

Supporting Information

Amino acid functionalized lignin polyampholyte as natural broad-spectrum antimicrobial agent for high-efficient personal protection

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It Includes 14 Pages, 4 Tables, 9 Figures.

Table S1. Molecular weight (M_w), polydispersity index ($P.D.I.$) and functional group content EHL and EHL-W.

Compound	M_w (Da)	M_n (Da)	$P.D.I.$	Ph-OH (mmol g ⁻¹)	-COOH (mmol g ⁻¹)
EHL	3100	1100	2.81	1.55±0.04	1.89±0.06
EHL-W	2100	800	2.61	1.98±0.02	2.02±0.03

Table S2. The reaction conditions and yields of amino acid modified EHL (EHL-AA-x).

EHL (g)	Water* (mL)	Amino acid	Amino acid/EHL (%)	Amino acid/Glyoxal (molar ratio)	Yield (%)
4.0	20	Arg	5	1:1	88.0
4.0	20	Arg	10	1:1	91.7
4.0	20	Arg	20	1:1	93.5
4.0	20	Arg	40	1:1	95.5
4.0	20	Lys	40	1:1	91.3
4.0	20	His	40	1:1	86.3

*The pH value of water was 12.

Table S3. The elemental contents of EHL-AA-x.

Compound	N%	C%	H%	O%*
EHL-Arg-5	2.8	55.0	6.5	35.7
EHL-Arg-10	4.4	56.0	6.8	32.8
EHL-Arg-20	6.0	54.4	6.7	32.9
EHL-Arg-40	6.7	54.8	7.0	31.5
EHL-Lys-40	6.4	56.4	7.3	29.9
EHL-His-40	5.9	55.6	6.6	31.9

*O%=100%-N%-C%-H%.

Table S4. The antibacterial effect of cotton fabric containing antibacterial agent.

Samples	Treating time (min)	Bactericidal efficiency (%)	Antiadhesive Properties	References
Metal-organic framework based filter	30	>99.99	No	Ref 8
Zinc oxide nanoparticles coated fabrics	30	100	No	Ref 47
Silver nanoparticles coated fabrics	30	99	No	Ref 47
Titanium dioxide nanoparticles coated fabrics	30	85	No	Ref 47
Antibacterial nanogels coated fabrics	720	>99.99	Good	Ref 12
Our work	30	95	Good	-

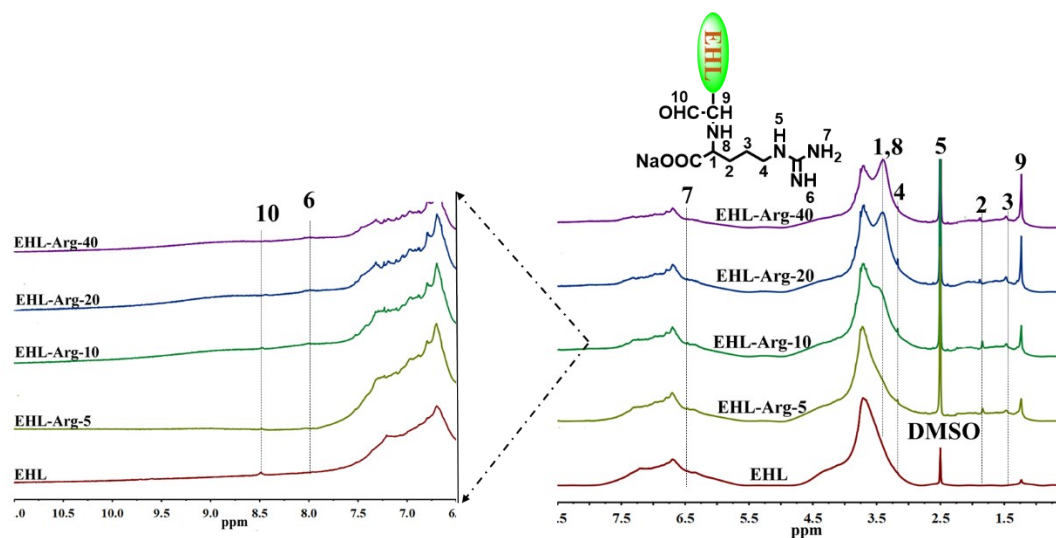


Figure S1. The ^1H NMR spectra of EHL and EHL-Arg-x.

EHL-Arg-x: ^1H NMR (600 MHz, $\text{DMSO}-d_6$): δ 1.23 (H-9, aliphatic H in EHL), 1.47 (H-3), 1.88 (H-2), 2.5 (H-5, $\text{DMSO}-d_6$), 3.18 (H-4), 3.4 (H-1, H-8), 3.71 ($-\text{OCH}_3$ in EHL), 6.46–7.31 (H-7, Ar-H in EHL), 7.95 (H-6), 8.50 (H-10, $-\text{CHO}$ in EHL) ppm.

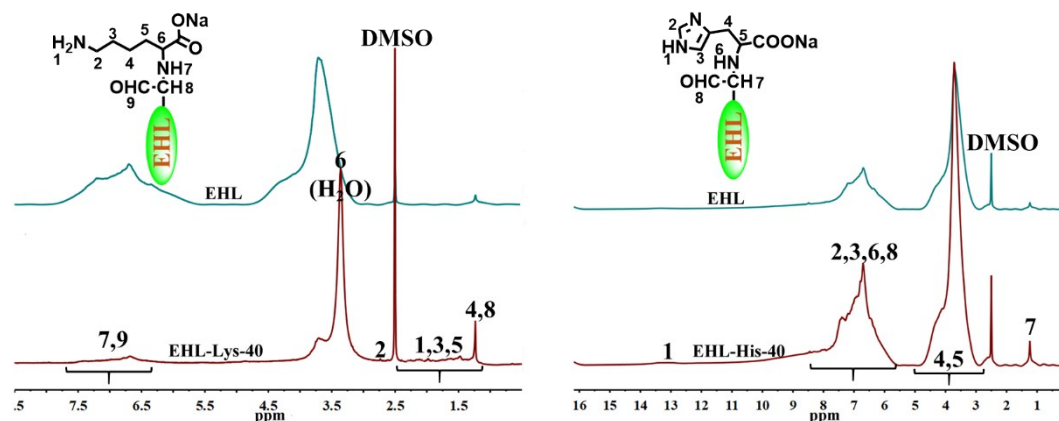


Figure S2. The ¹H NMR spectra of EHL, EHL-Lys-40 and EHL-His-40.

EHL-Lys-40: ¹H NMR (600 MHz, DMSO-*d*₆): δ 1.23 (H-4, H-8 aliphatic H in EHL), 1.48-2.01 (H-1, H-3, H-5), 2.5 (DMSO-*d*₆), 2.74 (H-2), 3.36 (H-6, H₂O), 3.71 (-OCH₃ in EHL), 6.46-8.50 (H-7, H-9, Ar-H and -CHO in EHL) ppm.

EHL-His-40: ¹H NMR (600 MHz, DMSO-*d*₆): δ 1.23 (H-7 aliphatic H in EHL), 2.5 (DMSO-*d*₆), 2.61-5.25 (H-4, H-5), 6.46-8.50 (H-2, H-3, H-6, H-8, Ar-H and -CHO in EHL), 11.3 (H-1) ppm.

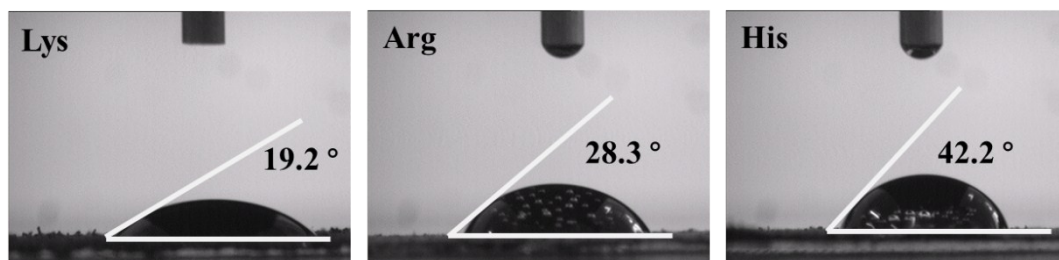


Figure S3. The water contact angles of three natural base amino acids including Lys, Arg and His.

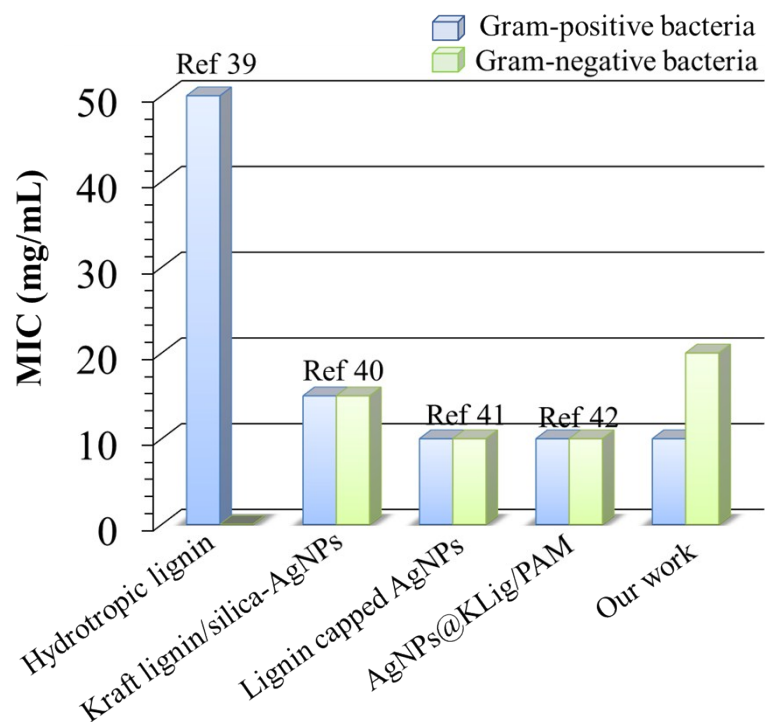


Figure S4. MIC values of lignin based antibacterial materials.

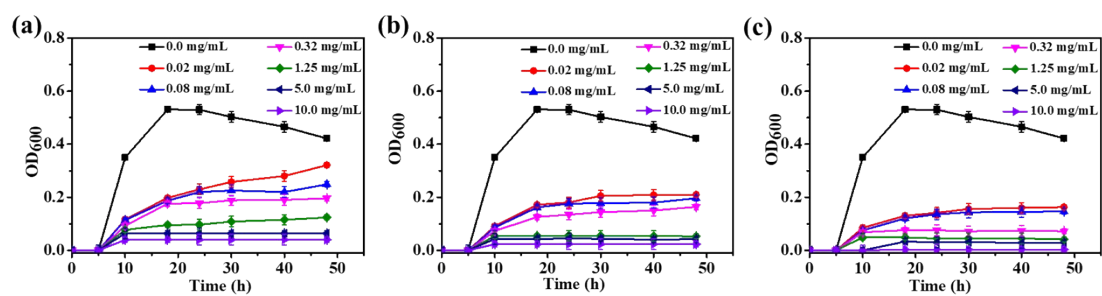


Figure S5. OD_{600} regrowth curves of *S. aureus* suspension treated with different concentrations of samples: (a) EHL-Lys-40; (b) EHL-His-40; (c) EHL-Arg-40.

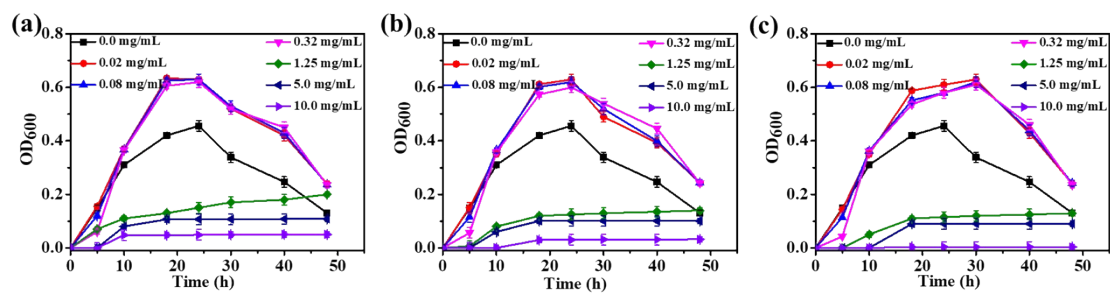


Figure S6. OD regrowth curves of *E. faecalis* suspension treated with different concentrations of samples: (a) EHL-Lys-40; (b) EHL-His-40; (c) EHL-Arg-40.

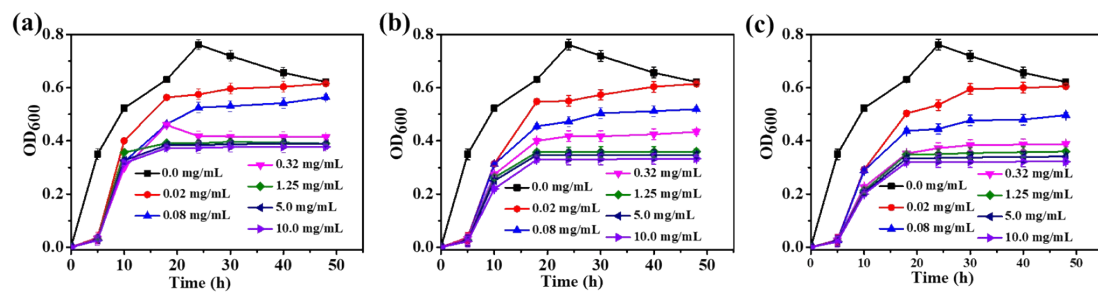


Figure S7. OD regrowth curves of *E. coli* suspension treated with different concentrations of samples: (a) EHL-Lys-40; (b) EHL-His-40; (c) EHL-Arg-40.

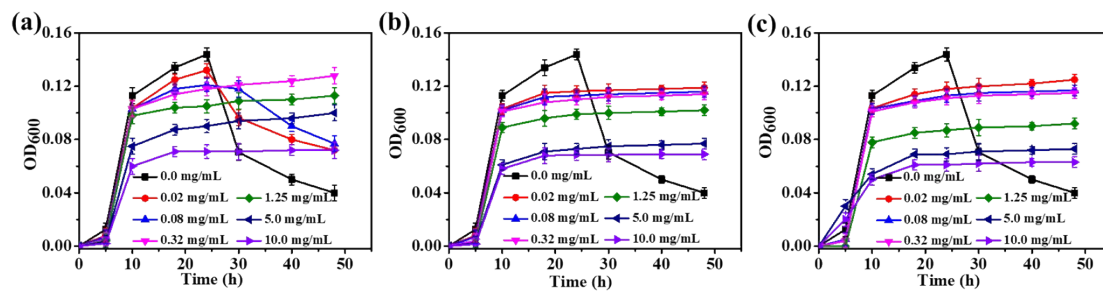


Figure S8. OD regrowth curves of *P. aeruginosa* suspension treated with different concentrations of samples: (a) EHL-Lys-40; (b) EHL-His-40; (c) EHL-Arg-40.

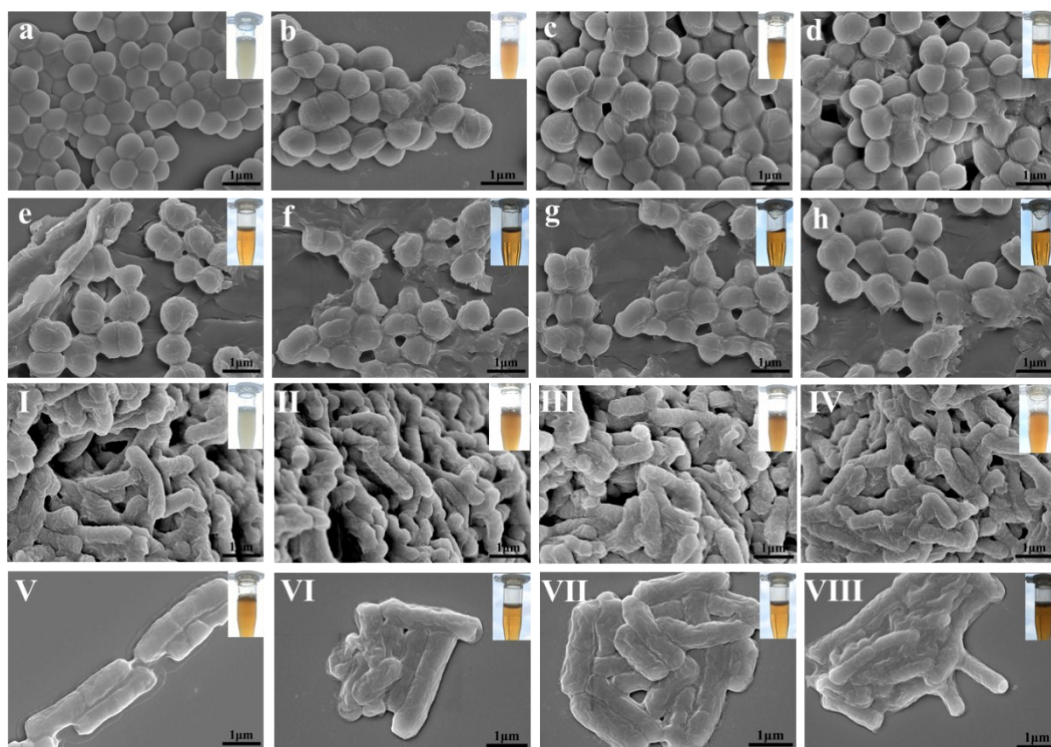


Figure S9. Morphology changes of Gram-positive and Gram-negative bacteria before and after EHL-AA-X treating. SEM images of *S. aureus* (a-h) and *E. coli* (I-VIII): (a, I) Untreated bacteria; Other bacteria treated with 10 mg/mL EHL-W and EHL-AA-x including (b, II) EHL-W; (c, III) EHL-Arg-5; (d, IV) EHL-Arg-10; (e, V) EHL-Arg-20; (f, VI) EHL-Arg-40; (g, VII) EHL-His-40; (h, VIII) EHL-Lys-40. Insert: The appearance of bacteria before and after antibacterial treating.