Supporting Information

Two-step gas expansion and liquid phase exfoliation strategy for production of N-doped defect-rich transition metal dichalcogenides nanosheets and their antibacterial applications

Tao Wang,[a,b] Xiao Zhang,[b] Linqiang Mei,[b,c] Dongqing Ma,[b] You Liao,[b] Yan Zu,[b] Peng Xu,[b] Wenyan Yin,[b,*] Zhanjun Gu,[b,c,*]

[a]College of Materials and Chemistry and Chemical Engineering, Chengdu University of Technology, Chengdu 610059, China.

[b]Key Laboratory for Biomedical Effects of Nanomaterials and Nanosafety, Institute of High Energy Physics and CAS Center for Excellence in Nanoscience National Center for Nanoscience and Technology of China, Chinese Academy of Sciences, Beijing 100049, China.

[c]Center of Materials Science and Optoelectronics Engineering, College of Materials Science and Optoelectronic Technology, University of Chinese Academy of Sciences, Beijing 100049, China

E-mail: yinwy@ihep.ac.cn; zjgu@ihep.ac.cn

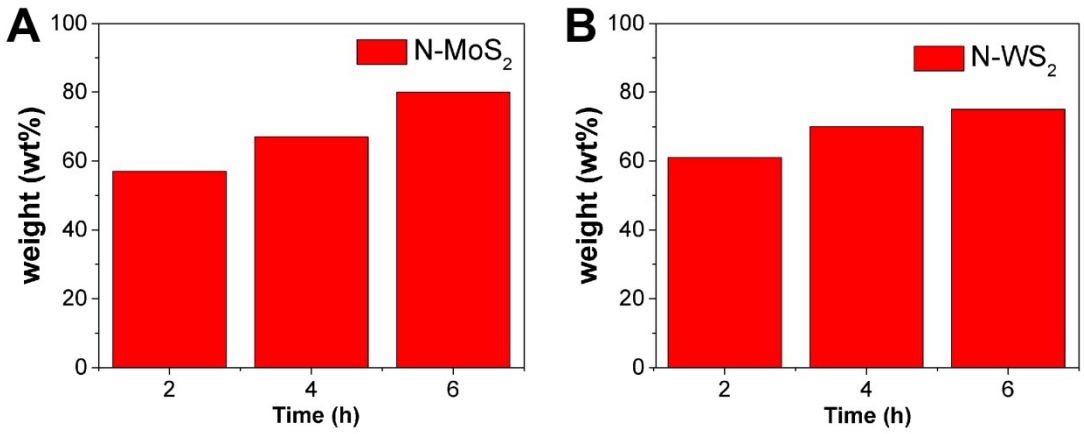

Figure S1. The comparisons of yields of defect-rich (A) N-MoS_2 and (B) N-WS_2 NSs after probe-ultrasonication for 2, 4, and 6 h.

Table S1 Summary the yield of MoS_2 NSs synthesized by liquid-phase exfoliation methods.

Synthetic method	Sonication time	Maximum yield	Ref.
Liquid-mediated exfoliation (sonication in NMP)	2 h	14%	1-4
	12 h	2.4%	
	60 h	8%	
	140 h	40%	
Liquid-mediated exfoliation (sonication in 45% EtOH/H_2O)	8 h	< 1%	5
Liquid-mediated exfoliation (high-shear exfoliation)	1 mg/min	1%	6
BSA-induced liquid exfoliation	48 h	27.2%	7
Salt-assisted ball-milling and sonication-assisted solvent exfoliation	-	25.5%	8
Unimer-Assisted Exfoliation	1 h	4.51%	9
Liquid-mediated exfoliation (sonication in sodium cholate (SC))	16 h	17%	10
Liquid-mediated exfoliation (the assistance of NaOH in NMP)	2 h	65%	11
Liquid-mediated exfoliation (oleum treatment, adding CS sonication)	2 h	62%	12

Table S2 Summary the yield of WS_2 NSs synthesized by liquid-phase exfoliation methods.

Synthetic method	Sonication time	Maximum yield	Ref.
Liquid-mediated exfoliation (iterative centrifugation cascades)	-	75%	13
Liquid-mediated exfoliation (microwave-assisted)	30 min	50%	14
Liquid-mediated exfoliation (by a modified LPE method)	2 h	22%	15
Liquid-mediated exfoliation (rapid quenching method)	20 min	>60%	16
Liquid-mediated exfoliation (high-boiling-point solvents)	24 h	2.7%	17
Liquid-mediated exfoliation (high-shear exfoliation)	3 h	<1%	18
Liquid-mediated exfoliation (polar micromolecular solvent)	48 h	18%	19

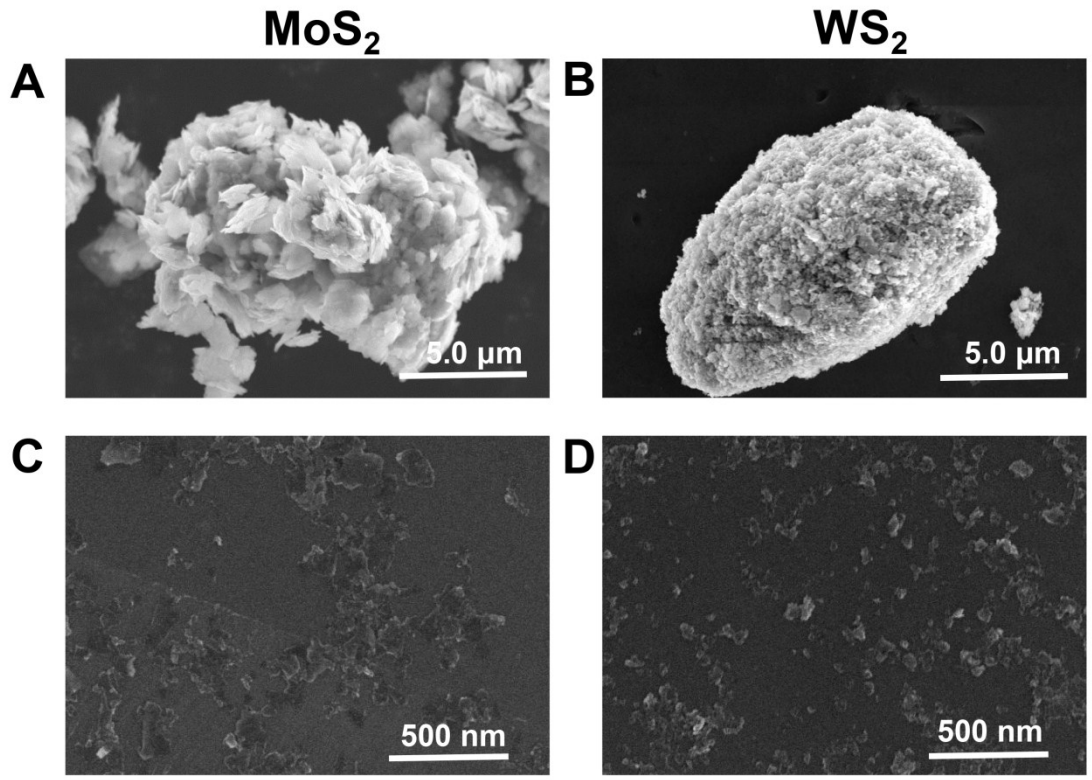

Figure S2. FE-SEM images of (A, B) bulk MoS_2 and bulk WS_2, (C, D) N-MoS_2 and N-WS_2 NSs.

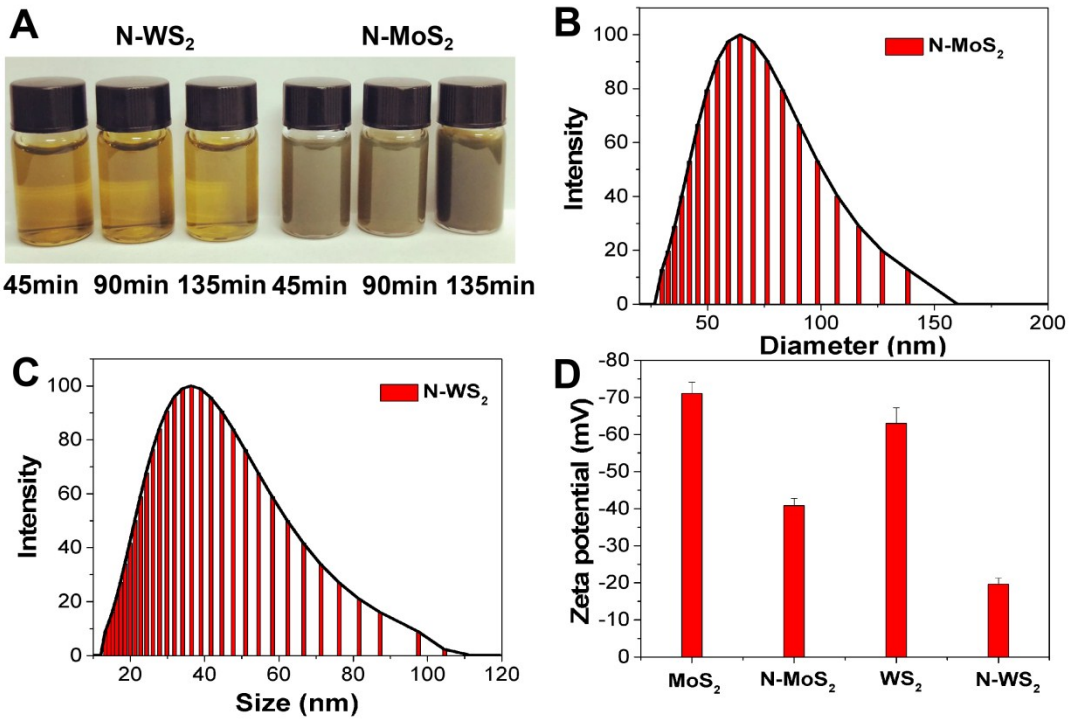

Figure S3. (A) Photographs of defect-rich N-MoS_2 and N-WS_2 NSs dispersed in water for a week after different exfoliation times. (B, C) Size distributions of N-MoS_2 and N-WS_2 NSs. (D) Comparison of zeta potential of MoS_2 and WS_2 NSs without/with N-doping. Concentration: N-MoS_2 (80 μg/mL), N-WS_2 (100 μg/mL).

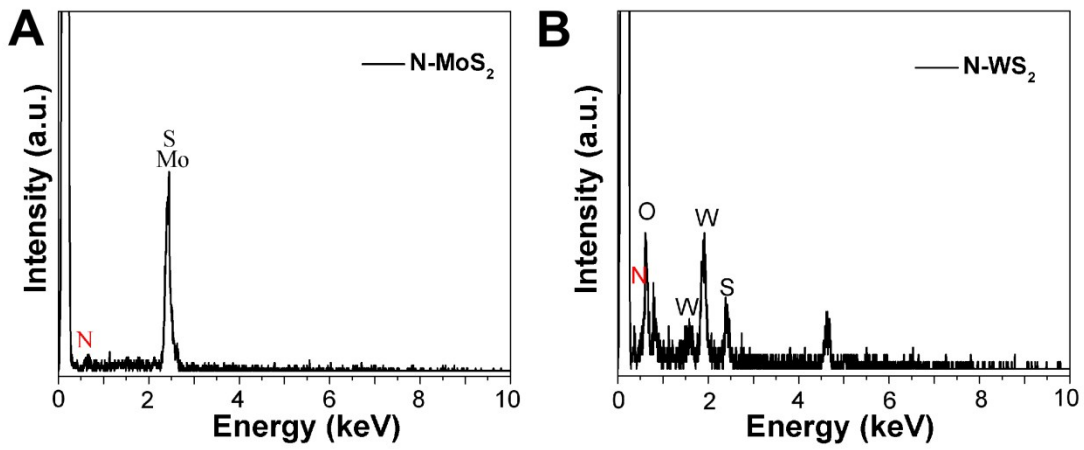

Figure S4. EDS of (A) N-MoS_2 and (B) N-WS_2 NSs.

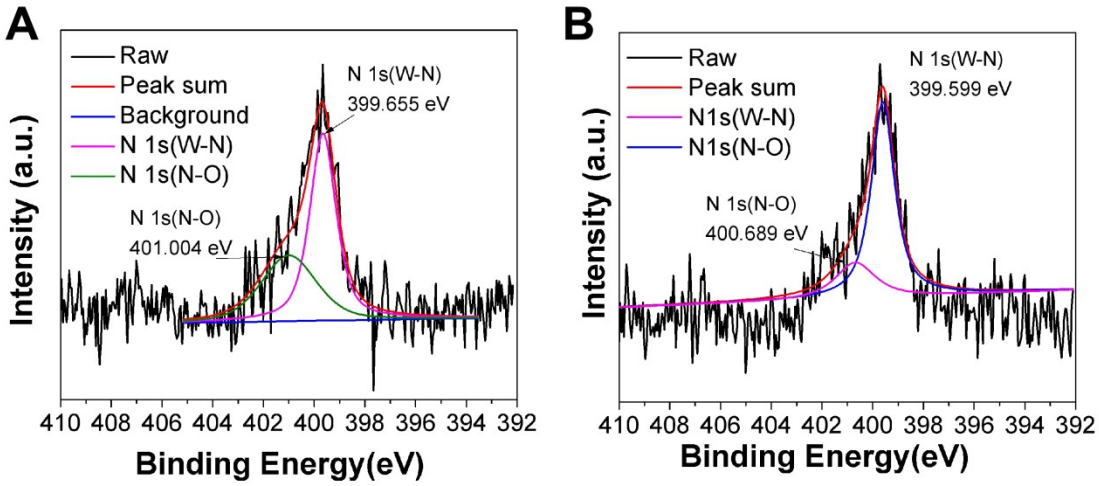

Figure S5. XPS of (A) N-WS_2 and (B) N-WS_2 obtained by using 200 mg and 400 mg of urea as N source. The percentage content (wt.%) of N in N-WS_2 was 0.75 %, 1.7 % respectively.

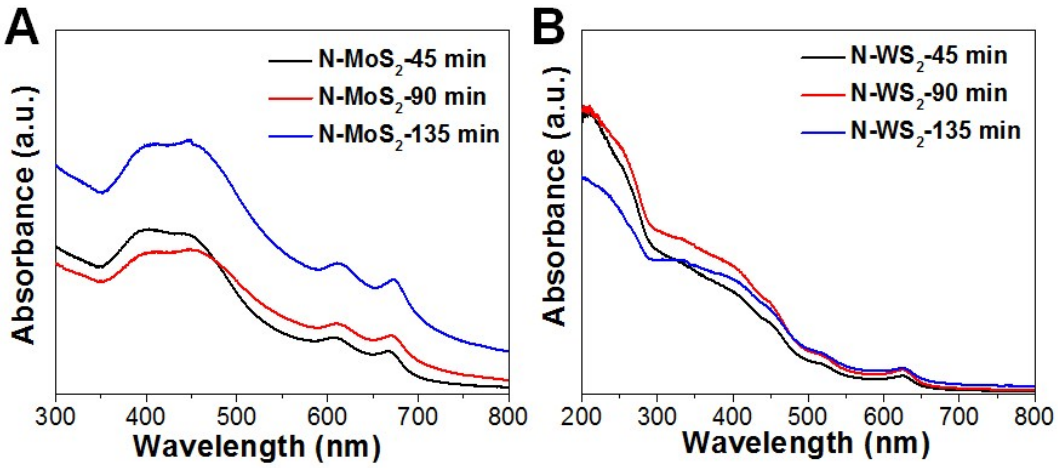

Figure S6. UV-vis-NIR absorption spectra of (A) defect-rich N-MoS_2, (B) defect-rich N-WS_2 NSs with different intercalation times.

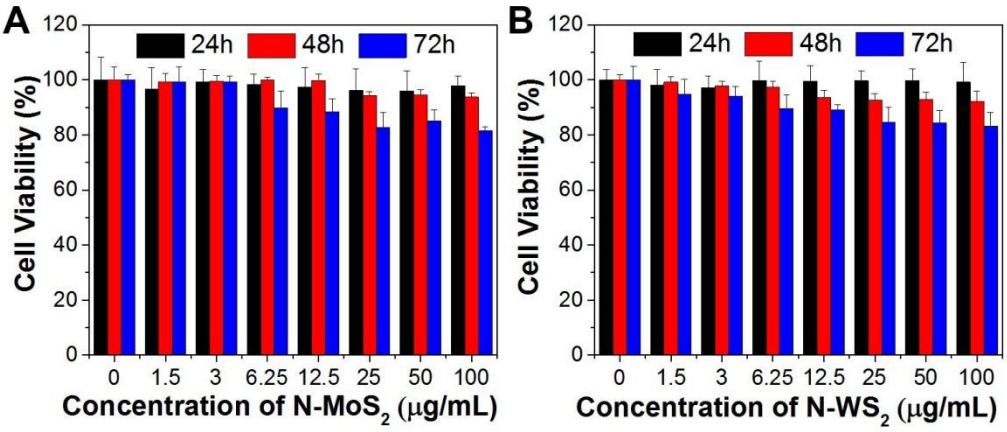

Figure S7. Cytotoxicity of different concentrations of (A) N-MoS_2 and (B) N-WS_2 NSs incubated with HUVEC cells for 24 h, 48 h, and 72 h.

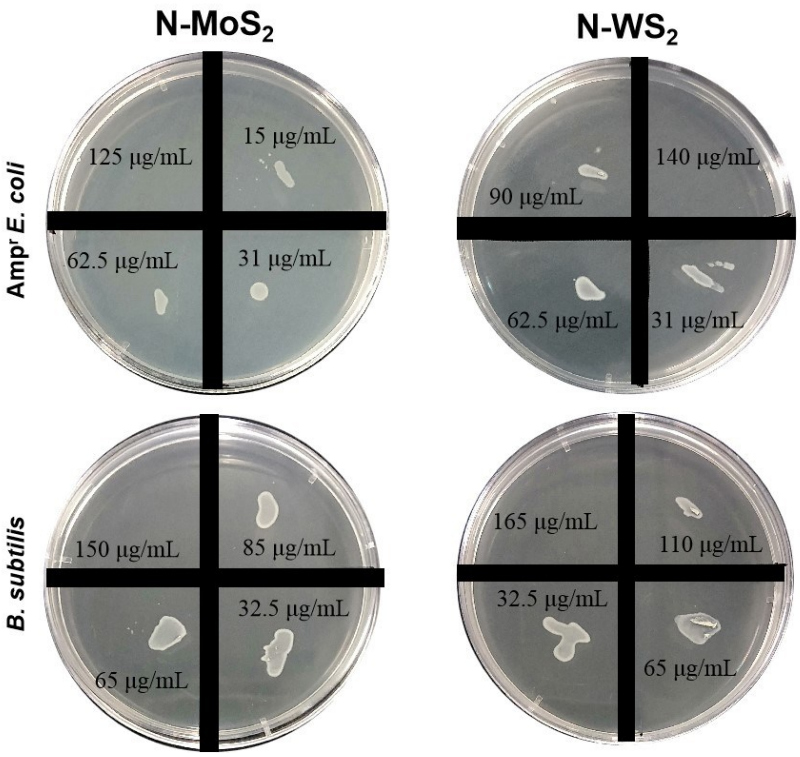

Figure S8. Plate images of Amp^r *E. coli* and *B. subtilis* after treating with different N-MoS_2, N-WS_2 NSs.

Table S3.MIC_{50} and MBC of N-MoS_2 and N-WS_2 against Amp^r *E. coli* and *B.subtilis.*

	Amp^r E. coli MIC_{50}	*Amp^r E. coli* MBC	*B. subtilis* MIC_{50}	*B. subtilis* MBC
N-MoS_2	19.99 μg/mL	125 μg/mL	24.3 μg/mL	150 μg/mL
N-WS_2	42.3 μg/mL	140 μg/mL	47.8 μg/mL	165 μg/mL

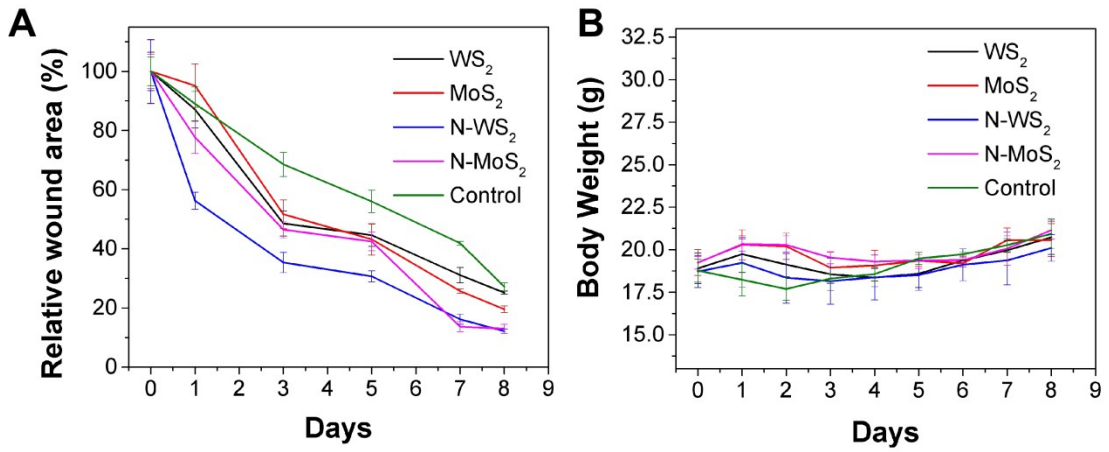

Figure S9. (A) Graphical representation of relative area of wound during 8 days. (B) Changes in mice body weight during 8 days of treatment.

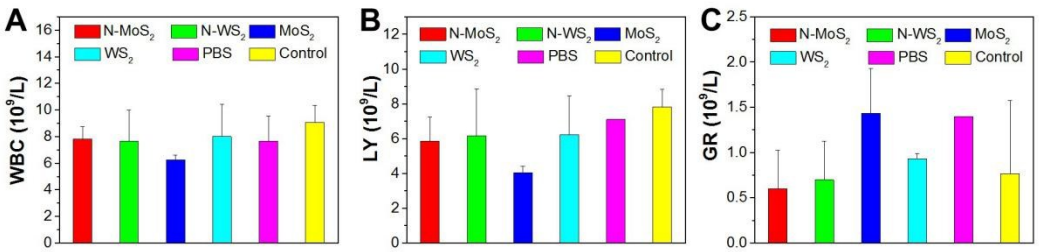

Figure S10. (A) white blood cell (WBC), (B) lymphocytes (LY), and (C) granulocytes (GR) of mice wound treating by WS_2, MoS_2, N-WS_2, N-MoS_2, PBS, and control without wound on day 8.

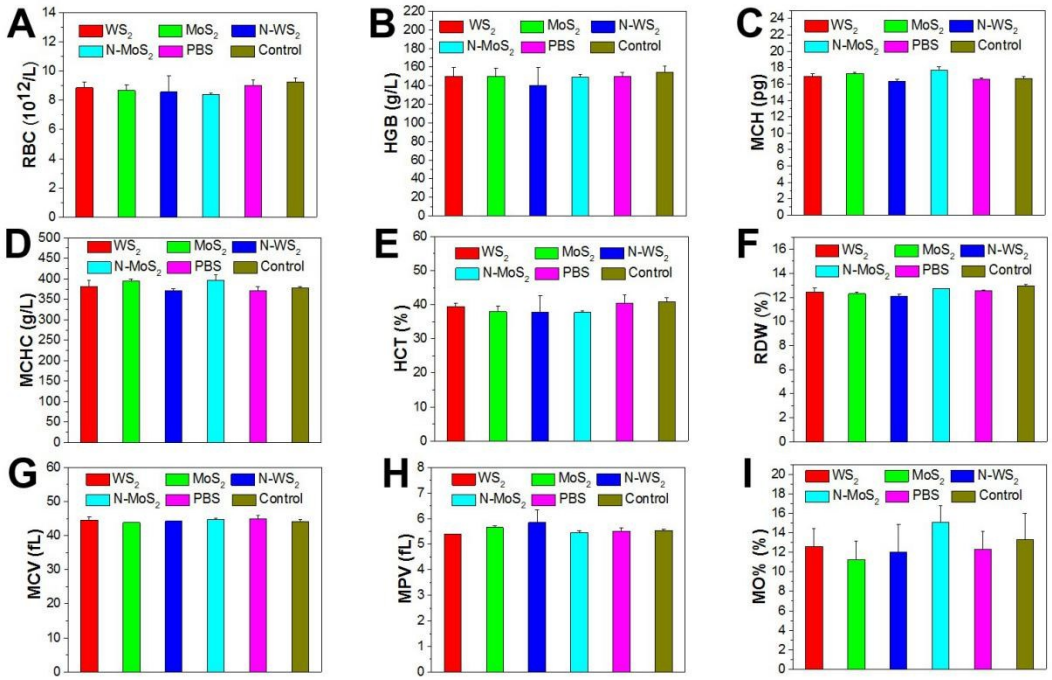

Figure S11. Blood routine analysis of mice suffering from Amp^r *E. coli*-infected wound treatment at the eighth day.

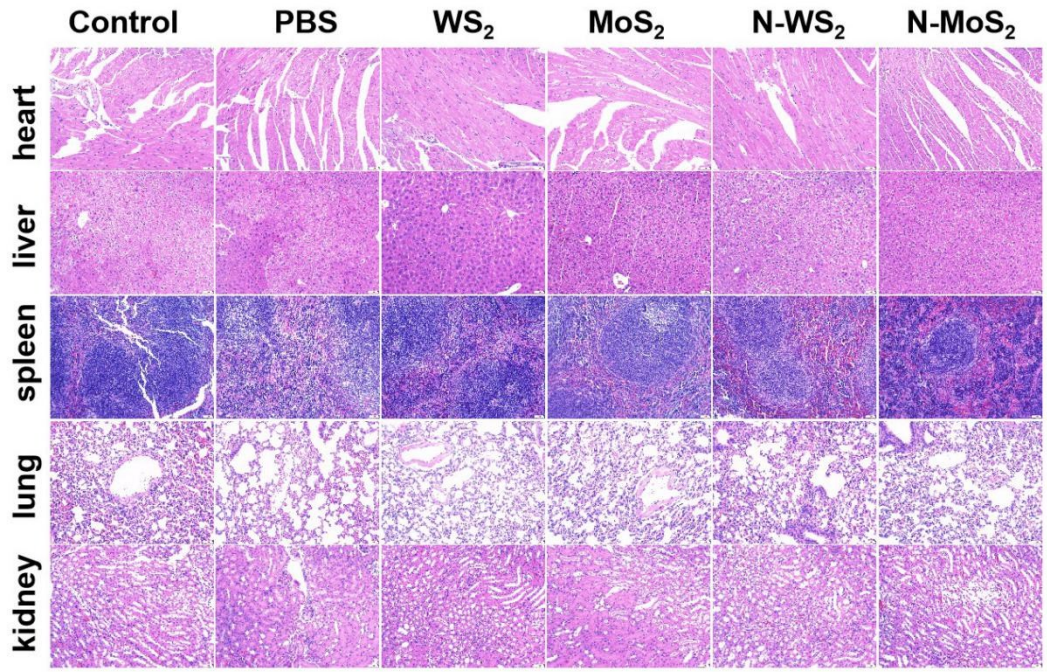

Figure S12. Histopathology analysis of the main organs that were treated with different groups at the eighth day.

References

1. J. N. Coleman, M. Lotya, A. O'Neill, S. D. Bergin, P. J. King, U. Khan, K. Young, A. Gaucher, S. De, R. J. Smith, I. V. Shvets, S. K. Arora, G. Stanton, H.-Y. Kim, K. Lee, G. T. Kim, G. S. Duesberg, T. Hallam, J. J. Boland, J. J. Wang, J. F. Donegan, J. C. Grunlan, G. Moriarty, A. Shmeliov, R. J. Nicholls, J. M. Perkins, E. M. Grieveson, K. Theuwissen, D. W. McComb, P. D. Nellist and V. Nicolosi, *Science*, 2011, **331**, 568-571.
2. A. O'Neill, U. Khan and J. N. Coleman, *Chem. Mater.*, 2012, **24**, 2414-2421.
3. K. Lee, H.-Y. Kim, M. Lotya, J. N. Coleman, G.-T. Kim and G. S. Duesberg, *Adv. Mater.*, 2011, **23**, 4178-4182.
4. V. Pachauri, K. Kern and K. Balasubramanian, *APL Mater.*, 2013, **1**, 032102.
5. K.-G. Zhou, N.-N. Mao, H.-X. Wang, Y. Peng and H.-L. Zhang, *Angew. Chem. Int. Ed.*, 2011, **50**, 10839-10842.
6. E. Varrla, C. Backes, K. R. Paton, A. Harvey, Z. Gholamvand, J. McCauley and J. N. Coleman, *Chem. Mater.*, 2015, **27**, 1129-1139.
7. G. Guan, S. Zhang, S. Liu, Y. Cai, M. Low, C. P. Teng, I. Y. Phang, Y. Cheng, K. L. Duei, B. M. Srinivasan, Y. Zheng, Y.-W. Zhang and M.-Y. Han, *J. Am. Chem. Soc.*, 2015, **137**, 6152-6155.
8. C. Han, Y. Zhang, P. Gao, S. Chen, X. Liu, Y. Mi, J. Zhang, Y. Ma, W. Jiang and J. Chang, *Nano Lett.*, 2017, **17**, 7767-7772.
9. C. Yeon, I. Lee, G.-H. Kim and S. J. Yun, *Langmuir*, 2017, **33**, 1217-1226.
10. R. J. Smith, P. J. King, M. Lotya, C. Wirtz, U. Khan, S. De, A. O'Neill, G. S. Duesberg, J. C. Grunlan, G. Moriarty, J. Chen, J. Wang, A. I. Minett, V. Nicolosi and J. N. Coleman, *Adv. Mater.*, 2011, **23**, 3944-3948.
11. G. S. Bang, K. W. Nam, J. Y. Kim, J. Shin, J. W. Choi and S.-Y. Choi, *ACS Appl. Mater. Interfaces*, 2014, **6**, 7084-7089.
12. W. Yin, L. Yan, J. Yu, G. Tian, L. Zhou, X. Zheng, X. Zhang, Y. Yong, J. Li, Z. Gu and Y. Zhao, *ACS Nano*, 2014, **8**, 6922-6933.
13. C. Backes, B. M. Szydłowska, A. Harvey, S. Yuan, V. Vega-Mayoral, B. R. Davies, P.-l. Zhao, D. Hanlon, E. J. G. Santos, M. I. Katsnelson, W. J. Blau, C. Gadermaier and J. N. Coleman, *ACS Nano*, 2016, **10**, 1589-1601.
14. W. Wu, J. Xu, X. Tang, P. Xie, X. Liu, J. Xu, H. Zhou, D. Zhang and T. Fan, *Chem. Mater.*, 2018, **30**, 5932-5940.
15. D. Xu, P. Xu, Y. Zhu, W. Peng, Y. Li, G. Zhang, F. Zhang, T. E. Mallouk and X. Fan, *ACS Appl. Mater. Interfaces*, 2018, **10**, 2810-2818.
16. D. Chakravarty and D. J. Late, *Eur. J. Inorg. Chem.*, 2015, **2015**, 1973-1980.
17. R. Jha and P. K. Guha, *J. Mater. Sci.*, 2017, **52**, 7256-7268.

18. S. Biccai, S. Barwich, D. Boland, A. Harvey, D. Hanlon, N. McEvoy and J. N. Coleman, *2D Mater.*, 2018, **6**, 015008.

19. X. Hai, K. Chang, H. Pang, M. Li, P. Li, H. Liu, L. Shi and J. Ye, *J. Am. Chem. Soc.*, 2016, **138**, 14962-14969.