH surface was 66 times that of glass and 109 times that of aluminum, while the GLIκCH surface was 45 times that of glass and 74 times that of aluminum.

Figure 1d reveals that at -20°C, the freezing delay time of water droplets on the EGIκCH, PGIκCH, and GLIκCH surfaces reached 1821 s, 1149 s, and 907 s, respectively, while the freezing delay time on glass and aluminum reached 16 s and 9 s, respectively. Specifically, the freezing delay time on the EGIκCH surface was about 114 times that of glass and about 202 times that of aluminum, the PGIκCH surface was about 72 times that of glass and about 128 times that of aluminum, and the GLIκCH surface was about 57 times that of glass and about 101 times that of aluminum.

From Figure 1e, we observe that at -25°C, the freezing delay time on the surfaces of EGIκCH, PGIκCH, and GLIκCH reached 1416 s, 996 s, and 783 s, respectively, compared to 10 s on glass and 6 s on aluminum. The freezing delay time on the EGIκCH surface was about 142 times that of glass and 236 times that of aluminum. For the PGIκCH surface, the freezing delay time was about 100 times that of glass and 166 times that of aluminum, and for the GLIκCH surface, it was 78 times that of glass and 131 times that of aluminum.

Finally, Figure 1f reveals that at -30°C, the freezing delay time on the EGIκCH, PGIκCH, and GLIκCH surfaces reached 1269 s, 914 s, and 715 s, respectively, while the freezing delay time on glass and aluminum reached just 7 s and 4 s, respectively. Compared with glass and aluminum surfaces, the EGIκCH, PGIκCH, and GLIκCH surfaces exhibited significantly longer freezing delay time, indicating superior anti-icing capabilities of these materials in low-temperature environments. The freezing delay time on the EGIκCH surface was approximately 181 times that of glass and about 317 times that of aluminum, the PGIκCH surface was 131 times that of glass and 229 times that of aluminum, and the GLIκCH surface was 102 times that of glass and 179 times that of aluminum.

The results indicate that compared with glass and aluminum, the EGIκCH, PGIκCH, and GLIκCH surfaces show significantly longer freezing delay time, effectively delaying the freezing of water droplets. This reflects their exceptional anti-freezing performance, which is attributed to the presence of more unfrozen bound water within these materials. Additionally, the organic alcohol components on the material surfaces exhibit greater fluidity, enhancing the interaction between the hydrophilic components and water molecules, which further improves their anti-icing capabilities. As a result, these materials demonstrate superior anti-icing performance in low-temperature environments, making them highly promising for anti-icing applications. Furthermore, data comparisons reveal that EGIκCH outperforms PGIκCH and GLIκCH in anti-icing performance. Specifically, EGIκCH effectively suppresses ice crystal formation, granting it exceptional anti-icing properties and significantly delaying the formation of ice layers. Additionally, EGIκCH shows better stability in low-temperature environments, making it more suitable for various harsh climates, giving it a significant edge in anti-icing applications. Although PGIκCH and GLIκCH are slightly less effective in terms of anti-icing performance, they still possess certain anti-icing capabilities under different conditions, with the choice of material depending on the specific needs and environmental factors. In conclusion, the use of these materials significantly enhances surface anti-icing performance, prolonging the freezing time of water droplets in low-temperature environments. EGIκCH, PGIκCH, and GLIκCH exhibit superior anti-icing capabilities in low-temperature environments, holding vast potential for practical applications.

4. Conclusion

In this study, we successfully developed hydrogels that combine transparency with excellent anti-icing performance. By incorporating ethylene glycol, propylene glycol, and glycerol into κ-carrageenan hydrogels, we significantly improved the anti-icing properties of the material in low-temperature environments, while maintaining good mechanical stability and optical transparency. Experimental results show that at -30°C, the freezing delay time on the surface of the ethylene glycol-impregnated κ-carrageenan hydrogel reached 1269 s, approximately 96 times that of ordinary glass, demonstrating exceptional anti-icing performance. This performance is primarily due to the introduction of ethylene glycol, which effectively lowers the freezing point of water and suppresses ice nucleation and growth by weakening hydrogen bonds between water molecules. Additionally, the network structure of the hydrogel further delays the phase transition by binding liquid water. Notably, this hydrogel maintains good transparency while achieving efficient anti-icing, providing a solid foundation for its practical applications in anti-icing fields. This study provides a new idea for the preparation of anti-icing materials.

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