Supporting Information:

Rationally Designed, Multifunctional Self-Assembled Nanoparticles for Covalently Networked, Flexible and Self-Healable Superhydrophobic Composite Films

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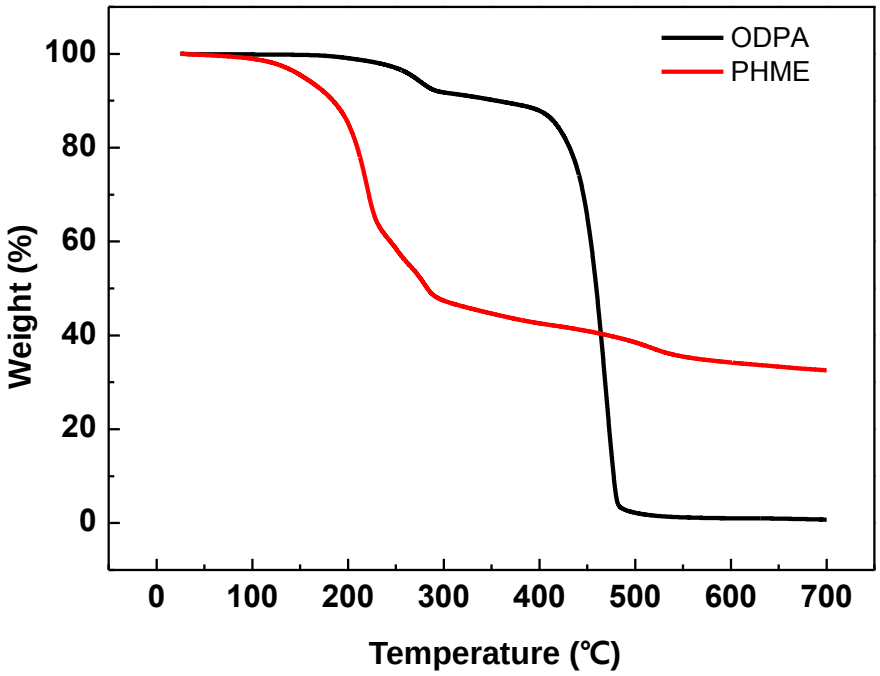


Figure S1. TGA data of ODPA and PHME molecules.

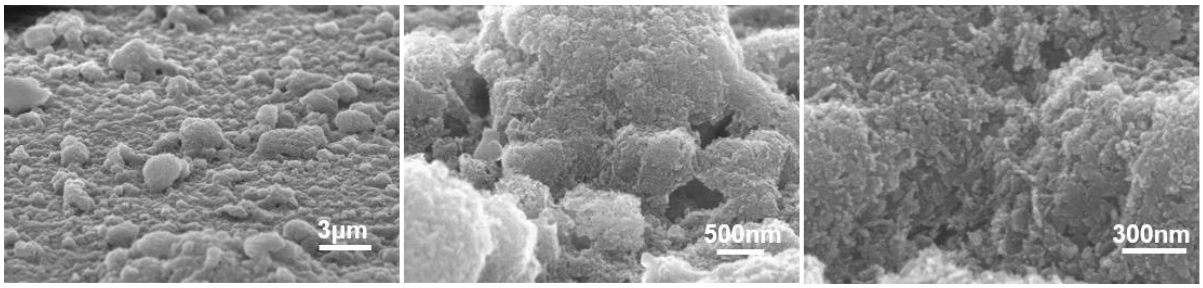
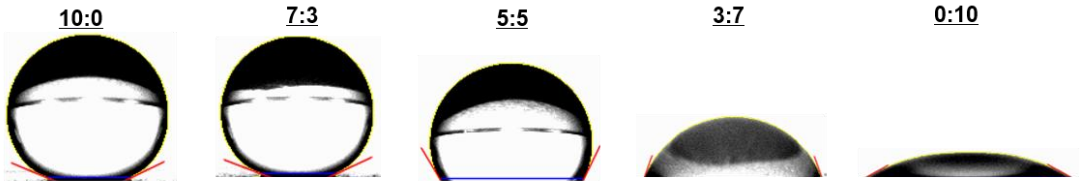
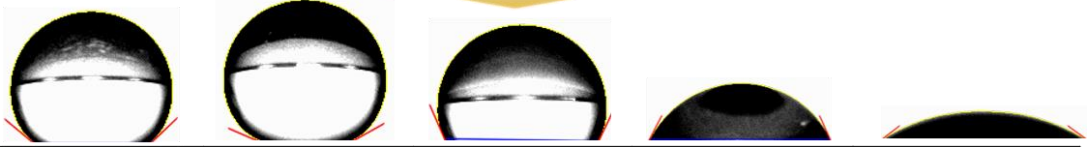


Figure S2. Tilted SEM images of the superhydrophobic composite film of multifunctional NPs with a representative SAM ratio of ODPA to PHME (7:3) in different magnifications. The tilted views at varied magnifications show the micro-sized papilla-like features formed from NPs, representing hierarchical micro/nano dual-scale structures. The samples were tilted at 15°.



	10:0	7:3	5:5	3:7	0:10
WCA (°)	156.3 (±0.5)	155.3 (±0.3)	118.2 (±0.4)	70.9 (±1.5)	29.1 (±1.5)
SA (°)	3	5	81	NA	NA

Sonication



	10:0	7:3	5:5	3:7	0:10
WCA (°)	137.8 (±4.0)	153.6 (±0.5)	116.8 (±0.6)	68.5 (±1.0)	29.5 (±1.5)
SA (°)	70	6	83	NA	NA

Figure S3. Contact angles and sliding angles of water droplets on the composite films of multifunctional NPs with various ratios of ODPA to PHME SAMs before and after ultrasonication treatment.

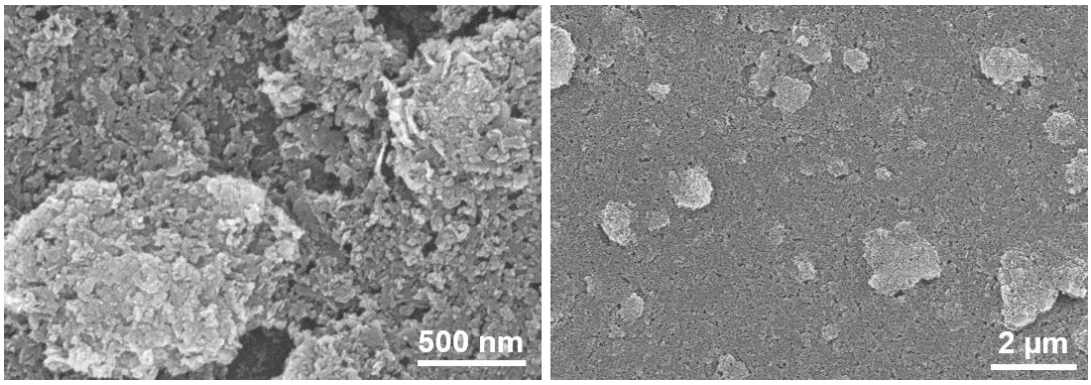


Figure S4. Top-down SEM images of the composite film of multifunctional NPs with an optimized SAM ratio of 7:3 after repeatedly bending 100 times. The SEM images, taken at different magnifications, show that the hierarchical micro/nano dual-scale surface roughness can be retained after the repeated bending process.

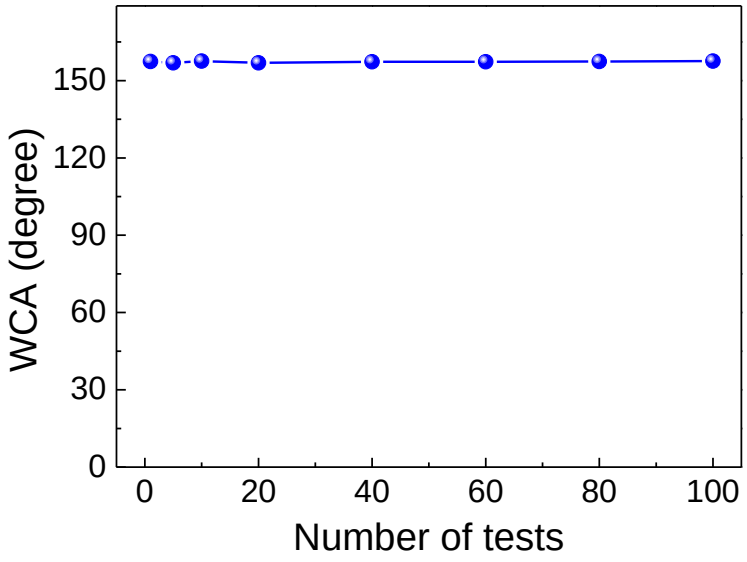


Figure S5. WCA of the hybrid composite film of multifunctional NPs with the SAM ratio of ODPA to PHME of 10:0 as a function of the number of bending tests for 100 cycles under a bending stress at the bending radius of 6.5 mm.

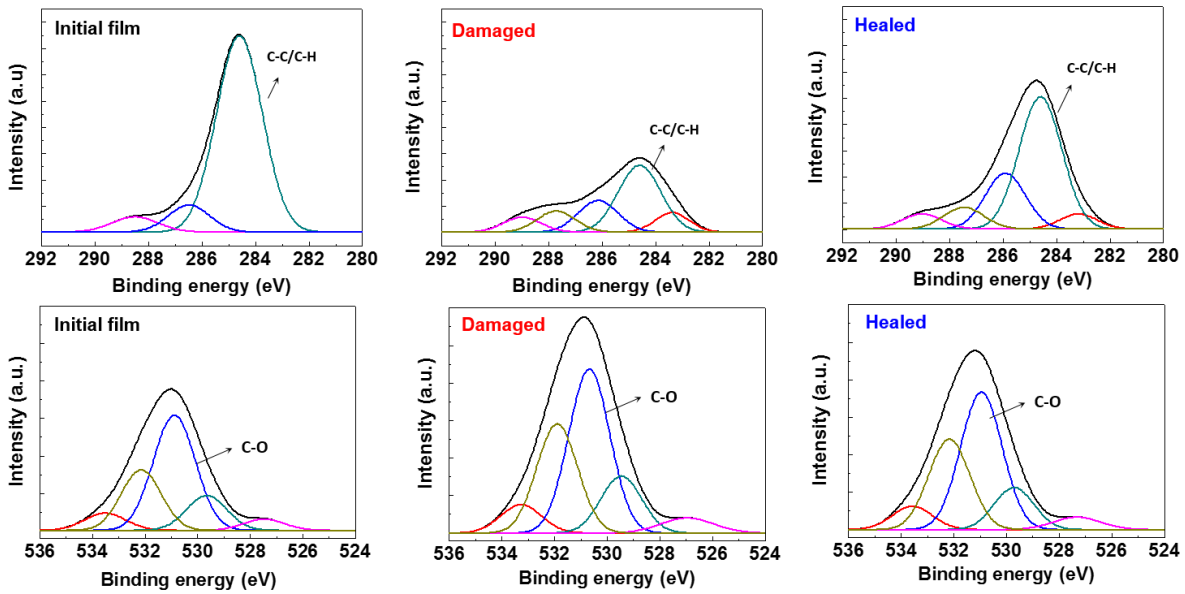


Figure S6. XPS spectra of the multifunctional NP-based superhydrophobic composite film (7:3) after UVO damage and subsequent self-healing. The composite film was treated with UVO (2 min) to induce chemical damage and then with heat (100 ℃, 30 min) to recover. After the heating process, the respective C-C/C-H and C-O peaks of the damaged film were restored to the initial superhydrophobic state.

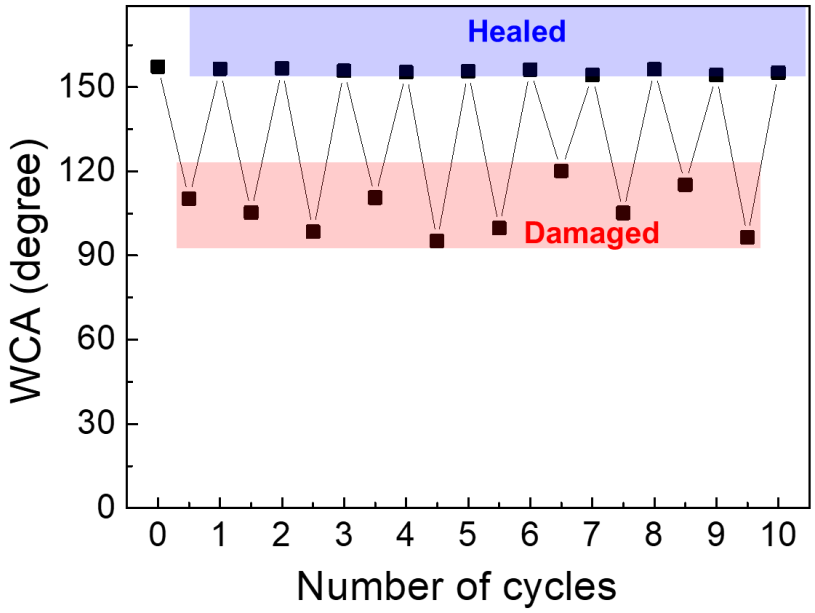


Figure S7. Change in WCAs of the hybrid composite film of multifunctional NPs with the SAM ratio of ODPA to PHME of 10:0 for 10 cycles of damage by immersion in a strong acid solution (6M HCl) and healing by heating.

Table S1. Comparison of the self-healing performance of the multifunctional NP-based superhydrophobic composite film with other self-healable superhydrophobic composite films.

Coating materials	Damage source	Initial WCA (°)	Damaged WCA (°)	Healed WCA (°)	# of cycles	Reference
Multifunctional NPs/HDDA	UV Ozone	>150	~ 0	>150	10	This study
	HCl (6M)	>150	93-107	>150	10	
PDA/PDMS	O_2 Plasma	>160	~ 50	>160	8	[63]
SPEEK/POTS	O_2 Plasma	>150	~ 0	>150	6	[64]
PAA/PAH–SPEEK	O_2 Plasma	>160	~ 0	>160	9	[65]
PS/SiO_2 NPs with PDMS	O_2 Plasma	>150	~ 0	>150	9	[66]
PDA@ODA–Fe_3O_4	O_2 Plasma	>150	~ 0	>150	10	[67]
PAL@fluoroPOS/PFDTES	O_2 Plasma	>150	~ 0	>150	5	[68]
DOPA–silica trimethylsilyl modified (DSTM) gel	HCl (1M)	>150	~ 127	>150	8	[69]
F-POSS/APP/bPEI	O_2 Plasma	>160	~ 0	>160	9	[70]