Supporting Information

Enhancing the Efficiency of PVDF-Based Piezoelectric Catalysis through Water-Induced Polarization and Micro-Nano Composite Strategy

Haitao Li,[a] Yingying Zhang,[a,b] Han Dai,[a] Veronica Pereira,[c] Junfeng Zhao*[a] and Hiang Kwee Lee*[c, d]*

[a]*Laboratory of Advanced Light Alloy Materials and Devices, Postdoctoral Workstation of Nanshan Group Co., Ltd., Yantai Nanshan University, Longkou 265713, China.*

[b]*School of Chemistry and Chemical Engineering, Yangzhou University, Yangzhou, 225002, PR China.*

[c]*Division of Chemistry and Biological Chemistry, School of Chemistry, Chemical Engineering and Biotechnology, Nanyang Technological University, 21 Nanyang Link, 637371 Singapore*

[d]*Institute of Materials Research and Engineering, The Agency for Science, Technology and Research (A*STAR), 2 Fusionopolis Way, #08-03, Innovis 138634, Singapore*

Corresponding Author

E-mail address: htli@yzu.edu.cn (Haitao Li); zhaojunfengcc@163.com (Junfeng Zhao); hiangkwee@ntu.edu.sg (Hiang Kwee Lee)

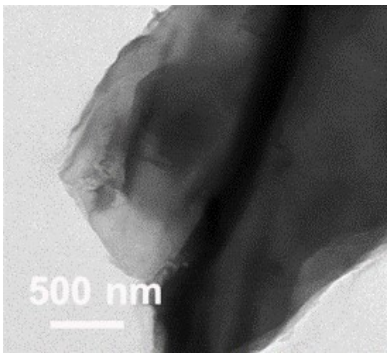

Figure S1. TEM image of rGO.

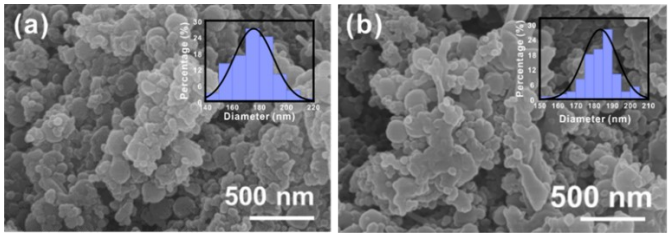

Figure S2. SEM images of (a) self-polarized PVDF (SP) and (b) self-polarized rGO/PVDF (SP/rGO).

The calculation of β phase content and related crystallinity

(1) The calculation of β phase: FTIR absorption was assumed to obey the Lambert-Beer law, the amount of the β-phase (F(β)) of PVDF was calculated using Eq (S1)[1].

$$F(\beta) = \frac{A_\beta}{1.26A_\alpha + A_\beta} \times 100\ \% \quad \text{(S1)}$$

Here, A_α and A_β, respectively, correspond to the absorbance of the α (762 cm^{-1}) and β phases (839 cm^{-1}) of PVDF. The β phase content of original PVDF, SP and SP/$rGO_{1.5}$ are 14%, 91%, 95%, respectively.

(2) The calculation of related crystallinity： The crystallinities of original PVDF, SP and SP/$rGO_{1.5}$ are calculated according to their temperature increase curves using Eq. (S2)[2].

$$X_C = \frac{\Delta H_f}{\Delta H_f^* \cdot \varphi} \times 100\ \% \quad \text{(S2)}$$

Where ΔH_f is the sample enthalpy of fusion, calculated from heating DSC curve, ΔH^*_f is the heat of fusion of perfectly crystalline PVDF from literature (104.7 J g^{-1}) and ϕ is the weight fraction of PVDF in the samples. The crystallinity for original PVDF, SCP_0 and $SCP_{1.5}$ are 23 %, 37%, 40%, respectively.

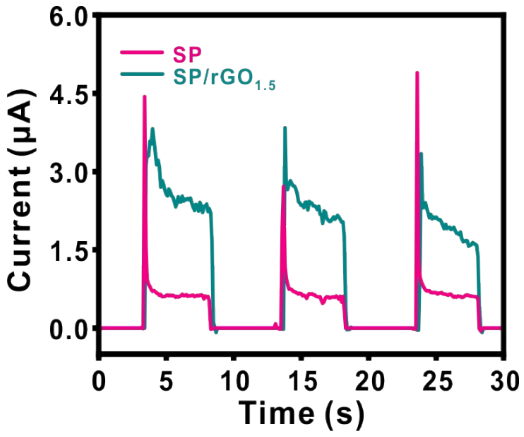

Figure S3. Piezoelectric current of (a) SP and (b) SP/$rGO_{1.5}$.

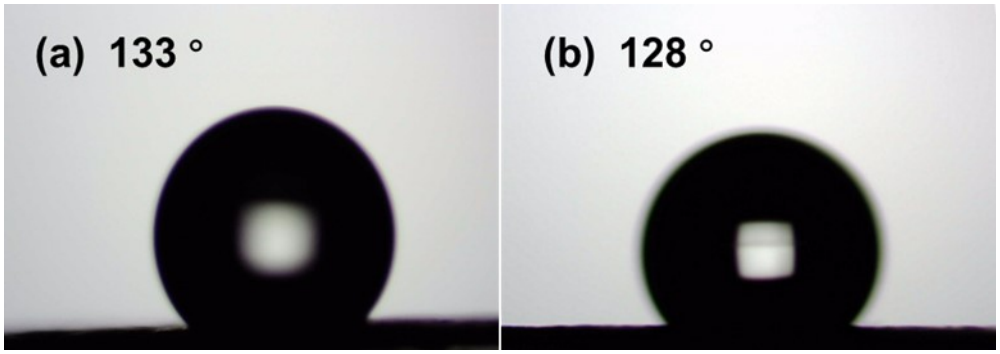

Figure S4. Contact angle images of a sessile water droplet on (a) SP and (b) SP/$rGO_{1.5}$.

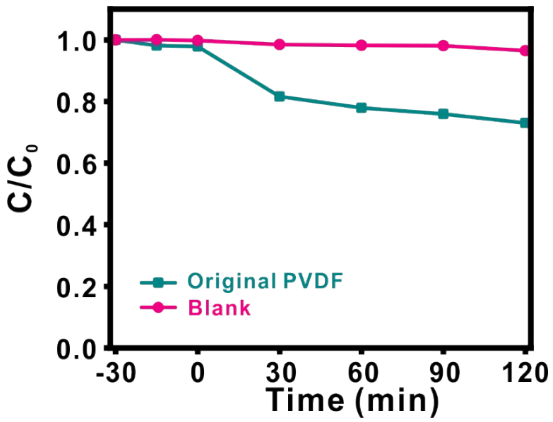

Figure S5. Piezocatalytic degradation of 15 mL RhB solution (100 mg L^{-1}， pH=7, 30 ℃) under 240 W ultrasonication. Comparison of normalized absorbance between 15 mg pristine PVDF and blank control.

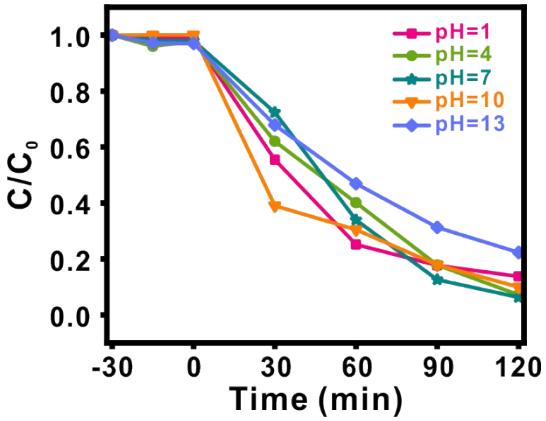

Figure S6. Time-dependent plot of normalized RhB concentration (15 mL, 100 mg L^{-1}; 30 ℃) under 240 W ultrasonication at different pH environment (1 - 13) and in the presence of SP/$rGO_{1.5}$ (15 mg).

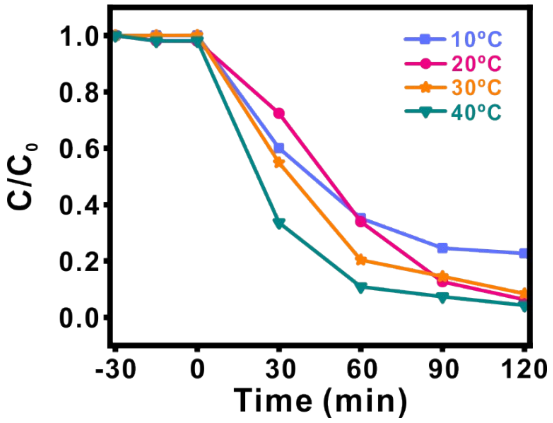

Figure S7. Time-dependent plot of normalized RhB concentration (15 mL; pH =7; 30 ℃) under 240 W ultrasonication at different reaction temperature (10 - 40℃) and in presence of SP/$rGO_{1.5}$ (15 mg).

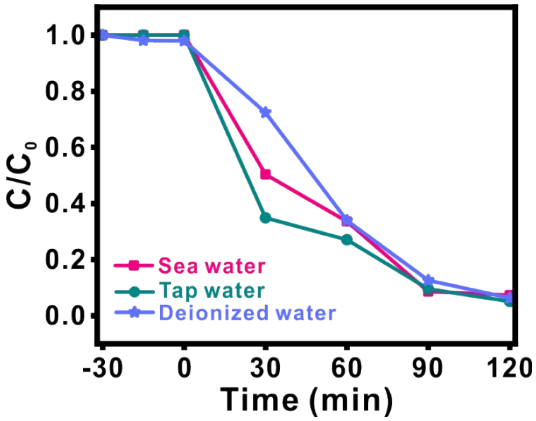

Figure S8. Time-dependent plot of normalized RhB concentration (15 mL; pH =7; 30 ℃) under 240 W ultrasonication when using different water samples (DI water, tap water and sea water) and in the presence of SP/r$GO_{1.5}$ (15 mg).

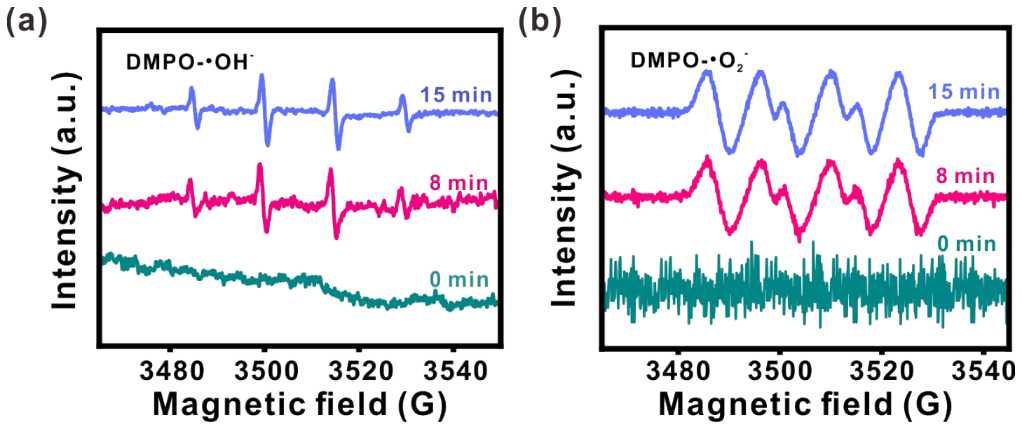

Figure S9. (a) DMPO-·OH and (b) DMPO-·O_2- obtained from SP/r$GO_{1.5}$ with different ultrasonication time.

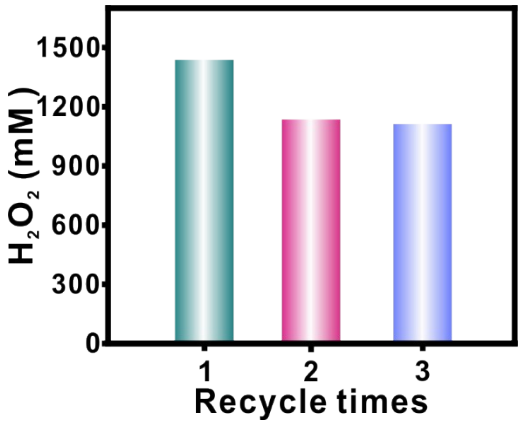

Figure S10. Comparison of H_2O_2 production rate by SP/$rGO_{1.5}$ over three successive cycles.

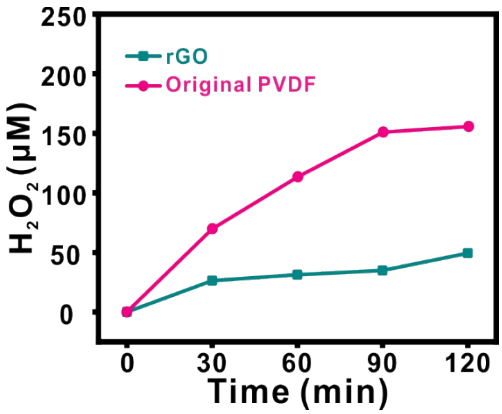

Figure S11. Time-dependent piezocatalytic H_2O_2 productions by rGO and pristine PVDF under 300 W ultrasonication.

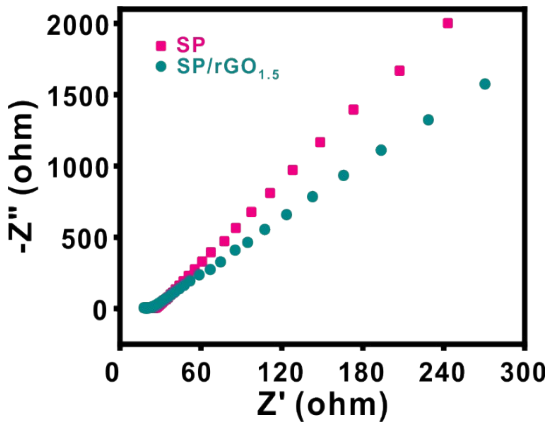

Figure S12. Electrochemical impedance spectroscopy characterization of SP and SP/$rGO_{1.5}$.

Table S1. Comparison of the organic degradation and H_2O_2 production by SP/$rGO_{1.5}$ with other previously reported piezoelectric catalysts.

Material of catalyst, Synthesis method	Content of catalyst	Reaction solution	Frequency and power of ultrasound	Time, min	Degradation ratio, %	Generating rate of H_2O_2, mmol g_{cat}^{-1} h^{-1}
Nano kaolinite-MWCNT/PVDF film[3] one-step solution casting method	1x1cm	15 mL, 2.5 ppm RhB	33 kHz, 50 W	45	~96, 99	
Ag@$LiNbO_3$/ PVDF[4] The solvent casting method	d=2.5 cm	10 mL, 10 mg/L MB	40 kHz, 70 W	180	~89	
αFe_2O_3/PVDF[5] electrospinning	7 mg	20 mL, 10 mg/L MB	18 kHz, 250 W	60	~60.4	
BNT/PVDF[6], a "sol-gel-elec-trospinning" method	100 mg	100 mL, 50 mg/L RhB	45 kHz, 200 W	180	~76.6	
BNBT-x[7], one-step sol-vothermal method.	100 mg	50 mL, 10 mg/L RhB	18 kHz, 250 W	60	~73	
PCN/PVDF-HFP[8]	50 mg	8 mL H_2O containing 2mL EtOH	40 kHz, 300 W	120		0.668
SiO_2/PVDF-HFP[9]	50 mg	8 mL H_2O containing 2mL EtOH	40 kHz, 300 W	60		0.492
CNT/PVDF[10] phase separation by nanosization	15 mg	15 mL, 40 mg L^{-1} RhB	40 kHz, 240 W	120	~98	
	25 mg	8 mL H_2O containing 2mL EtOH	40 kHz, 300 W	120		13.51
rGO/PVDF phase separation by nanosization	15 mg	15 mL, 100 mg L^{-1} RhB	40 kHz, 240 W	120	~93.7	
	5 mg	8 mL H_2O containing 2mL EtOH	40 kHz, 300 W	120		95.8

Note and references

1. *S. Huang, S. Hong, Y. Su, Y. Jiang, S. Fukushima, T. M. Gill, N. E. D. Yilmaz, S. Tiwari, K.-i. Nomura, R. K. Kalia and Flame, Combust Flame, 2020,* ***219****, 467-477.*
2. *C. Merlini, G. Barra, T. M. Araujo and A. Pegoretti, RSC Adv, 2014,* ***4****, 15749-15758.*
3. *D. Mondal, S. Bardhan, N. Das, J. Roy, S. Ghosh, A. Maity, S. Roy, R. Basu and S. Das, Nano Energy, 2022,* ***104****.*
4. *G. Singh, M. Sharma and R. Vaish, ACS Appl Mater Interfaces, 2021,* ***13****, 22914-22925.*
5. *F. Orudzhev, S. Ramazanov, D. Sobola, P. Kaspar, T. Trčka, K. Částková, J. Kastyl, I. Zvereva, C. Wang, D. Selimov, R. Gulakhmedov, M. Abdurakhmanov, A. Shuaibov and M. Kadiev, Nano Energy, 2021,* ***90****.*
6. *X. Zhou, Q. Sun, Z. Xiao, H. Luo and D. Zhang, J. Environ, 2022,* ***10****.*
7. *L. Jiang, N. Xie, Y. Hou, H. Fu, J. Zhang, H. Gao and Y. Liao, Catal Commun, 2023,* ***181****, 106735.*
8. *Z. Chen, J. Zhuang, C. Liu, M. Chai, S. Zhang, K. Teng, T. Cao, Y. Zhang, Y. Hu, L. Zhao and Q. An, ChemElectroChem, 2022,* ***9****.*
9. *L. Wang, Z. Chen, Y. Zhang, C. Liu, J. Yuan, Y. Liu, W. Ge, S. Lin, Q. An and Z. Feng, Chem Asian J, 2022,* ***17****, e202200278.*
10. *Y. Zhang, C. Chong, W. Tong, H. Li, H. K. Lee and J. Han, Colloids and Surfaces A: Physicochemical and Engineering Aspects, 2023,* ***677****.*