Supporting Information

The Controllable Generation of Interfacial Gas Structures on Graphite Surface by Substrate Hydrophobicity and Gas Oversaturation in Water

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1. *The procedure of the new method producing interfacial gas domains.*

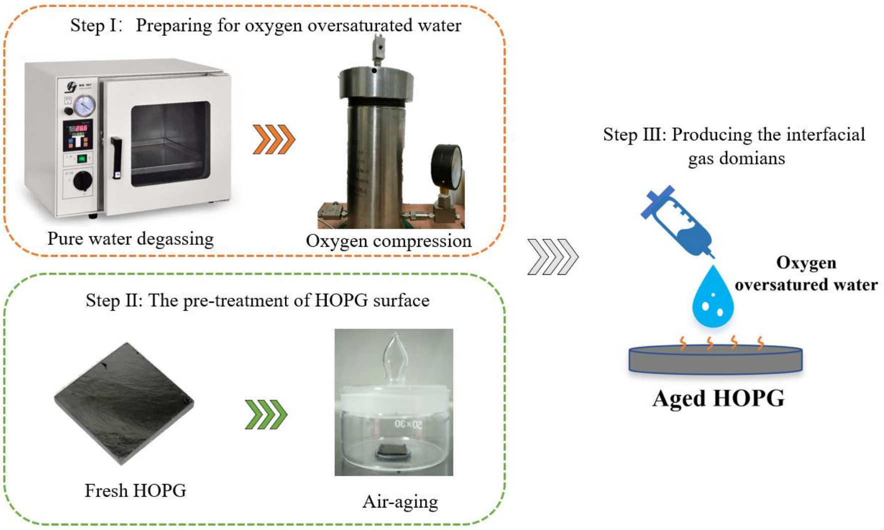


Figure S1. The schematic graph of the new method.

To control the substrate hydrophobicity and gas oversaturation in water, we pre-treated the HOPG surface and pure water before the experiment respectively. The pure water was degassed by the thawing-degassing cycle to remove the air in water, and then compressed the degassed water in oxygen at certain pressure for 5 hours. The fresh HOPG was stored in a ground glass bottles for the aging process in air. Finally, adding the oxygen oversaturated water to the aged HOPG surface produced the interfacial gas domains.

2. ***The in-situ height and frequency shift information of the HOPG surface with different air-aging time.***

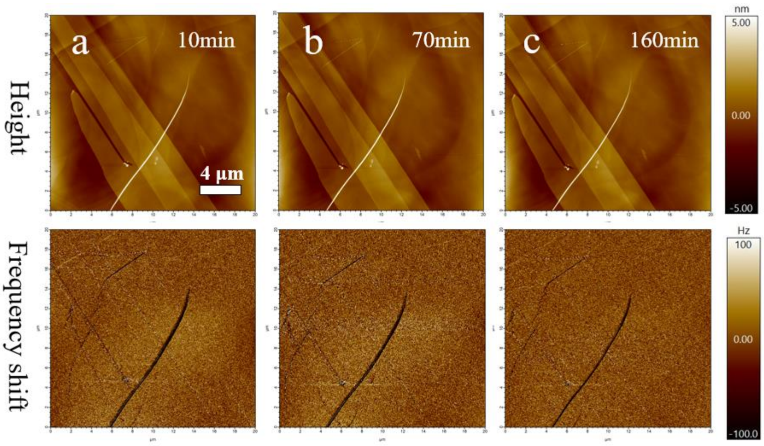


Figure S2. (a-c) The in-situ height and frequency images of the cleaved HOPG surface (20 μm^2) exposed in air for 10, 70, 160 minutes.

We in situ measured the HOPG surface in the air over different times by the AMFM-AFM with the high-sensitivity. According to the comparison of height and frequency shift images, we did not observe new structures formed at the aged surface after exposure to air over a few hours.

3. *Comparing the nucleation of interfacial gas domains at the air-aged HOPG surface in degassed water and cold water.*

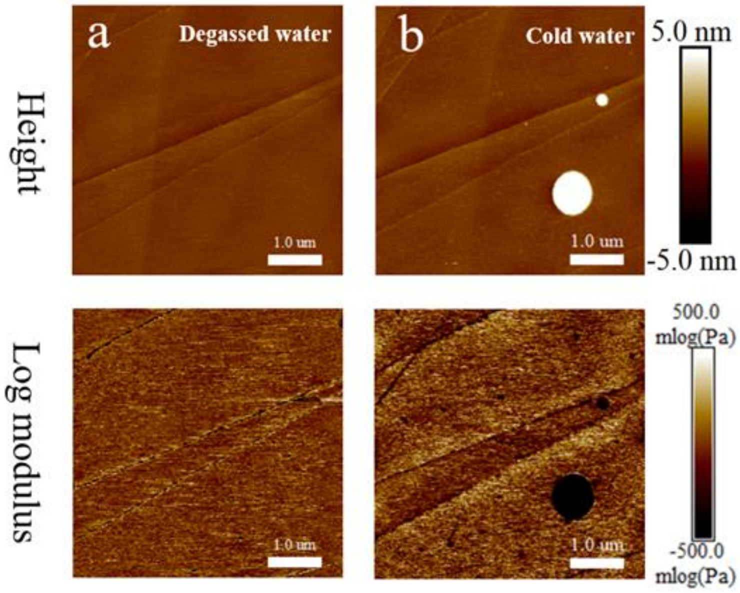


Figure S3. The height and modulus images of the HOPG surface with the air-aging for 60 min, then immersed in degassed water (DO= 2.3 mg/L) and cold water (DO= 11.5 mg/L).

To reinforce the point that the observed layers are gaseous, we conducted in-situ imaging to compare the nucleation of gas domains in degassed water and cold oversaturated water. The degassed water was injected into the surface that aged in the air for 60 mins firstly. As shown in Figure S3b, no nucleation happened in the degassed water. We replaced the degassed water with cold water (4°C storage for 24 hours). In the cold water, we observed surface nanobubbles formed at the surface.

4. *The infrared signals of absorbed hydrocarbons of the different regions of a HOPG surface exposed to the air.*

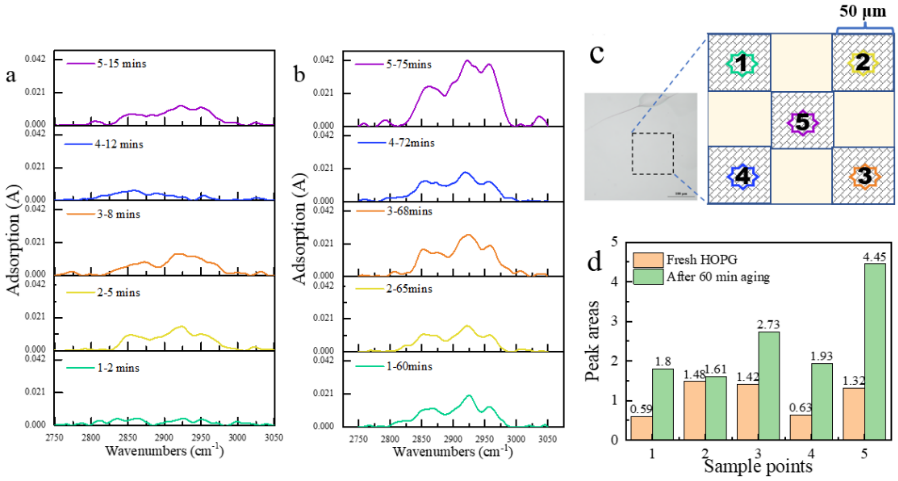


Figure S4. The C—H stretching vibration signals at five adjacent regions (50×50 μm^2). (a) the measured regions of the fresh HOPG surface; (b) the measured regions of the aged HOPG surface; (c) the location of these measured regions; (d) The histogram of the integrated peak areas before and after the air-aging.

To reveal the distribution of absorbed hydrocarbons during the air-aging, we measured the five adjacent regions of the HOPG surface before and after the 60 min air-aging. Acquiring the spectrum of five regions at fresh HOPG surface cost about 15 min in the air. The spectrum results show that the C—H signals with different intensities in the different regions. Especially for region No.4, Despite this area being exposed to the air for 12 min, we did not measure the signal peaks. After the 60 min air-aging, the increasing signal abundances were observed in all measured regions. However, the increments of every region of the aged HOPG surface are various as shown in Figure S4 (d). The observations indicated that the absorbed hydrocarbons are increasing and are not distributed homogeneously during the air-aging.

2. the different efficiency in producing visible micropancakes of two methods

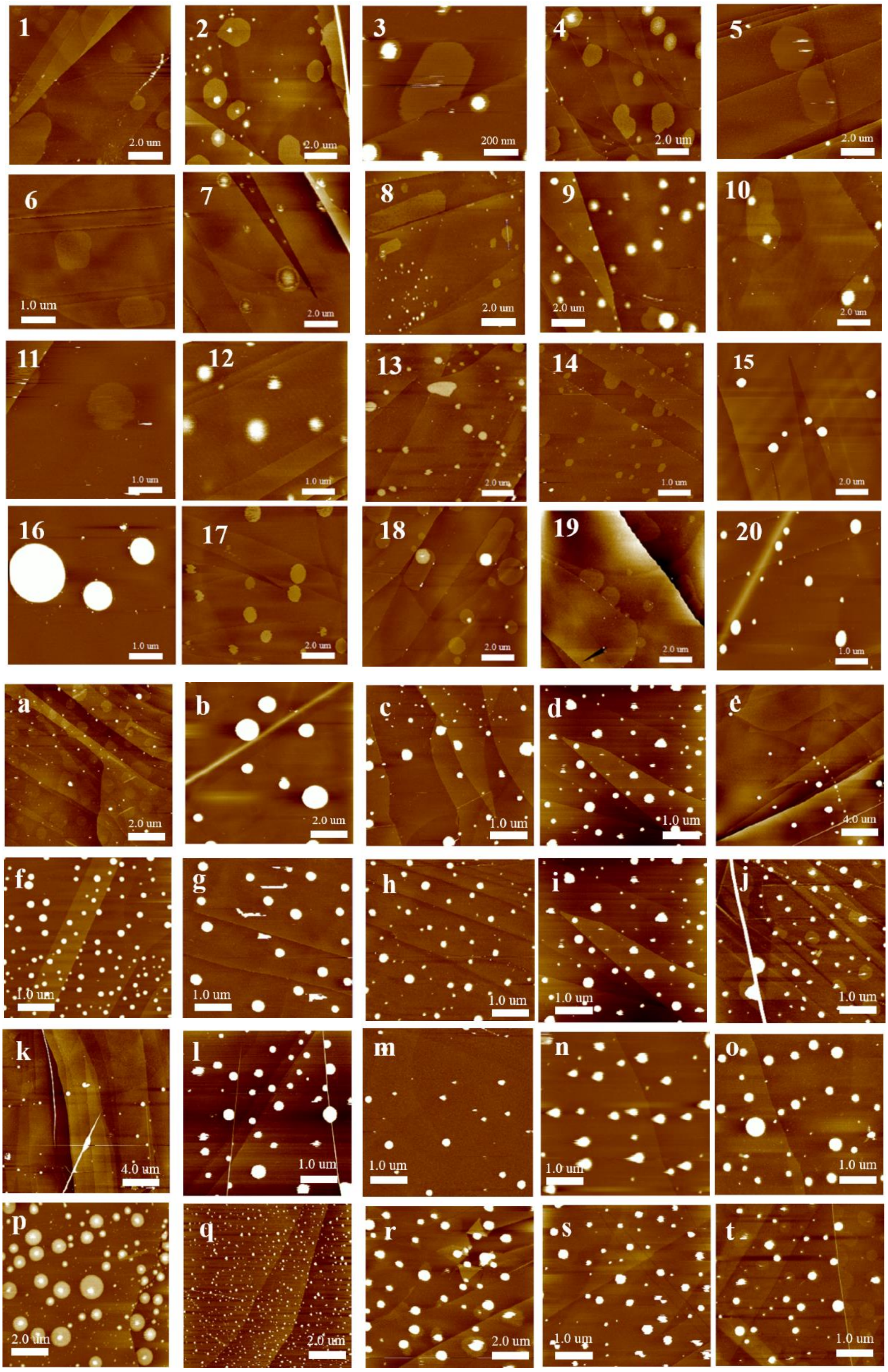
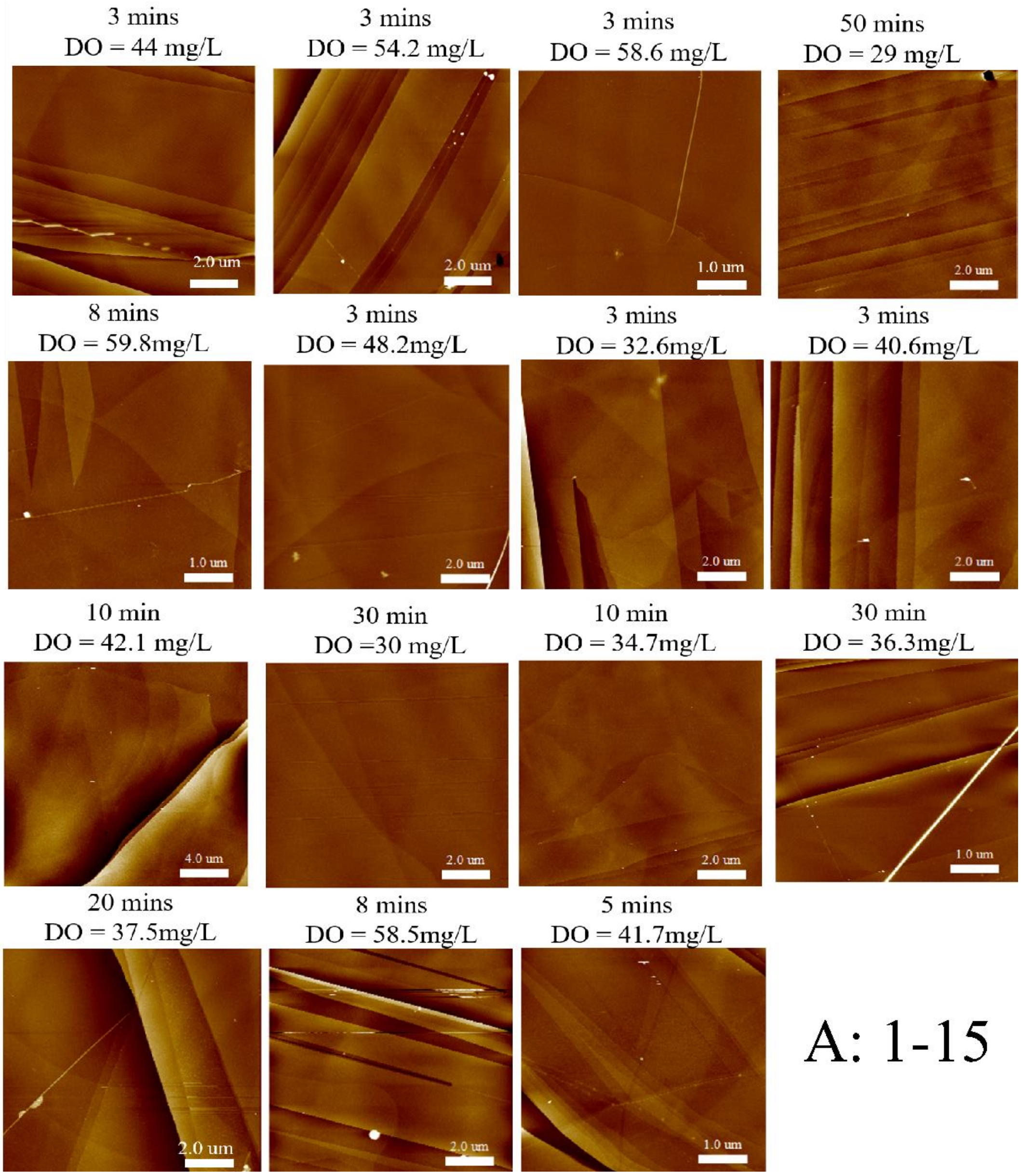
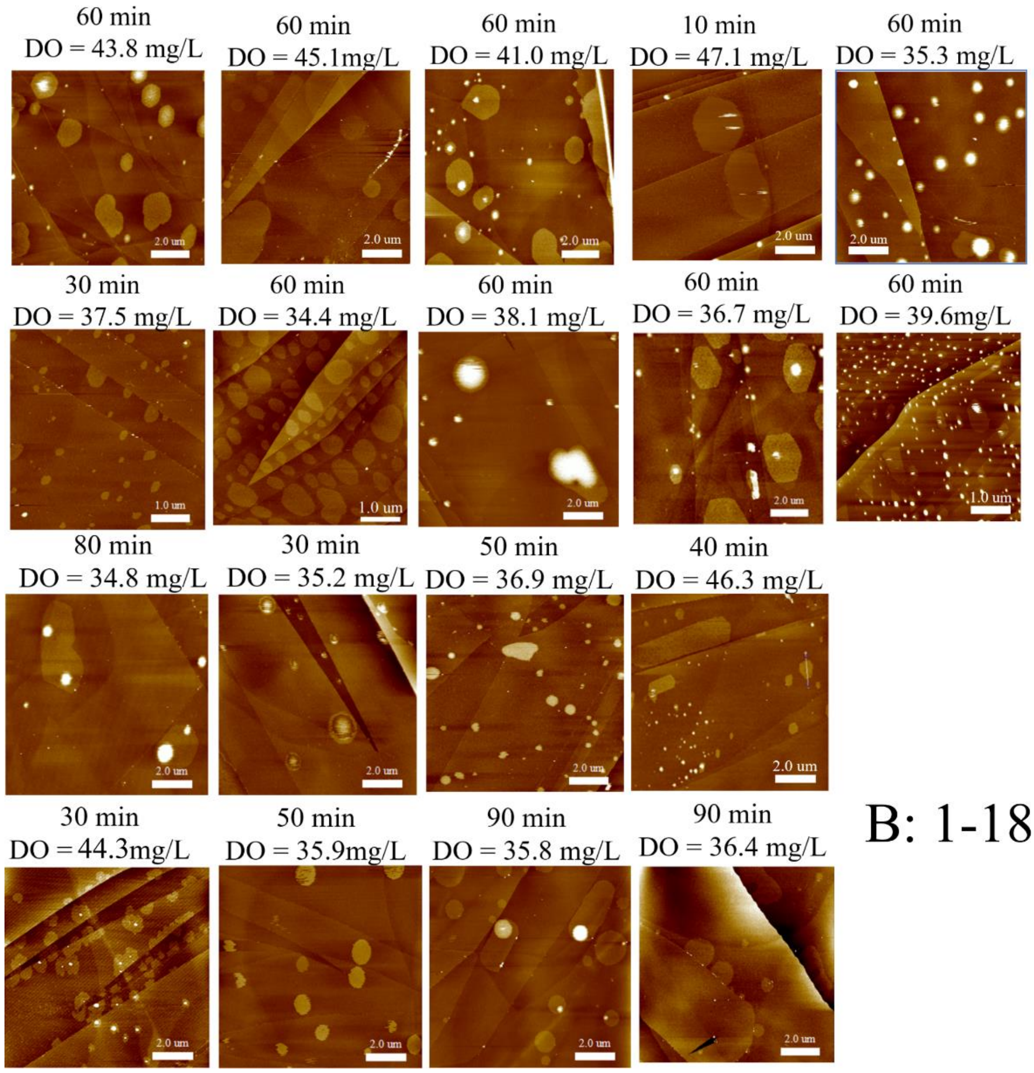


Figure S5. The 20 AFM height images of the interfacial gas domains produced by the new method (1-20) and the ethanol-water exchange (a-t) respectively. For the new method, only four planes (12, 15, 16, 20) observed the alone nanobubbles. For the solvent exchanges, five out of twenty images (a, j, p, r, t) measured the pancakes surrounded the nanobubbles.

3. The formation of gas domains under different oxygen contents in water and aging-time of surfaces



A: 1-15


60 min
DO = 43.8 mg/L
2.0 um
60 min
DO = 45.1mg/L
2.0 um
60 min
DO = 41.0 mg/L
2.0 um
10 min
DO = 47.1 mg/L
2.0 um
60 min
DO = 35.3 mg/L
2.0 um
30 min
DO = 37.5 mg/L
1.0 um
60 min
DO = 34.4 mg/L
1.0 um
60 min
DO = 38.1 mg/L
2.0 um
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DO = 36.7 mg/L
2.0 um
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DO = 39.6mg/L
1.0 um
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DO = 35.2 mg/L
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DO = 36.9 mg/L
2.0 um
40 min
DO = 46.3 mg/L
2.0 um
B: 1-18
30 min
DO = 44.3mg/L
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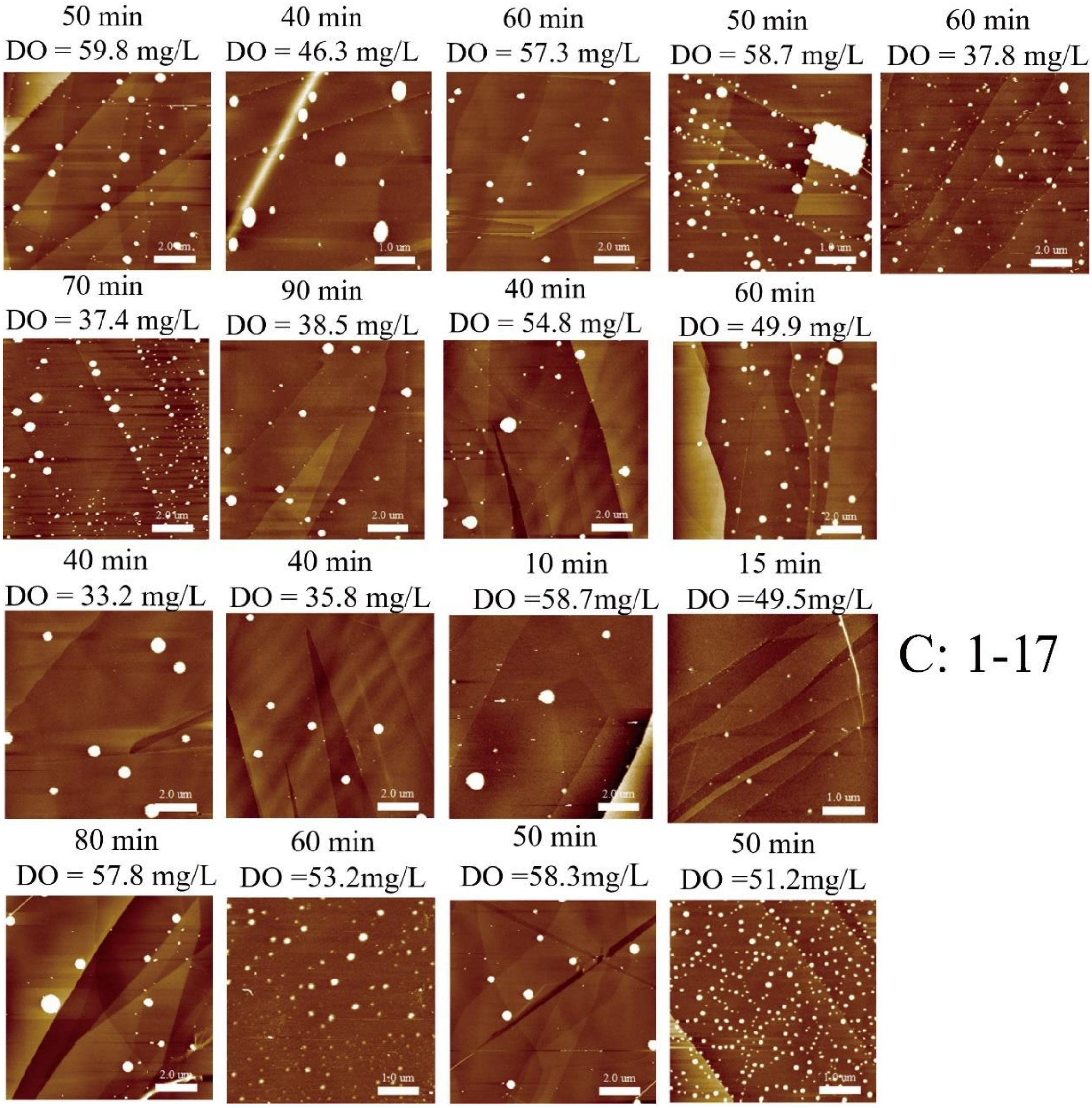

Figure S6. The 50 AFM images exhibited the formation states of gas domains under different DO values of water and aging time of surfaces. Images A 1-15 show the formation of interfacial gas phases with lower efficiency; B 1-18: micropancakes including individual and coexistence formed with the higher efficiency; C 1-17: only nanobubbles formation.