

14 **Index**

15 **Supplementary Figures**

16 Figure S1 CLSM visualization of polysaccharides and proteins.

17 Figure S2 TEM-EDS elemental mapping of carbon (C), nitrogen (N), oxygen (O), and
18 sulfur (S) across the organic coating and sulfur core of bio-S⁰ variants.

19 Figure S3 Cryo-XPS spectra of all bio-S⁰ variants.

20 Figure S4 Correlation analysis between organic coating thickness and physicochemical
21 properties.

22 Figure S5 XRD patterns of S⁰ (purity $\geq 99.9\%$) after the same four treatments.

23 Figure S6 Raman spectra of S⁰ (purity $\geq 99.9\%$) after the same four treatments.

24 Figure S7 Correlation analysis between coating thickness and nitrate removal rate.

25 Figure S8 Sulfate generation (a, b) and theoretical vs. actual yield differences (c, d) in
26 the first (a, c) and second (b, d) rounds of denitrification experiments with bio-S⁰
27 variants.

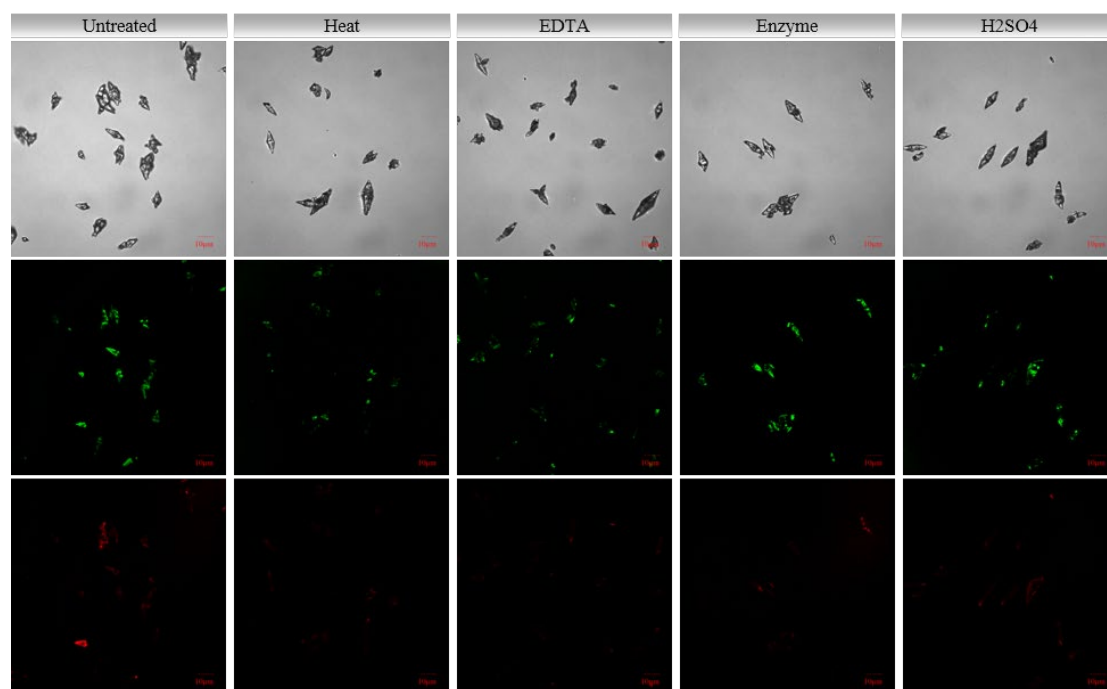
28 Figure S9 Sulfur consumption rates calculated from sulfate production in the second
29 round.

30 Figure S10 Correlation heatmap showing relationships among organic coating
31 structural parameters, physicochemical properties, denitrification performance, and key
32 microbial genera.

33 **Supplementary Tables**

34 Table S1. Alpha diversity indices of microbial communities in different bio-S⁰ variants
35 after the second-round denitrification experiments.

36 **Supplementary Figures**



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38 Figure S1 CLSM visualization of polysaccharides and proteins.

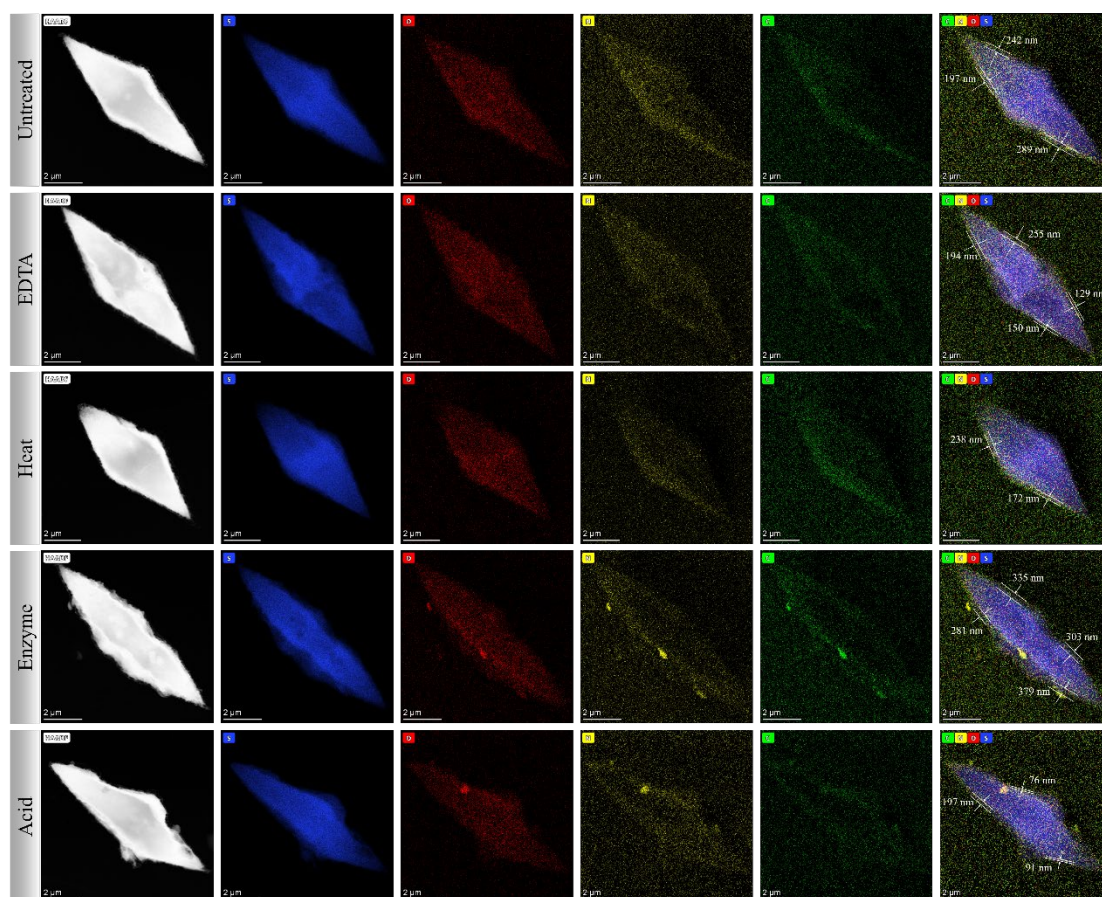
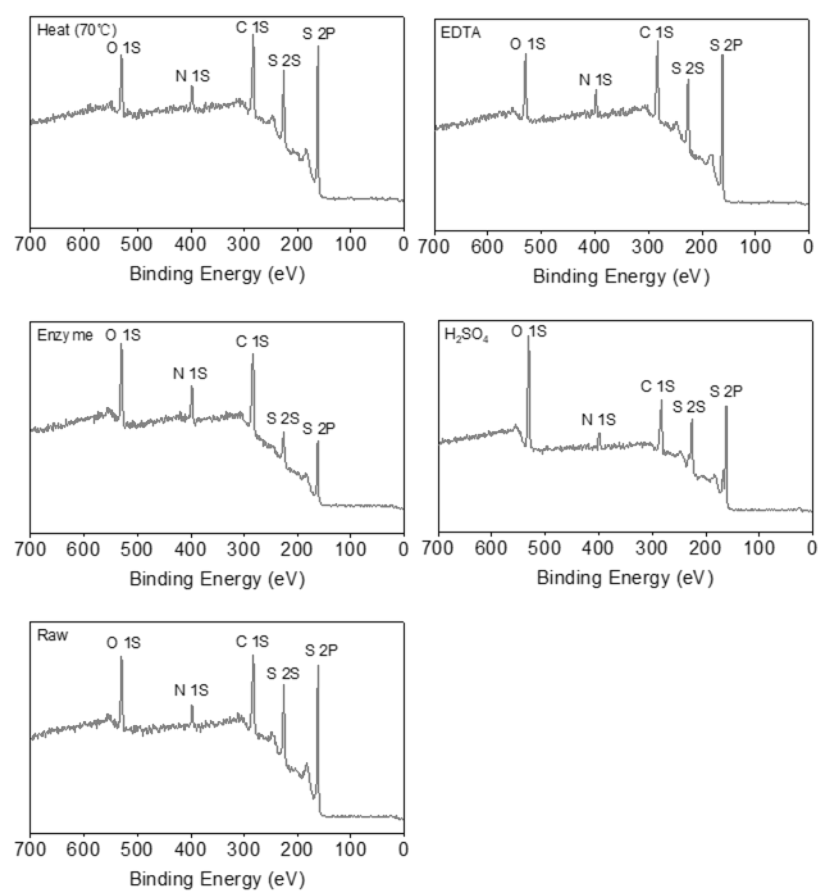
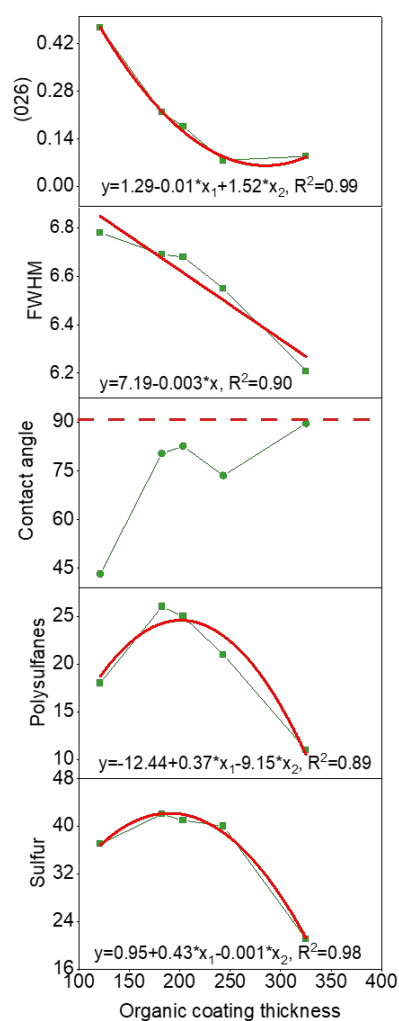


Figure S2 TEM-EDS elemental mapping of carbon (C), nitrogen (N), oxygen (O), and sulfur (S) across the organic coating and sulfur core of bio-S⁰ variants.



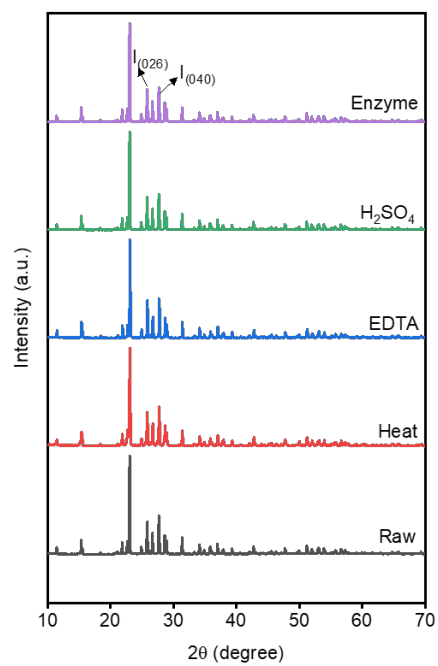
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43 Figure S3 Cryo-XPS spectra of all bio-S⁰ variants.



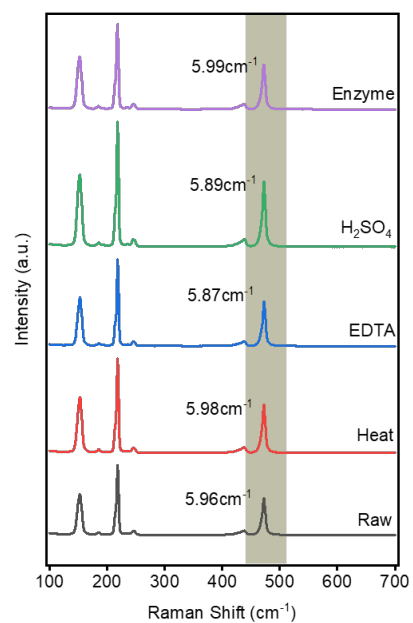
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45 Figure S4 Correlation analysis between organic coating thickness and physicochemical
 46 properties.



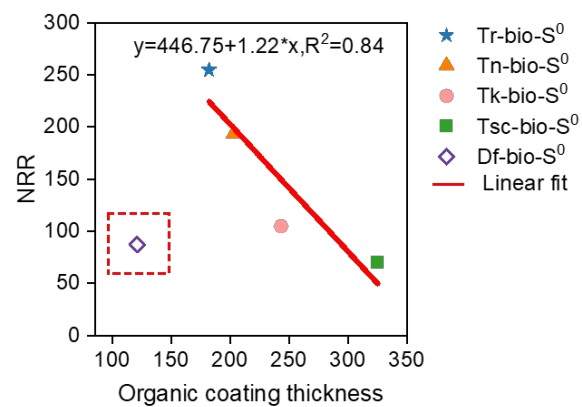
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48 Figure S5 XRD patterns of S^0 (purity $\geq 99.9\%$) after the same four treatments.



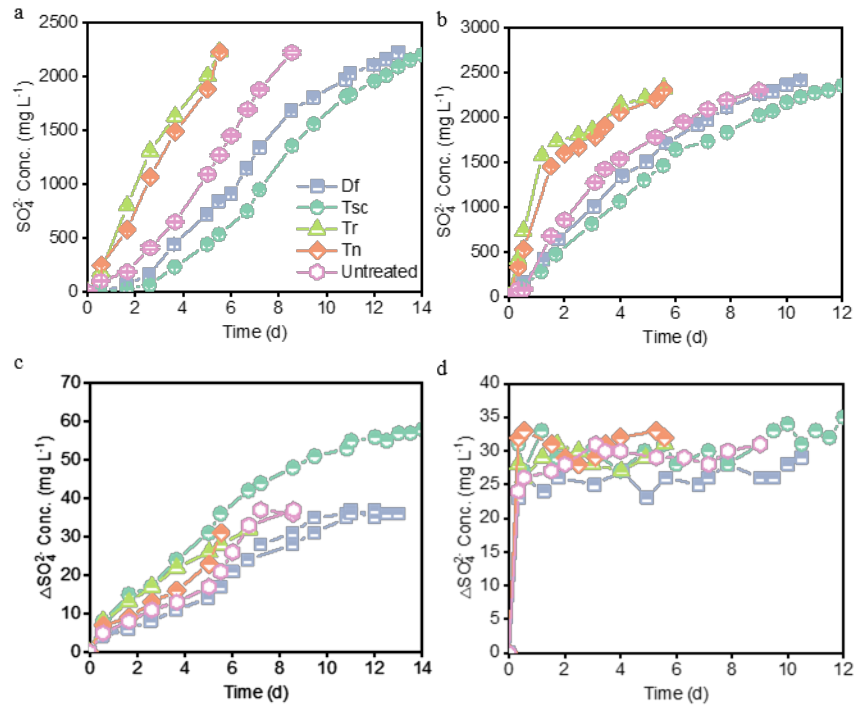
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50 Figure S6 Raman spectra of S^0 (purity $\geq 99.9\%$) after the same four treatments.



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52 Figure S7 Correlation analysis between coating thickness and nitrate removal rate.

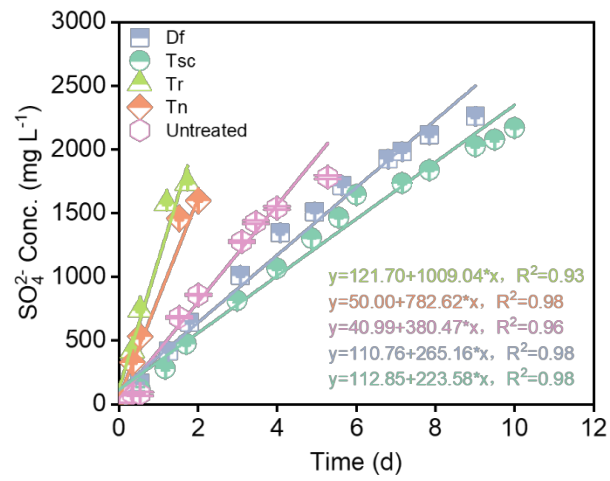


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55 the first (a, c) and second (b, d) rounds of denitrification experiments with bio-S⁰

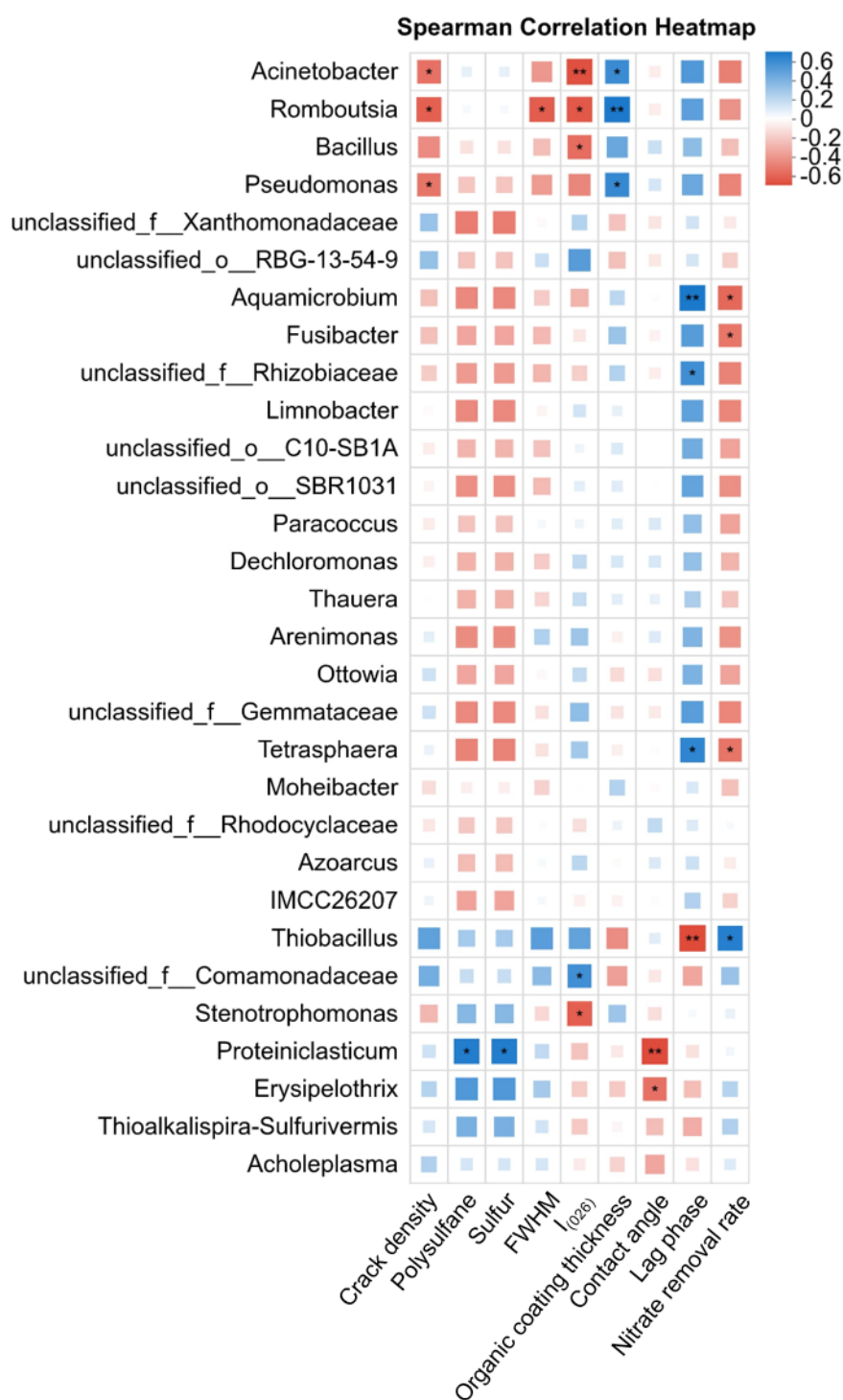
56 variants.



57

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59 round.



60

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 63 microbial genera.

64 **Supplementary Tables**

65 Table S1 Alpha diversity indices of microbial communities in different bio-S⁰ variants
66 after the second-round denitrification experiments.

		ACE	CHAO	SHANNON	SOBS
EDTA	Average	865.177	832.492	1.894	659.000
	SD	71.411	62.460	0.345	45.174
HEAT	Average	855.675	831.688	2.121	636.333
	SD	166.029	167.617	0.247	123.982
ENZYME	Average	1202.446	1181.558	3.886	956.333
	SD	63.738	66.546	0.200	66.620
ACID	Average	848.193	821.783	2.315	635.333
	SD	141.965	169.766	0.126	121.525
UNTREATED	Average	881.118	848.258	2.966	634.000
	SD	73.066	72.305	0.148	63.640

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