Supporting Information

Co-depositing mussel-inspired nanohybrids onto 1D fibers under "green" conditions for significantly enhanced surfacial/interfacial properties

Xiaobin Yang,[†] Hongpeng Du,[‡] Songwei Li,[†] Zhenxing Wang,[†] and Lu Shao[†]*

[†] *MIIT Key Laboratory of Critical Materials Technology for New Energy Conversion and Storage, State Key Laboratory of Urban Water Resource and Environment, School of Chemistry and Chemical Engineering, Harbin Institute of Technology, Harbin 150001, PR China.*

[‡] *Hangzhou Fortune Gas Cryogenic Group CO., LTD, Hangzhou 310000, China.*

*Corresponding Author at: Harbin Institute of Technology, No 92, West Dazhi Street, Harbin 150001, China. Email address: odysseynus@hotmail.com, shaolu@hit.edu.cn (L. Shao)

Tel. +86-451-86413711； fax:+86-451-86418270

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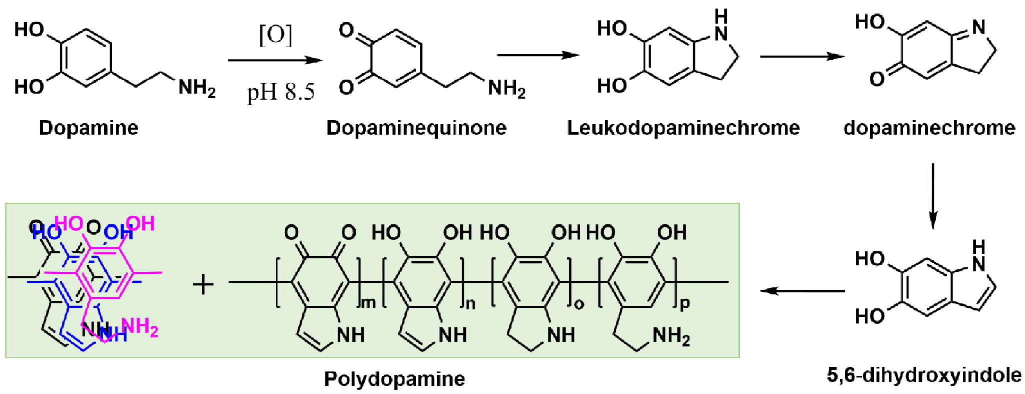

Figure S1. The typical polymerization mechanism of polydopamine (PDA).

As shown in **Figure S1**, dopamine first undergoes the initial oxidation to the corresponding quinone type and intramolecular cyclization via 1,4 Michael addition, resulting in leukodopaminechrome under alkaline conditions. Afterwards, it evolves into 5,6-dihydroxyindole (DHI) via a further oxidation and rearrangement, then DHI can be easily oxidized to 5,6-indolequinone. Afterwards, the synergetic effects combined DHI and its corresponding quinone type lead to the branching reactions, covalent binding among aryl rings, and the subsequent formation of multiple isomers of dimers, trimers and etc., resulting in the cross-linked polymer via the reverse dismutation transform between catechol and o-quinone species. The synergetic effects between the covalent bindings and noncovalent bindings (π–π stacking, hydrogen bonding, and charge transfer interactions) could result in adhesive deposition of PDA onto substrates.

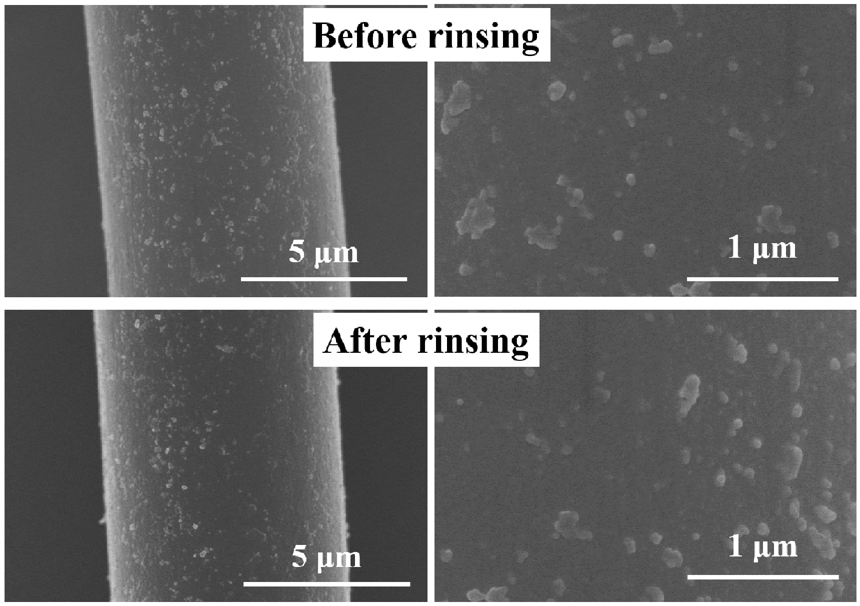

Figure S2. SEM images of the surfaces on CF-PDA/POSS before and after rinsing for about 3 h. As for rinsing test, CFs were fixed on the inner wall of a beaker by aluminum foil tape, then rinsing with water under high speed stirring (about 3000 r/min).

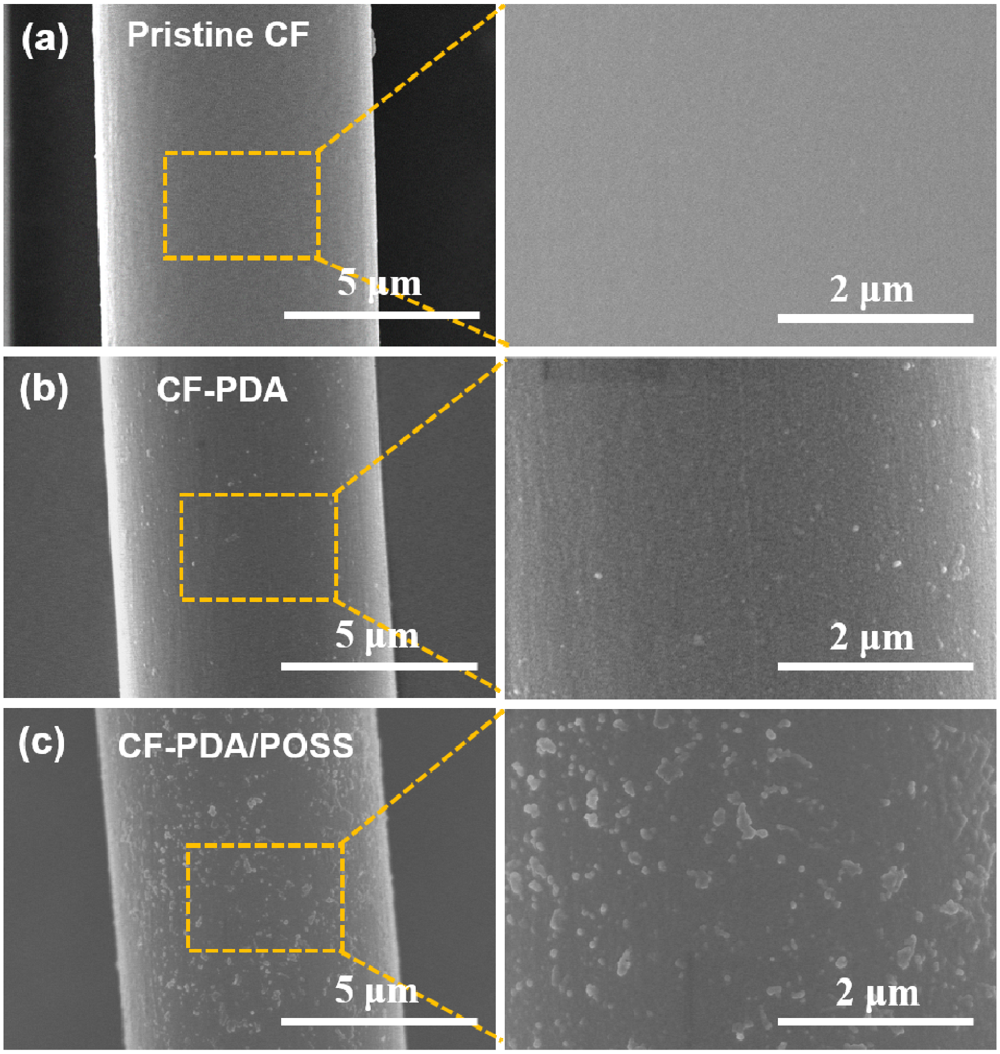

Figure S3. SEM images on the surfaces of CFs, including (a) pristine CFs, (b) CF-PDA, and (c) CF-PDA/POSS.

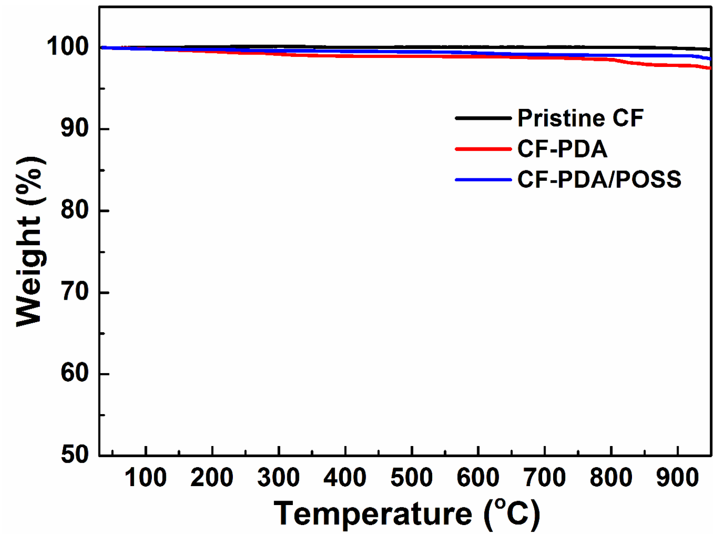

Figure S4. TGA traces of pristine CF (black line), CF-PDA (red line), and CF-PDA/POSS (blue line).

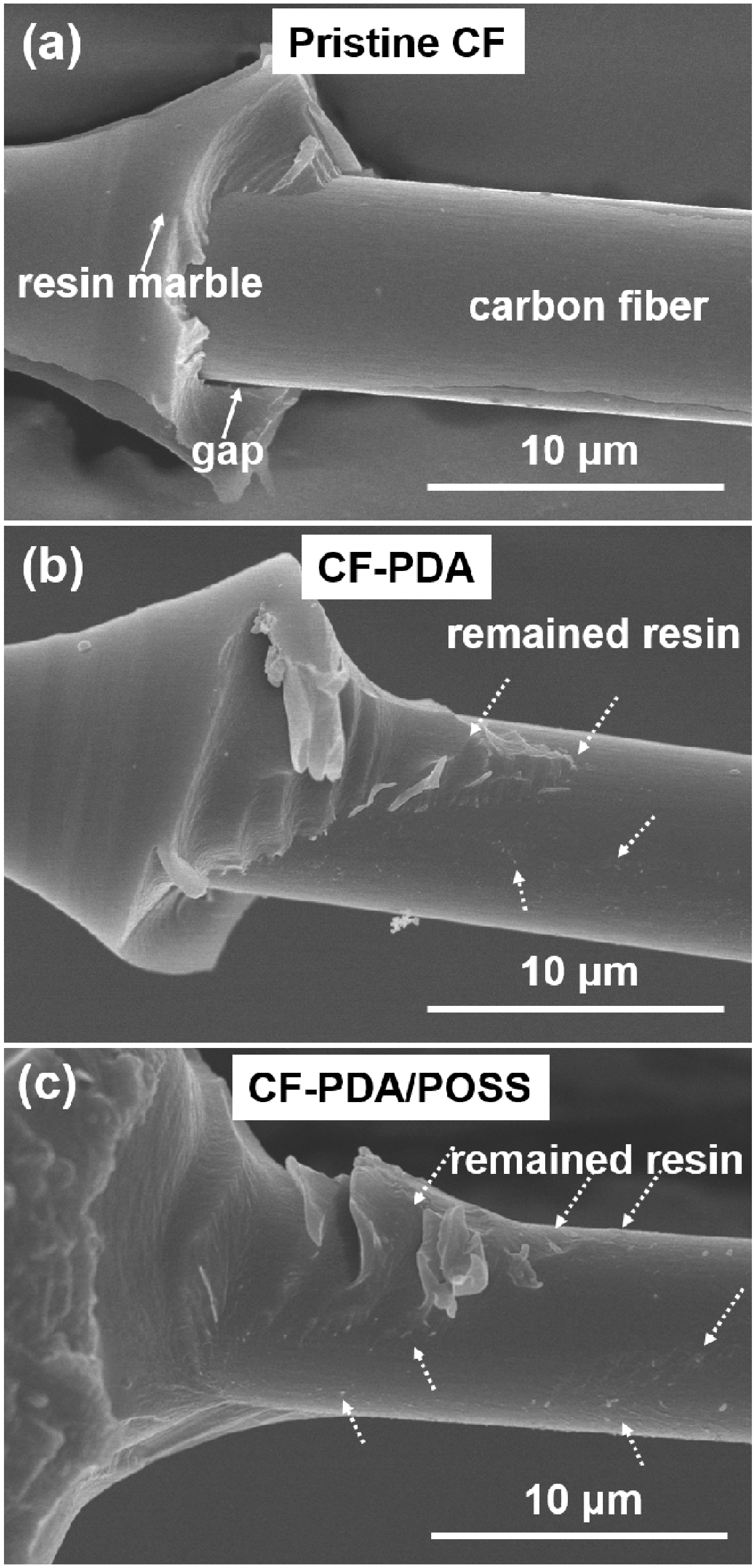
(a)
Pristine CF
resin marble
carbon fiber
gap
10 μm
(b)
CF-PDA
remained resin
10 μm
(c)
CF-PDA/POSS
remained resin
10 μm

Figure S5. SEM images of the samples (a) the pristine CF, (b) CF-PDA, and (c) CF-PDA/POSS after IFSS measurement through pulling out the cured resin marbles from the single filament. The exposed fiber surface of pristine CF was almost neat and there appeared gap between fiber and resin. There reamined few epoxy freagments on the surfaces of CF-PDA, more freagments on CF-PDA/POSS, simultanously showed benign connection between fiber and resin.

Table S1. Incubation conditions and maximum IFSS values about grafting POSS on the fiber surfaces.

Fiber types	Modification conditions	Topmost incubation temperature (^{o}C)	Number of incubation steps	Maximum IFSS (MPa)	References
CF	Strong acid, THF	80	3	80.6	1
CF	Strong acid, THF	80	4	95.9	2
CF	Strong acid, THF	78	3	88	3
CF	Strong acid, THF, toluene	120	5	-	4
CF	Strong acid, THF, DMF, $SOCl_2$	80	4	-	5
CF	Strong acid, THF, DMF, methanol	80	5	106.48	6
PBO	Strong acid, THF	150	4	74.1	7
PBO	Plasma, THF	80	2	54.9	8
CF	Strong acid, DMF, $SOCl_2$	78	4	-	9
CF	Strong acid, NMP, $SOCl_2$	80	4	-	10
CF	Strong acid, methylbezene	110	4	93	11
CF	Strong acid, acetone	78	4	~90	12
CNF	Strong acid, THF, DMAc, catalyst, pre-modified fibers	100	2	-	13
CF	**Aqueous system**	**Room temperature**	**1**	**117.1**	**This work**

*The abbreviations of "CF, PBO, CNF, THF, DMF, NMP, DMAc, $SOCl_2$, and IFSS" refer to carbon fiber, poly (*p*-phenylene benzobisoxazole), carbon nano-fiber, tetrahydrofuran, dimethyl formamide, N-methyl pyrrolidone, dimethylacetamide, sulfoxide chloride, and interfacial shear strength, respectively. The reagents of THF, DMF, NMP, DMAc, $SOCl_2$ are organic solvents.

As shown in **Table S1**, we could find that the modification methods towards fibers with POSS are either involved with strong acids, organic reagents, the high energy consumption, or the multi-step manipulation. However, our strategy is to co-deposit mussel-inspired nanohybrid coating onto carbon fiber (CF) surface *via* co-incubation in the dopamine (DA) and octaammonium polyhedral oligomeric silsesquioxane (POSS-NH_2) mixed aqueous solution. The deposition procedure was conducted under ambient conditions, and no harsh conditions and toxic solvents were needed. Thus, our reported facile strategy presents a eco-friendly and promising platform to modify 1D fiber surface for advanced composite materials.

Table S2. Surface energy evaluation of pristine CF, CF-PDA, CF-PDA/POSS.

Samples	Surface energy (mN m^{-1})		
	γ_f^p	γ_f^d	γ_f
pristine CF	38.7	1.3	40.0
CF-PDA	50.9	2.5	53.4
CF-PDA/POSS	65.7	0.4	66.1

Table S3. IFSS of CF/epoxy resin composites.

CF Type	Epoxy Type	Curing Agent	Maximum IFSS (MPa)	References
T700	WSR618 (E-51)	Methyl Tetrahyelrophthalic Anhydride	80.6	1
T700	WSR618 (E-51)	Methyl Tetrahyelrophthalic Anhydride	95.9	2
JT-400A-3K	WSR618 (E-51)	H-256	106.48	6
T700	WSR618 (E-51)	Methyl Tetrahyelrophthalic Anhydride	78.6	14
T700	E-20, E-54	DDM, DDS, 2-Ethyl-4-methylimidazole	98.1	15
T700	YD-128	EC301	57.2	16
T700	E-51	T-31	52.3	17
T700	E-5228	-	76	18
T700	HS5382	Tetrahydrophthalic Anhydride	64.8	19
T700	EPON 828	m-phenylenediamine	81	20
T300	EPON 618, 650	DBP	91	21
T300	-	-	87.77	22
T700	-	-	89.39	22
T600	DW2	Cycloaliphatic Amine	60.26	23
T700	DW2	Cycloaliphatic Amine	67.53	23
STS5631	DW2	Cycloaliphatic Amine	80.09	23
-	E-51	H-256	118.5	24
-	E-51	H-256	118.7	25
T300	AG-80	p-diaminodiphenylsulfone	106.55	26
C320.00A	HexPly 8552	-	~115	27
-	E-51	-	113	28
T700	E-51	H-256	103	29
T700	E-51	H-256	117.1	This work

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