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Fast Capture, Collection, and Targeted Transfer of Underwater Gas Bubbles Using Janus-Faced Carbon Cloth Prepared by a Novel and Simple Strategy

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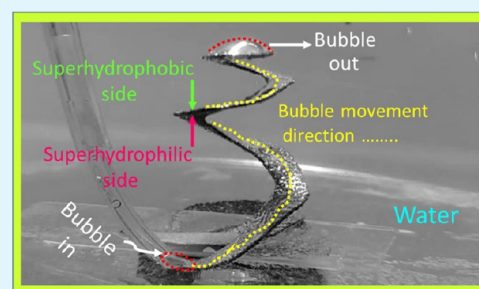


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Supporting Information

ABSTRACT: Transportation of bubbles in liquids in a controlled fashion is a challenging task and an important subject in numerous industrial processes, including elimination of corrosive gas bubbles in fluid transportation pipes, water electrolysis, reactions between gases, heat transfer, etc. Using superaerophilic surfaces represents a promising solution for bubble movement in a programmed way. Here, a novel and low-cost method is introduced for the preparation of Janus-faced carbon cloth (Janus-CC) using poly(dimethylsiloxane) (PDMS) coating and then burning one side of the carbon cloth/PDMS on an alcoholic burner. The results show that the superhydrophobic face behaves as a superaerophilic surface, while the superhydrophilic side is aerophobic underwater. Subsequently, the Janus-CC is applied for pumpless transport of underwater gas bubbles even under harsh conditions. The movement of gas bubbles on the surface of the Janus-CC is interpreted based on the formed gaseous film on the aerophilic side of the Janus-CC. Various applications of the prepared Janus-CC for underwater bubble transportation, such as underwater gas distributor, gas collector membrane, gas transport for chemical reactions, unidirectional gas membrane, and elimination of gas bubbles in transport pipe, are presented.



KEYWORDS: Janus-faced carbon cloth, bubble transfer, poly(dimethylsiloxane) (PDMS), superaerophilic, superhydrophobic, superhydrophilic, gas bubble capture

INTRODUCTION

Wettability at solid/liquid/gas interface, which is inspired by natural surfaces, has been widely studied and has many practical applications including anti-icing, oil–water separation, self-cleaning, fog harvesting, anticorrosion, microfluidics, etc.^{1–3}

Janus materials with opposite wettability (superhydrophobicity and superhydrophilicity) have attracted a lot of attention. One of the most famous natural examples of Janus surfaces is the lotus leaf; its upper side is superhydrophobic (water contact angle $> 150^\circ$) and oleophilic (oil contact angle $< 90^\circ$), while the lower side is hydrophilic (water contact angle $< 90^\circ$) and underwater oleophobic (oil contact angle $> 90^\circ$).⁴ Janus materials have a variety of practical applications, including fog harvesting,^{5–8} membrane distillation,^{9–11} humidity sensing,¹² directional liquid transfer,^{13–15} solar desalination,^{16–20} and oil–water separation,^{21–24} and one of the applications that has recently been considered by researchers is the collection and transportation of gas bubbles.²⁵ The presence of bubbles in liquid systems is significant due to their application in various cases like wastewater treatment,²⁶ minerals recovery,²⁷ and solar energy harvesting. On the other hand, gas bubbles can have negative effects on pipes under certain conditions. Indeed, the presence of gas bubbles leads to an increase in the rate of corrosion and hydrogenation of the

pipes. For example, hydrogen sulfide, carbon dioxide, and oxygen bubbles lead to corrosion of the pipes.^{28,29} In recent years, controlled transportation of bubbles has become a very important challenge. Therefore, the attention of a number of researchers was oriented toward making samples that can transport and collect bubbles in a controlled fashion using elastic liquid infused materials,³⁰ superaerophilic geometry-gradient channels,³¹ slippery liquid infused porous surfaces (SLIPs),³² Janus surfaces,³³ etc.

Underwater superaerophobic (bubble contact angle $> 150^\circ$) or aerophobic (bubble contact angle $> 90^\circ$) and superaerophilic (bubble contact angle $< 10^\circ$)^{1–3} surfaces have been applied to collect and transport subaqueous gas bubbles.^{34,35} Superhydrophobic surfaces are underwater superaerophilic and superoleophilic, while superhydrophilic surfaces are underwater superaerophobic and superoleophobic.^{36,37} Therefore, these surfaces possess bubble capture and antibubble proper-

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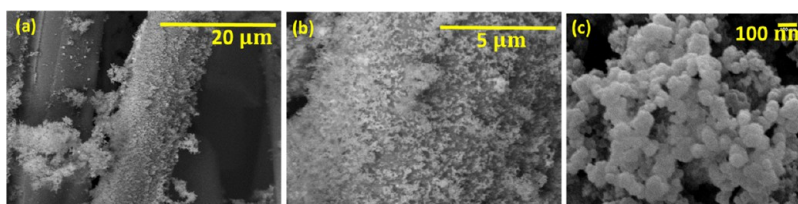


Figure 1. FE-SEM images at various magnifications of the flame-exposed side (superhydrophilic) of the Janus-CC.

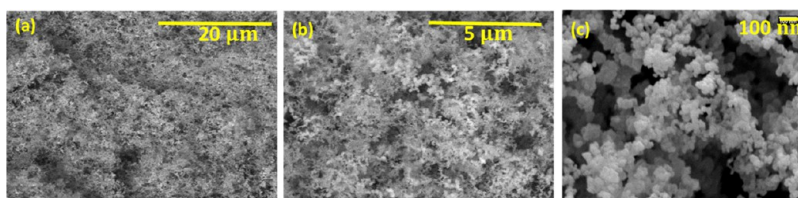


Figure 2. FE-SEM images at various magnifications of the side nonexposed to flame (superhydrophobic) of the Janus-CC.

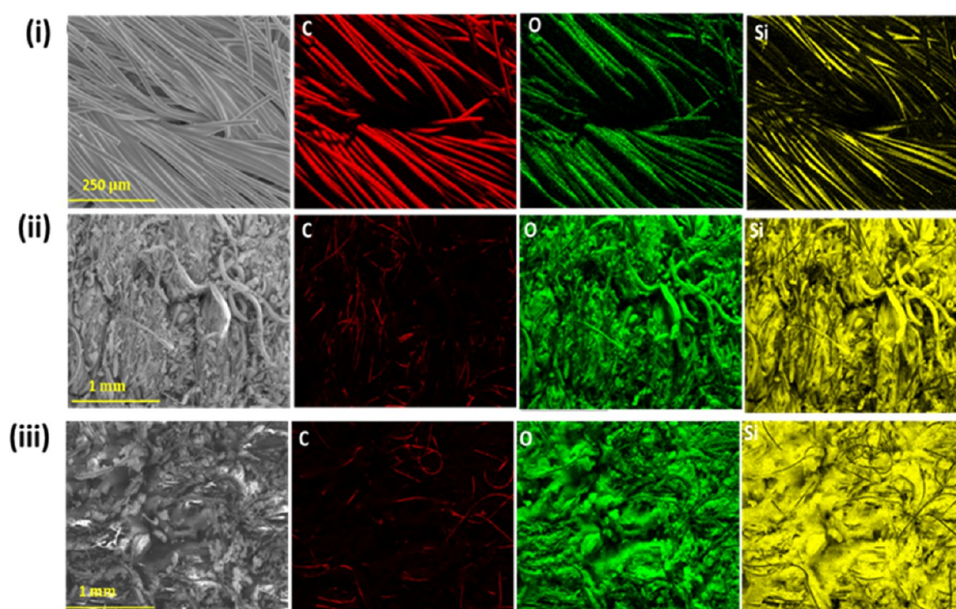


Figure 3. Element mapping images of (i) superhydrophobic carbon cloth, (ii) Janus-CC superhydrophilic side, and (iii) Janus-CC superhydrophobic side.

64 ties and could be applied to collect or repel bubbles,
65 respectively.

66 Hu et al. synthesized a Janus membrane using a femtosecond
67 laser-assisted ablation and nanoparticles deposition approach
68 to achieve underwater unidirectional bubble infiltration.³⁸
69 Wang et al. prepared a Janus sample by spraying polydopamine
70 on a polypropylene surface, and the synthesized sample was
71 used for dye adsorption and bubble aeration.³⁹ Pei et al.
72 designed a Janus mesh with the ability to unidirectionally
73 transfer gas bubbles. The bubbles passed through the mesh
74 from the superaerophobic side to the superaerophilic side
75 underwater, but transportation in the opposite direction was
76 not possible.⁴⁰ Chen and co-workers produced arrays of
77 polydimethylsiloxane with the ability to control the bubble
78 wettability gradient for underwater bubble unidirectional
79 spontaneous transport.⁴¹

80 In recent years, the research activity in this field has been
81 focused on the design of cost-effective, environmentally
82 friendly, and low-energy methods for controlled transmission
83 and removal of gas bubbles. Herein, a novel strategy was

demonstrated for the preparation of Janus carbon cloth (Janus-
84 CC) using PDMS coating and then burning one side of the
85 sample on an alcohol burner. The prepared sample was
86 characterized by different methods for its morphology,
87 chemical composition, and wetting properties. The underwater
88 bubble behavior on both sides of the Janus-CC was
89 investigated, and the results showed that one side of the
90 Janus-CC was superaerophilic, while the other side was
91 aerophobic. The results obtained in this study hold great
92 promise for designing cost-effective Janus materials for
93 effective capture and controlled transportation of bubbles in
94 fluids. 95

96 ■ RESULTS AND DISCUSSION

97 **Characterization.** The FE-SEM images in Figure S1,
98 Supporting Information, revealed that the surface of the
99 superhydrophobic PDMS-coated CC was smooth. The
100 morphology of both sides of the CC changed dramatically
101 after one side of the fabric CC was exposed to an alcohol 101 112

102 burner flame, Figures 1a–c and 2a–c. The appearance of
 103 nanoparticles on both sides of the sample was evident (size <
 104 100 nm). It should be noted that the size of the nanoparticles
 105 on the face nonexposed to flame was smaller than that on the
 106 other side, i.e., flame-exposed face (Figures 1c and 2c).
 107 However, the deposited layer of nanoparticles on the side
 108 nonexposed to flame of the Janus-CC was much thicker than
 109 the flame-exposed side, which could be seen with the naked
 110 eye.

111 Figure S2a–c depicts the EDX elemental analysis results of
 112 the superhydrophobic PDMS-coated CC and both sides
 113 (superhydrophobic and superhydrophilic) of the Janus-CC.
 114 The EDX plot of the superhydrophobic CC contains C, O, and
 115 Si peaks, indicating the presence of a PDMS layer on the CC
 116 surface. The increased weight percentages of O and Si on both
 117 sides of the Janus-CC clearly reveal the formation of SiO₂
 118 particles.

119 Elemental mapping images of the superhydrophobic and
 120 both sides of the Janus-CC are displayed in Figure 3i–iii. It
 121 could be clearly seen that C, O, and Si are uniformly
 122 distributed on all three surfaces. It is noteworthy that for the
 123 Janus-CC sample, the carbon abundance is lower but the O
 124 and Si contents are higher than for the superhydrophobic
 125 sample, confirming the formation of SiO₂ nanoparticles on
 126 both sides of the Janus-CC.

127 The ATR-IR spectra of both sides of the Janus-CC are
 128 depicted in Figure S3. For both sides of the sample, the broad
 129 peak at 1080–1090 cm⁻¹ is assigned to the Si–O–Si bond
 130 stretching of SiO₂ due to the formation of SiO₂ nanoparticles
 131 on both sides of the Janus-CC. For the superhydrophobic side,
 132 a peak at 803 cm⁻¹, which belongs to the Si–CH₃ bending, is
 133 observed, indicating the presence of PDMS and its
 134 oligomers.^{42–44}

135 To identify the elements on the surface and their chemical
 136 environment, XPS analysis was performed on pristine CC,
 137 superhydrophobic CC (PDMS-coated CC), and both sides of
 138 the Janus-CC samples. The survey spectrum of pristine CC is
 139 shown in Figure S4a. It comprises two peaks at 285.0 and
 140 532.0 eV corresponding to C 1s and O 1s, respectively. As
 141 determined from this spectrum, about 95% of the CC surface is
 142 composed of carbon atoms. The high-resolution XPS spectrum
 143 of the C 1s is depicted in Figure S4b. The fitting results reveal
 144 that the mentioned peak consists of three bands: the most
 145 intense peak at 285.0 eV belongs to carbon–carbon (C–C) or
 146 hydrogen (C–H) bonds, the band at 286.0 eV is due to C–O,
 147 and the peak at 290.2 eV is ascribed to COO groups.
 148 Therefore, it can be concluded that the pristine carbon cloth
 149 surface mainly comprises carbon, along with a small amount of
 150 oxygen-containing functional groups, originating from surface
 151 contamination or slight oxidation of the CC surface during
 152 storage and handling. Figure S5a exhibits the survey XPS
 153 spectrum of the superhydrophobic (PDMS-coated) CC. It
 154 consists of three elements: Si 2p (103.1 eV), C 1s (285.0 eV),
 155 and O 1s (532.0 eV) with different atomic percentages; the
 156 percentage of oxygen on the surface of this sample is high,
 157 which is related to the PDMS coating. The high-resolution
 158 XPS spectrum of C 1s (Figure S5b) could be fitted with the
 159 main component at 285.0 eV corresponding to C–C/C–H
 160 bonds. The Si 2p band (Figure S5c) comprises a main
 161 component at 103.1 eV assigned to silicon bonded to oxygen
 162 (Si–O_x), confirming the presence of PDMS on the surface of
 163 this sample. For the superhydrophilic side of the Janus-CC, the
 164 wide XPS spectrum in Figure 4a reveals three peaks at 285.0,

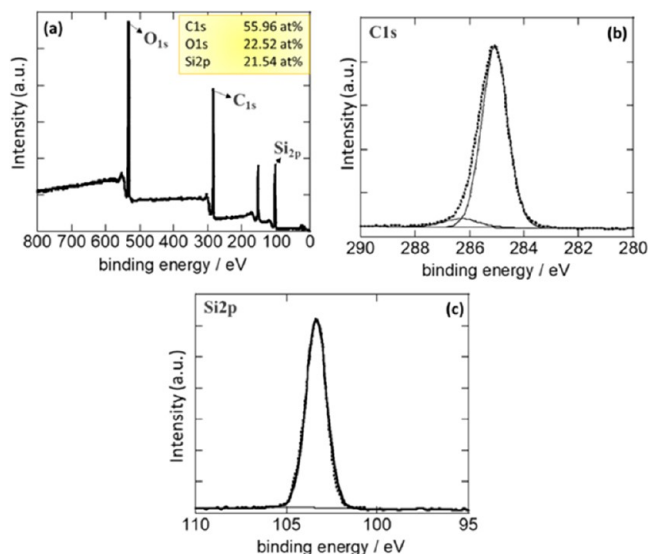


Figure 4. (a) XPS survey spectrum and high-resolution spectra of (b) C 1s and (c) Si 2p of the superhydrophilic side of the Janus-CC sample.

103.1, and 532.0 eV ascribed to C 1s, Si 2p, and O 1s, 165
 respectively. Figure 4b displays the high-resolution spectrum of 166
 the C 1s. It could be curve-fitted with two peaks at 285.0 eV 167
 (C–C/C–H) and 286.0 eV (C–O). Figure 4c depicts the 168
 high-resolution spectrum of Si 2p, which consists of only one 169
 peak at 103.5 eV due to SiO₂. The results infer that the 170
 superhydrophilic side of the Janus-CC sample is essentially 171
 composed of SiO₂ and carbon. Figure 5a exhibits the survey 172 15

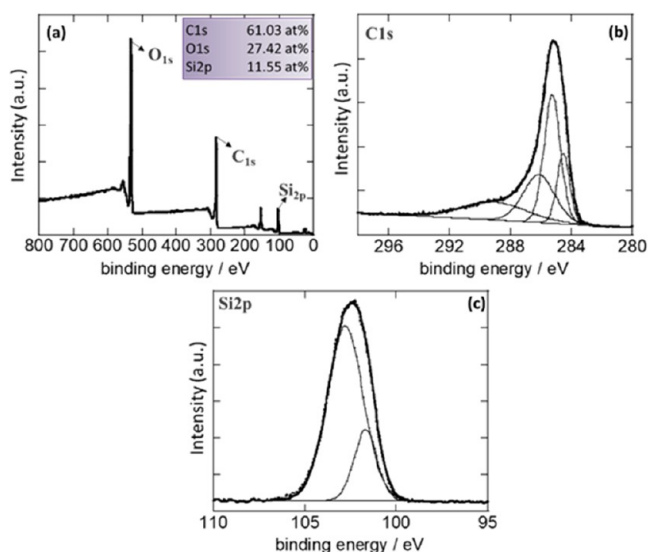


Figure 5. (a) XPS survey spectrum and high-resolution spectra of (b) C 1s and (c) Si 2p of the superhydrophobic side of the Janus-CC sample.

XPS spectrum of the superhydrophobic side of the Janus-CC 173
 sample. It features similar peaks at 103.1, 285.0, and 532.0 eV 174
 ascribed respectively to Si 2p, C 1s, and O 1s. The high- 175
 resolution spectrum of C 1s (Figure 5b) could be 176
 deconvoluted into several bands at 284.5 eV (Si–C), 285.5 177
 eV (C–C/C–H), 286.5 eV (C–O), and 289.1 eV (COO). 178
 These peaks originate from PDMS and its oligomers on the 179

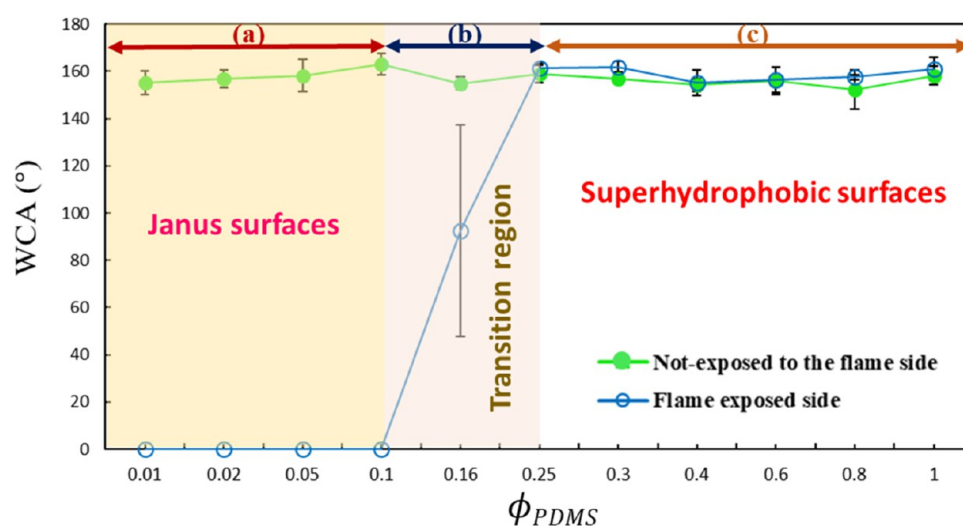


Figure 6. Diagram of WCA changes on both sides of the prepared Janus-CC samples as a function of ϕ_{PDMS} .

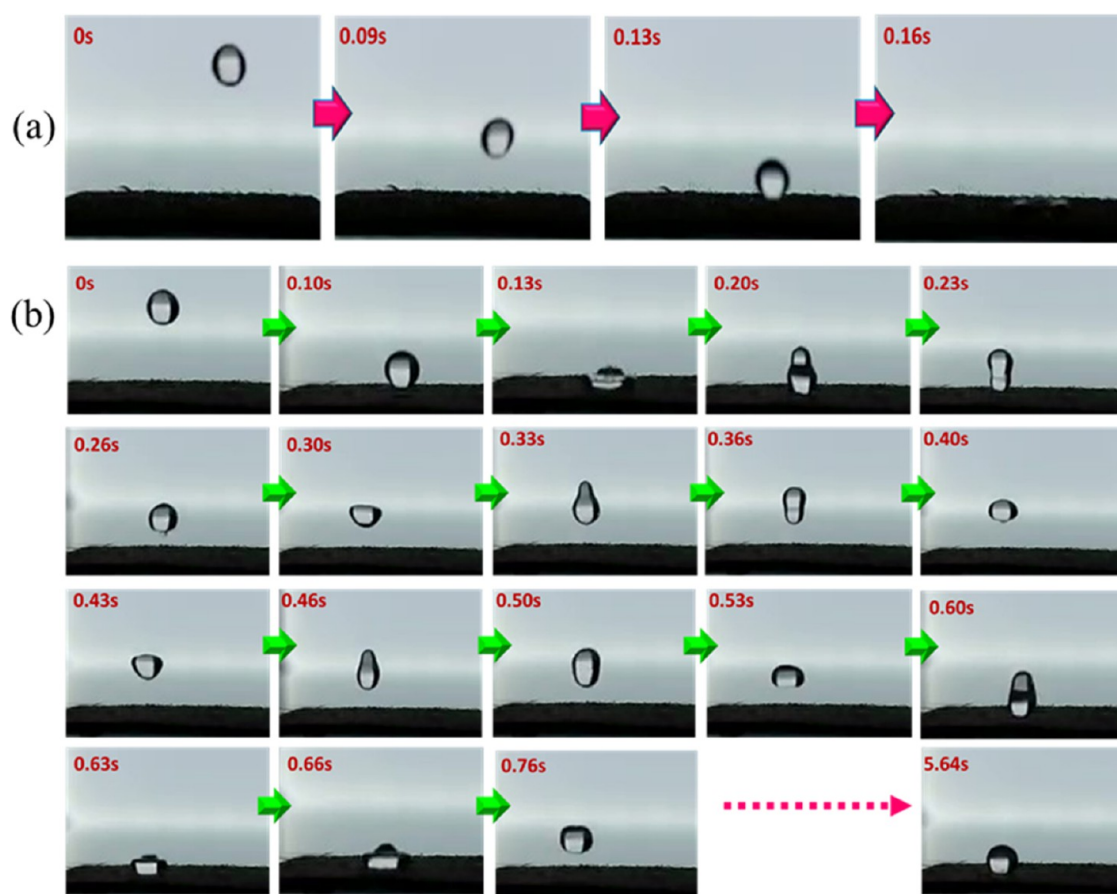


Figure 7. Sequential images of the impact of a falling water droplet on the (a) superhydrophilic and (b) superhydrophobic sides of the Janus-CC sample.

180 superhydrophobic side of the Janus-CC. The high-resolution
181 spectrum of Si 2p (Figure 5c) could be fitted with two peaks at
182 102.1 and 103.6 eV related to PDMS and SiO₂, respectively.⁴²
183 Therefore, the presence of SiO₂ nanoparticles adhering to the
184 carbon surface on the superhydrophobic side of the Janus-CC
185 sample is clearly confirmed. Therefore, based on all of the
186 studied characterization methods, it can be concluded that the
187 superhydrophilic side of the Janus sample consists of SiO₂
188 nanoparticles deposited on carbon cloth, whereas the super-

hydrophobic side comprises a very thin layer of PDMS and its
oligomers coated on the surface of SiO₂ nanoparticles
deposited on the carbon cloth.

Based on the obtained analysis results, the proposed
structure of the prepared Janus-CC is schematically presented
in Figure S6b.

The effect of different amounts of PDMS on the wetting
properties of both sides (exposed and nonexposed to the
flame) of the synthesized sample was investigated through

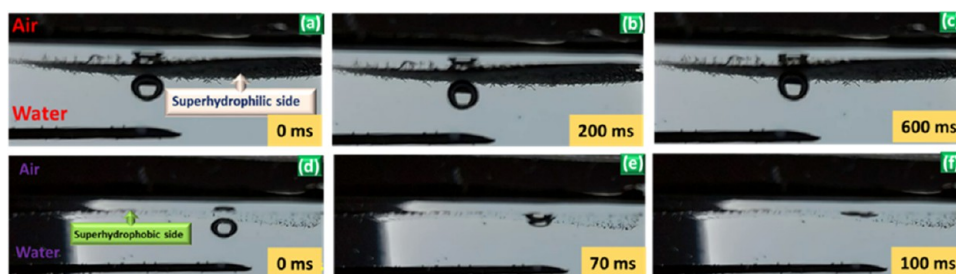


Figure 8. Bubble behavior at the air–water interface of the (a–c) superhydrophilic and (d–f) superhydrophobic sides in contact with water.

198 water contact angle (WCA) and sliding angle (SA) measure-
 199 ments. Figure 6 presents the diagram of the measured WCA
 200 versus ϕ_{PDMS} , which can be divided into three separate
 201 sections. In region “a” ($0.01 \leq \phi_{\text{PDMS}} \leq 0.1$), the prepared
 202 samples are Janus; thus, the flame-exposed side is super-
 203 hydrophilic and the side nonexposed to flame is super-
 204 hydrophobic. In region “b” ($0.1 < \phi_{\text{PDMS}} \leq 0.25$), the WCA of
 205 the flame-exposed side changes from 0 to 155° ; therefore, the
 206 wettability of this side changes from superhydrophilic to
 207 superhydrophobic, while the WCA of the other side remains
 208 high and constant ($\sim 155^\circ$; superhydrophobic). In region “c”
 209 ($0.25 < \phi_{\text{PDMS}} \leq 1$), the synthesized samples are completely
 210 superhydrophobic. Figure S7 shows the sliding angle on the
 211 surface of the synthesized samples. At $0.01 \leq \phi_{\text{PDMS}} \leq 0.1$, the
 212 flame-exposed side has a superhydrophilic property and is
 213 wetted completely by water. However, at $\phi_{\text{PDMS}} > 0.2$, the SA
 214 is lower than 2° . For the side nonexposed to flame, the sliding
 215 angle decreases from 5° ($\phi_{\text{PDMS}} = 0.01$) to 2° ($0.05 \leq \phi_{\text{PDMS}}$).
 216 According to the results of Figures 6 and S7, $\phi_{\text{PDMS}} = 0.1$ was
 217 selected for the fabrication of the Janus-CC.

218 It has been previously reported that the thermal decom-
 219 position of PDMS, in air, leads to the formation of a mixture of
 220 oligomers, carbon dioxide, and a very soft powder of SiO_2 .⁴²
 221 Therefore, it could be concluded that when the PDMS-coated
 222 CC was exposed to an alcohol lamp flame, a small amount of
 223 SiO_2 was formed on the higher-temperature side, which leads
 224 to surface superhydrophilicity. However, a large amount of
 225 SiO_2 is also formed on the backside (PDMS-coated CC
 226 nonexposed to flame) of the CC, which is the reason for the
 227 whitening of the surface (Figure S6). On the side nonexposed
 228 to flame of the CC, the deposition of SiO_2 nanoparticles led to
 229 an increase of surface roughness. The enhanced roughness
 230 combined with the adsorption of evaporated PDMS and its
 231 oligomers on the surface of SiO_2 nanoparticles confers a
 232 superhydrophobic character to the side nonexposed to flame of
 233 the CC. Therefore, a Janus-CC featuring both super-
 234 hydrophilic and superhydrophobic sides is formed easily.

235 To evaluate the stability of the wettability of both sides of
 236 the Janus-CC, WCA measurements at different time intervals
 237 were performed (Figure S8). Also, the water sliding angle was
 238 assessed at different time intervals, and no change was
 239 observed over a long period. Therefore, the wettability
 240 properties of the synthesized Janus-CC are stable for a long
 241 time (53 days).

242 **Investigation of Thermal Stability.** The thermal stability
 243 of the prepared Janus-CC was examined by heating in an
 244 electric furnace (Figure S9). The furnace temperature reached
 245 100°C at a speed of $10^\circ\text{C}/\text{min}$ and was kept at this
 246 temperature for 1 h; then, the temperature reached the desired
 247 temperature (200, 300, 400, and 500°C) and was maintained
 248 for 4 h. The WCA, measured on the superhydrophobic side of

the Janus-CC, was stable up to 400°C . However, at higher
 249 temperatures, because of the desorption and decomposition of
 250 adsorbed PDMS and its oligomers, the surface lost its
 251 hydrophobicity and became superhydrophilic.

**Falling Droplets on the Superhydrophilic and Super-
 hydrophobic Surfaces of the Janus-CC.** In this section, a
 254 water droplet was dropped on both sides of the Janus sample
 255 and the images were recorded with a slow-motion camera. The
 256 water droplet spreads quickly on the superhydrophilic side of
 257 the Janus-CC (less than 0.1 s) (Figure 7a). On the
 258 superhydrophobic side of the Janus-CC, the falling water
 259 droplet impacts the surface, slightly spreads, then shrinks,
 260 bounces several times, and finally rests on the surface with a
 261 spherical shape (Figure 7b). This behavior is characteristic of a
 262 superhydrophobic surface.^{46–48}

Determination of Surface Energy of Janus-CC. Figure
 264 S10 depicts the Zisman diagram for the determination of the
 265 surface energy of the superhydrophobic side of the Janus-CC.
 266 The obtained critical surface tension for the superhydrophobic
 267 side is about $40.7\text{ mN}/\text{m}$. So, any organic solvent with $\gamma \leq 40.7$
 268 mN/m can wet the superhydrophobic side of the Janus-CC.

Wettability of the Janus-CC at Different Interfaces.
 270 The wettability of the Janus-CC at the oil–water and air–
 271 water interfaces was investigated. Figure S11a shows a Janus-
 272 CC at the oil–water interface; the sample is placed in such a
 273 way that the superhydrophobic side is in contact with oil and
 274 the superhydrophilic side is in contact with water. As seen in
 275 the figure, despite the contact of the superhydrophobic surface
 276 with oil, the sample still retains its superhydrophobicity and is
 277 stable at the two-phase interface. On the other hand, a Janus-
 278 CC was placed in the opposite direction at the oil–water
 279 interface (Figure S11b) and water droplets were injected into
 280 the superhydrophilic side; the surface is still superhydrophilic
 281 and is wetted completely. The injection of water droplets
 282 continued until the surface became saturated, and then the
 283 collected water drained to the aqueous phase.

284 In another experiment, Janus-CC was placed at the air–
 285 water interface and the oil wettability was evaluated on both
 286 sides of the Janus sample. In Figure S12a, the super-
 287 hydrophobic side of the Janus-CC was in contact with water
 288 and then an oil droplet was injected into the surface. It was
 289 seen that the oil droplet rapidly absorbs and spreads on the
 290 superhydrophobic side of the Janus-CC surface, indicating its
 291 oleophilicity. Then, the sample was inverted such that the
 292 superhydrophilic side was in contact with the water (Figure
 293 S12b). When an oil droplet was injected, it maintained its
 294 spherical shape, confirming its oleophobicity. To further
 295 evaluate the performance of the Janus-CC, it was completely
 296 immersed in a water container and oil was injected into both
 297 sides. It is obvious that the superhydrophilic side of the Janus-
 298 CC is oil-repellent underwater and the oil droplet remains
 299

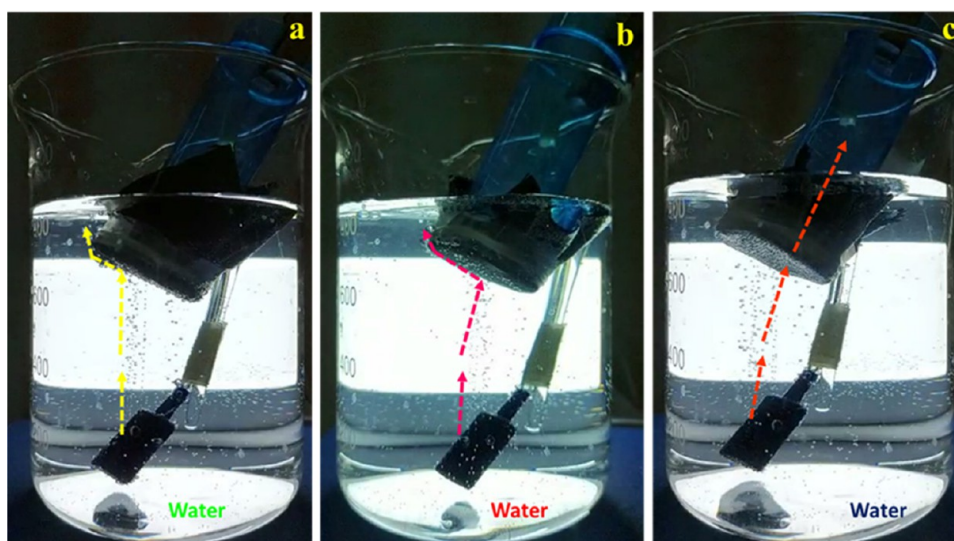


Figure 9. Continuous flow of tiny gas bubbles hitting (a) pristine carbon cloth, (b) superhydrophilic side of the Janus-CC, and (c) superhydrophobic side of the Janus-CC.

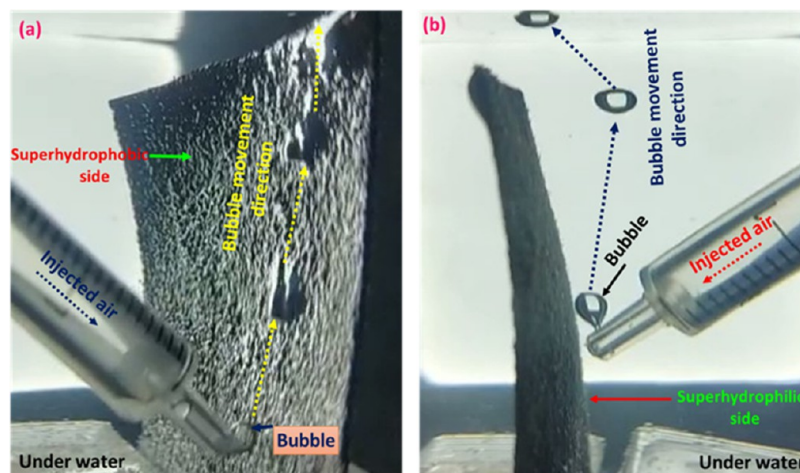


Figure 10. Injection of bubbles into the vertically positioned Janus-CC: (a) superhydrophobic side, and (b) superhydrophilic side.

spherical (Figure S13a). For the superhydrophobic side of the Janus-CC, the oil droplets spread immediately and the surface exhibits underwater oleophilicity (Figure S13b, Supporting Information). Figure S14 illustrates the lotus leaf property of the prepared Janus-CC, which floats stably at the air–water interface in such a way that the upper side is superhydrophobic and the bottom side is superhydrophilic and also underwater oleophobic.

Gas Bubble Behavior of the Janus-CC Positioned at the Air–Water Interface. The aerophilicity of the prepared Janus-CC was investigated by placing it at the air–water interface and then injecting a bubble. As shown in Figure 8a–c, when the superhydrophilic side is in contact with water, the bubble cannot pass through the Janus-CC because the superhydrophilic side is completely wetted and covered with water, creating a polar surface, which repels the bubble (nonpolar medium). On the other hand, when the superhydrophobic side is in contact with water, the bubble passes easily through the Janus-CC membrane within 100 ms (Figure 8d–f). In this case, the surface is not wetted by water and retains its nonpolar character and also porosity and hence attracts the bubble.

Underwater Collection of a Continuous Flow of Gas Bubbles. According to the behavior of the synthesized sample in dealing with bubbles, an experiment was designed using a continuous flow of tiny bubbles in such a way that a tube was covered with pristine CC or the superhydrophobic or superhydrophilic side of the Janus-CC, Figure 9. When the tiny gas bubbles hit the surface of pristine CC and superhydrophilic side of the Janus-CC, their direction changes and they move toward the surface of water, Figure 9a,b. In contrast, when the flow of the tiny gas bubbles hits the superhydrophobic side of the Janus-CC, the bubbles are absorbed by the surface and transferred into the tube, Figure 9c. This interesting behavior of the Janus-CC suggests that it can be used for the collection of gas bubbles released in aqueous media.

Injecting Gas Bubbles into the Vertically Aligned Janus-CC Immersed in Water. In this part, a Janus-CC was immersed vertically in a water container; then, gas bubbles were injected into both sides of the Janus-CC surface using a syringe. Interestingly, the superhydrophilic side repels the bubbles, while the superhydrophobic side captures all of the

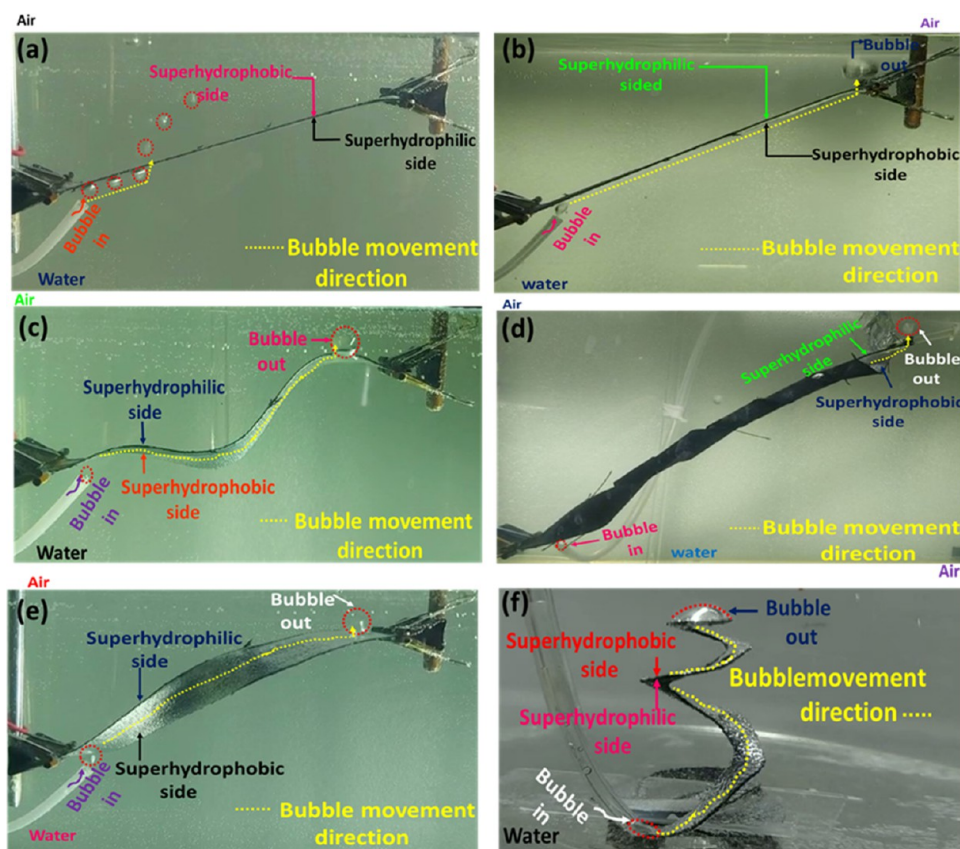


Figure 11. Bubble capture and transportation by the superhydrophobic side of the Janus-CC strip with different shapes. (a) Inclined shape with the superhydrophobic side at the bottom, (b) inclined shape with the superhydrophobic side at the bottom, (c) curved shape, (d) twisted with a high degree of torsion, (e) twisted with a low degree of torsion, and (f) spiral shape.

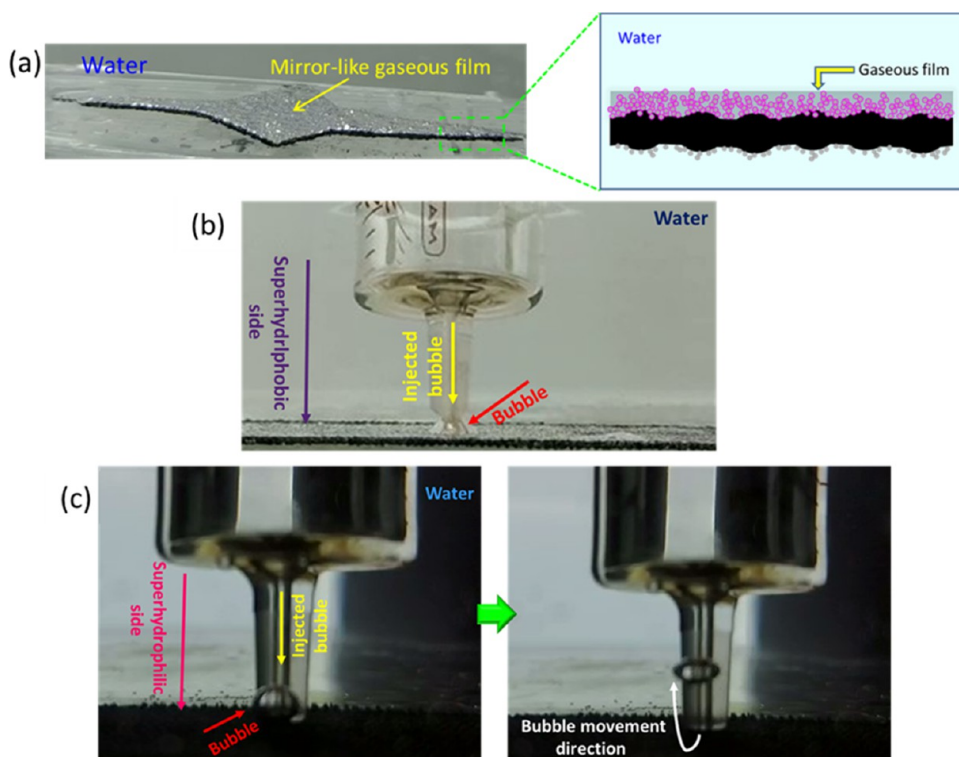


Figure 12. (a) Formation of a mirrorlike gaseous film on the superhydrophobic side of the Janus-CC immersed into water and underwater injection of a gas bubble into the (b) superhydrophobic and (c) the superhydrophilic sides of the Janus-CC.

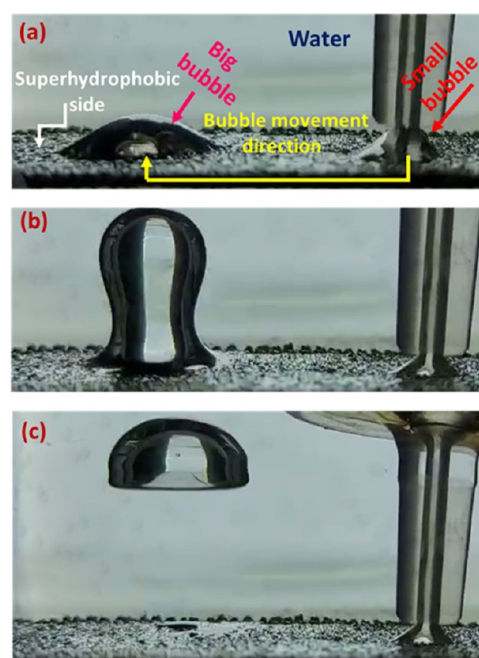


Figure 13. Transfer of a small bubble toward a larger one on the superhydrophobic surface via the gaseous film.

injected into another place on the surface, Figure 13b. The small bubble was absorbed and migrated through the gaseous film toward the larger bubble and then merged into a bigger one. The reason for the migration of the smaller bubble to the larger one is the difference in Laplace pressure. Finally, they merge and a big bubble detaches from the surface due to the buoyant force, Figure 13c and Video S3.

For a better understanding of the mechanism of bubble transfer on the superhydrophobic surface of the Janus-CC sample through the gaseous film, various designs of the Janus-CC were made and the bubble transfer abilities were investigated. In all designed patterns (flowerlike, triangular, etc.), the Janus sample has both narrow and wide parts (Figure 14). In all tested designs, it was observed that if the bubble was injected into the narrow part of the sample, it spontaneously and quickly transferred to the wider part through the gaseous film, whereas if the bubble was injected into the wide part of the sample, it was tethered and remained there in the form of a paraboloid of revolution shape. The reason for the spontaneous transfer of the bubble from the narrow to the wide part of the sample is due to the presence of higher structural Laplace pressure in the narrow part of the sample,⁴⁵ which causes the bubble to be transferred to the part with a lower structural Laplace pressure (wide part of the sample). For the flowerlike structure, it was seen that, by injecting the bubbles into the center of the sample (narrow part of the sample), the bubbles were distributed spontaneously and rapidly toward all petals (wide part of the sample). This interesting result indicates that the Janus-CC with the flowerlike pattern can be used for bubble distribution and transfer to different places, Figure 14 and Video S4.

In another experiment, the bubble transfer process on the superhydrophobic side of an inclined Janus-CC ($\alpha = 30^\circ$) with a rectangular shape (having no structural Laplace pressure) was examined (Video S5). At an angle of 30° when the bubble was injected into the lower part of the sample, it was transmitted through the gaseous film and finally exited from the upper part

injected bubbles and transfers them into the collecting tube (Figure 10 and Video S1).

Gas Bubble Absorption and Transportation. In this section, the performance of the Janus-CC in bubble transportation was further assessed. Initially, a long strip of Janus-CC was made and positioned in an inclined orientation. When the air pump is turned on, the gas bubbles leave the superhydrophobic side after a short distance and enter the bulk water, Figure 11a. For the reverse position, the superhydrophobic surface acts as a bridge, directing the incoming bubbles to the end of the strip, Figure 11b. To evaluate and demonstrate the ability of the Janus-CC strip for bubble transportation, the Janus strip was reformed into various shapes, including curved (Figure 11c), completely twisted (Figure 11d), partially twisted (Figure 11e), and spiral (Figure 11f). The results of the experiments clearly evidenced that, under any torsional conditions, the superhydrophobic side of the Janus strip is capable of targeted bubble transfer. Figure S15 depicts a close-up view of bubble transfer on the superhydrophobic side of the twisted Janus-CC strip with a low torsional degree. Also, Video S2 shows the excellent performance of the spiral Janus-CC strip for capturing and transporting gas bubbles.

To further test the ability of the superhydrophobic side of the Janus-CC to transfer bubbles, a thin strip of the sample was placed in a water container. The bottom part of the sample was positioned next to the bubble production tube, and its upper part was placed inside an inverted cylindrical glass containing colored water (Figure S16). As can be seen, the superhydrophobic surface of the Janus-CC acts as a bridge for the targeted transport of bubbles, and the reduction in the height of the colored water column clearly indicates the ability of the sample to transport bubbles.

The bubble transfer rate on an inclined Janus-CC strip was investigated at various tilting angles (α). The results in Figure S17 reveal that, by increasing α , the bubble transfer rate increases. It is worth noting that at very low angles (10°), the transfer rate is also high (rate > 16 cm/s).

The Janus-CC was attached to a glass slide surface and placed horizontally at the bottom of a water container. As shown in Figure 12a, the superhydrophobic side of Janus-CC exhibits a mirrorlike feature underwater due to the formation of a gaseous film on its surface. Since the superhydrophobic side is completely nonpolar and porous, air (as a nonpolar phase) is trapped inside it, and by placing the sample underwater, a gaseous film is formed at the solid–liquid interface, Figure 12a. This gaseous film is very stable, and when a bubble is injected into the superhydrophobic side, the bubble is tethered to the surface due to the cohesion force between the gas bubble and the gaseous film, Figure 12b. Therefore, underwater, the superhydrophobic side of Janus-CC is superhydrophobic, but when the bubble is injected into the superhydrophilic side of the Janus-CC, the bubble is repelled, Figure 12c, because the superhydrophilic side is completely covered by water and forms a polar medium and therefore is superhydrophilic.

Since the superhydrophobic side of the Janus-CC displays underwater superhydrophobic character, more detailed studies were performed. Figure 13 and Video S3 exhibit interesting behavior of the superhydrophobic side underwater. First, a bubble was injected into the surface, the bubble was absorbed and stayed on the surface in the form of a paraboloid of revolution shape (Figure 13a). Then, a smaller bubble was

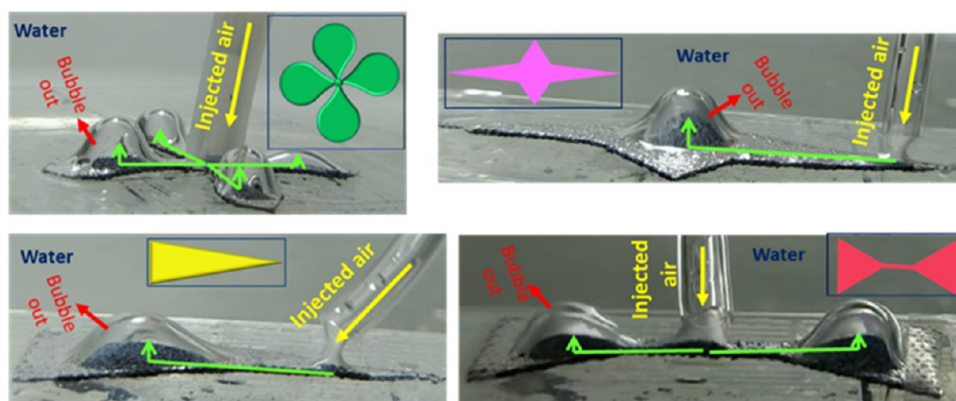


Figure 14. Movement of bubbles on the superaerophilic side of the Janus-CC from the narrow to wide part.

of the sample due to the buoyant force, Figure 15a. In the next experiment, the Janus-CC with a triangular shape was placed in

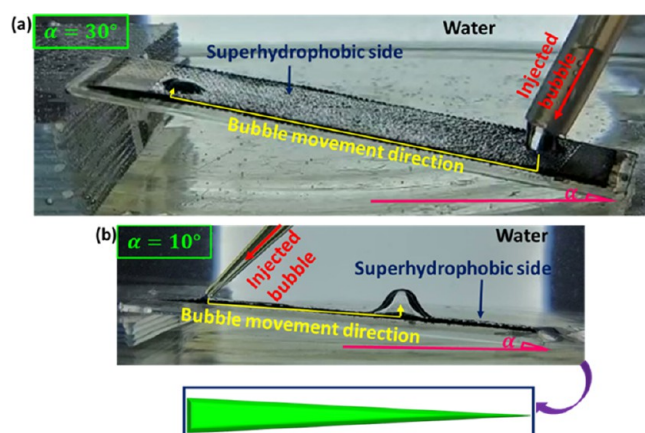


Figure 15. Bubble transfer on the superaerophilic side of an inclined Janus-CC: (a) on a rectangular sample and the movement is in the direction of the buoyant force and (b) on a triangular sample and the movement is in the opposite direction of the buoyant force.

an inclined position at an angle of 10° so that the vertex of the triangle points upward and its base faces downward. The bubble was injected into the vertex of the triangle, and surprisingly, it was observed that the bubble moved downward and exited from the bottom side near the triangle base, Figure 15b. The reason for this downward movement is the dominance of structural Laplace pressure of the narrow part of the surface over the buoyant force.

To evaluate the bubble transfer performance of the Janus-CC at high temperatures, the bubble movement in boiling water in the absence and presence of Janus-CC was investigated (Figure S18). The result revealed that the superaerophilic side of the Janus-CC captures the released bubbles and increases their upward movement. The obtained rate of bubble movement in bulk water is 0.05 m/s, while for Janus-CC, it is 0.31 m/s.

In the next part, an experiment was designed to evaluate the ability of the Janus-CC to transport bubbles under harsh physical conditions. In this experiment, air was pumped from the bottom side of a container of water and sand (Figure S19a). In the absence of the Janus-CC sample, the bubbles came out irregularly from different positions on the sand surface (Figure S19a). In contrast, when a Janus strip was

placed inside the water/sand container, all of the bubbles were captured by the superaerophilic side of the Janus-CC and transferred to the surface of the water in one direction (Figure S19b). As a result, the Janus-CC has retained its ability to transport bubbles under harsh physical conditions, and it can be used for gas bubble transport even inside the sand.

Applications of the Janus-CC for Gas Bubble Transfer. Unidirectional Gas Membrane. A setup was designed to assess the gas transfer into the liquid phase through a Janus-CC membrane. A Janus-CC piece was placed between two tubes as a membrane in a way that the superhydrophobic side was in the upward direction (in contact with the aqueous phase) and the superhydrophilic side was in the downward direction. In the next step, an alkaline solution of phenolphthalein was poured into the upper tube. The remarkable point is that, due to the superhydrophobicity of the upper side, the solution could not move downward and remained on the membrane. The lower tube was connected to a CO_2 source. It was obvious that the CO_2 gas was easily transferred from the superhydrophilic to the superhydrophobic side. With the entry of CO_2 gas bubbles into the alkaline solution of phenolphthalein, the pH of the solution decreased ($2\text{NaOH} + \text{CO}_2 \rightarrow \text{Na}_2\text{CO}_3 + \text{H}_2\text{O}$) and the color of the solution changed from pink to colorless within 49 s (Figure S20 and Video S5). So, the Janus-CC did not permit the aqueous solution to move downward, while allowing the gas to pass through it in the upward direction. The results clearly indicate that the Janus-CC could be successfully applied as a membrane (or valve) for gas entering into an aqueous solution.

Underwater Gas Transformation for Reaction. Simultaneous underwater transfer of CH_4 and air was examined by a Janus-CC. Two Janus strips were placed at the bottom of a water container in a way that the upper parts of the two carbon strips were put inside a glass syringe. Then, air was injected into the bottom part of one of the Janus strips and methane gas into the other one. It follows that CH_4 and air were transported into the glass syringe by the Janus strips and mixed with each other, confirmed using a flame at the tip of the syringe (Figure S21).

Debubbling in the Fluid Transfer Pipe. An experiment was performed to demonstrate the effective role of Janus-CC in debubbling in a fluid transfer pipe (Figure S22). Water stream and air were injected into a pipe having a duct for bubble exit; both bubbles and water droplets exited from the duct (Figure S22a). Upon placing a piece of a Janus-CC in the duct in such a way that the superhydrophobic (superaerophilic) side becomes the outer side, the moving bubbles were strongly

515 captured by the Janus-CC, but the water was repelled, and only
516 bubbles could exit from the duct (Figure S22b). This
517 experiment evidences the ability of Janus-CC to debubble
518 from the liquid stream in a pipe and prevent the secondary
519 problems caused by bubbles (such as corrosion of pipes).

520 ■ CONCLUSIONS

521 In this paper, Janus-faced carbon cloth (Janus-CC) was
522 successfully fabricated by a novel, cost-effective, fast, and
523 simple strategy. The hydrophilic side of the sample was
524 composed of a thin layer of deposited SiO₂ nanoparticles,
525 while its superhydrophobic side consisted of a thick layer of
526 SiO₂@PDMS. The superhydrophobic face of the Janus-CC
527 displayed a superaerophilic property underwater. Thanks to
528 the superaerophilic character, the Janus-CC demonstrated the
529 ability to spontaneously capture and directionally transport
530 underwater gas bubbles. The Janus-CC with a strip shape was
531 able to transfer gas bubbles at a long distance with a high speed
532 at any torsional degree. The underwater gaseous film formed
533 on the superaerophilic side of the Janus-CC was responsible
534 for its successful capture and transport of the bubbles. Our
535 Janus-CC was not only capable of the collection and controlled
536 transfer of bubbles but also featured the ability of unidirec-
537 tional penetration of underwater gas. The prepared Janus-CC
538 had high thermal, mechanical, and long-life stability. The
539 presented strategy and material provided new insights into the
540 fabrication of advanced surfaces with high performance for fast
541 capture, collection, and targeted transfer of underwater
542 bubbles. The prepared Janus-CC was successfully utilized as
543 a controlled underwater gas bubble transporter, a membrane
544 for unidirectional gas permeation and reaction in the aqueous
545 phase, an underwater gas transporter for reaction, and material
546 for debubbling of a fluid stream in a pipe.

547 ■ EXPERIMENTAL SECTION

548 **Materials.** Carbon cloth (CH-900–15) with a thickness of 0.50 ±
549 0.05 mm was purchased from Kuraray Chemical Co., Ltd., Japan.
550 Polydimethylsiloxane (PDMS, Sera Sense SF 350 Korea) and *n*-
551 hexane (≥95%, Merck Co.) were used as received.

552 **Preparation of Janus-Faced Carbon Cloth (Janus-CC).** The
553 process of Janus carbon cloth (Janus-CC) preparation is schematically
554 presented in Figure S20. The carbon cloth (CC) pieces were
555 immersed in a binary mixture of PDMS and *n*-hexane at different
556 volume fractions of PDMS ($\phi_{\text{PDMS}} = 0.01, 0.02, 0.05, 0.1, 0.25, 0.4,$
557 $0.6, 0.8,$ and 1) for 30 s. Then, the CC samples were left at room
558 temperature for 1 day for slow *n*-hexane evaporation. The obtained
559 CC at this stage was superhydrophobic.⁴⁹ Finally, one side of the
560 dried sample was placed on top of an alcohol burner flame for 60 or
561 90 s. At this stage, the flame (~500 °C)-exposed side of the PDMS-
562 modified CC lost its PDMS coating by evaporation and
563 decomposition and became superhydrophilic. On the other hand,
564 the other side of the CC (nonexposed to the flame) remained
565 superhydrophobic because of the deposition of the decomposed and
566 evaporated products of PDMS from the other side. Therefore, by this
567 novel and simple method, a Janus-CC was obtained (Figure S23).

568 ■ ASSOCIATED CONTENT

569 ■ Supporting Information

570 The Supporting Information is available free of charge at
571 <https://pubs.acs.org/doi/10.1021/acsami.2c12027>.

572 FE-SEM images of the superhydrophobic carbon cloth,
573 EDX spectra of superhydrophobic carbon cloth and
574 Janus-CC samples, ATR-IR spectra of the Janus-CC
575 sample, XPS survey spectra of samples, schematic of the

structure of Janus carbon cloth, sliding angle of the water
droplet on both sides of the Janus-CC, water contact
angles at different time intervals, stability of Janus-CC,
Zisman diagram of the Janus-CC, wettability of the
Janus-CC at the air–water and kerosene–water inter-
faces, underwater oleophobic/oleophilic properties of
the Janus-CC, lotus leaf behavior of the Janus-CC,
bubble transfer route on the Janus-CC twisted strip,
bubble movement rate diagram, images of bubble
movement at boiling temperature, movement path of
gas bubbles in sand–water, unidirectional gas mem-
brane, simultaneous underwater transfer of two gases by
two Janus carbon cloth, water and bubble flow in a pipe,
and scheme of Janus carbon cloth preparation method
(PDF)

Superhydrophilic side repelling the bubbles, while the
superhydrophobic side capturing all of the injected
bubbles and transferring them into the collecting tube
(AVI)

Excellent performance of the spiral Janus-CC strip for
capturing and transporting gas bubbles (AVI)

Interesting behavior of the superhydrophobic side
underwater (AVI)

Janus-CC with the flowerlike pattern used for bubble
distribution and transfer to different places (AVI)

Bubble transfer process on the superaerophilic side of
inclined Janus-CC ($\alpha = 30^\circ$) with a rectangular shape
(AVI)

Video S6 (AVI)

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