Electronic Supplementary Information (ESI)

pH-Dependent interaction mechanism of lignin nanofilms

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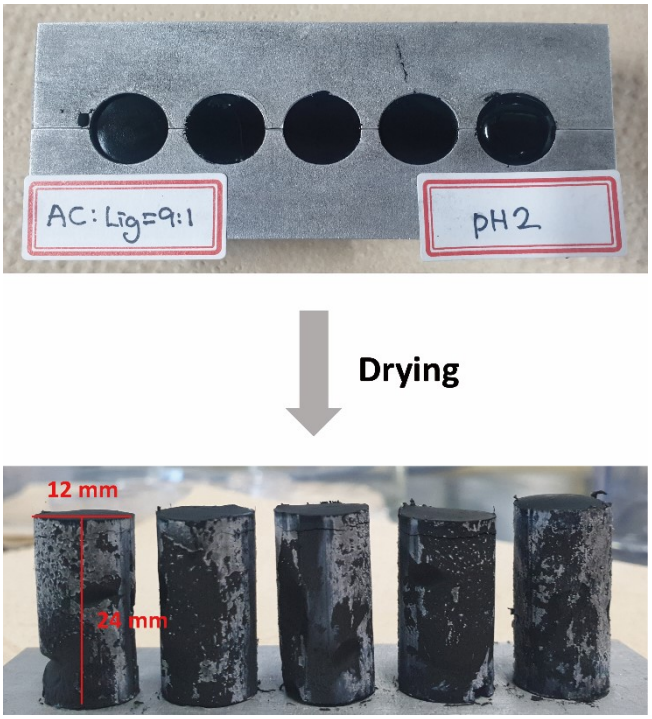

Fig. S1 Lignin-activated carbon composites specimen at pH 2 solution before drying and after drying.

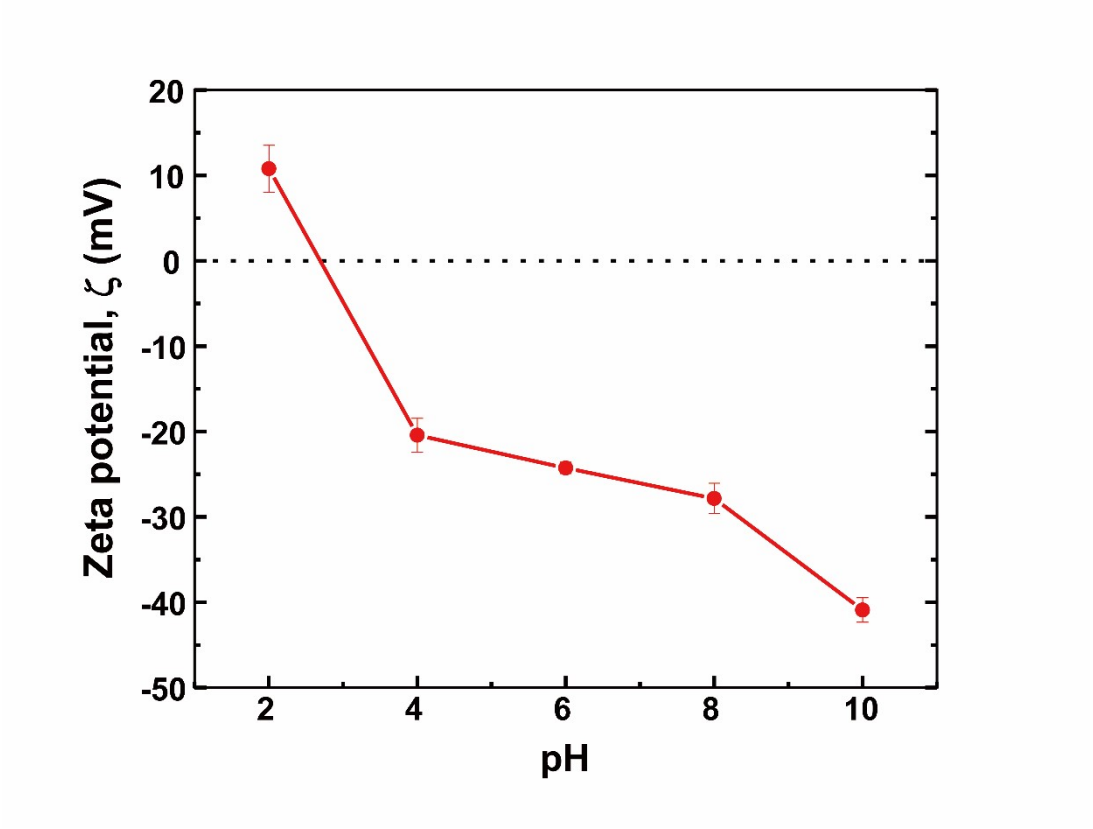

Fig. S2 Zeta potential of lignin at various pH solutions (pH 2 ~ pH 10), adjusted with HNO_3 and NaOH.

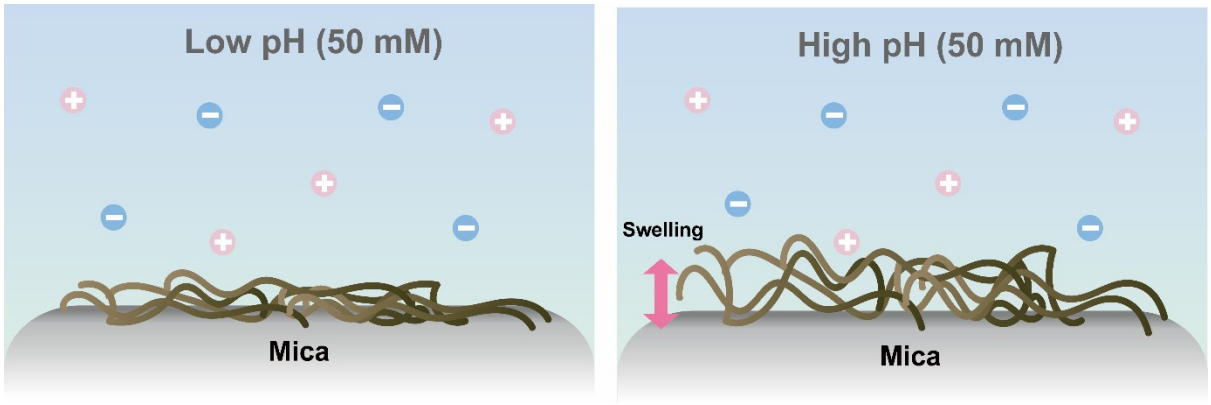

Fig. S3 Schematic illustration of lignin swelling in 50 mM solution at low pH and high pH, respectively.

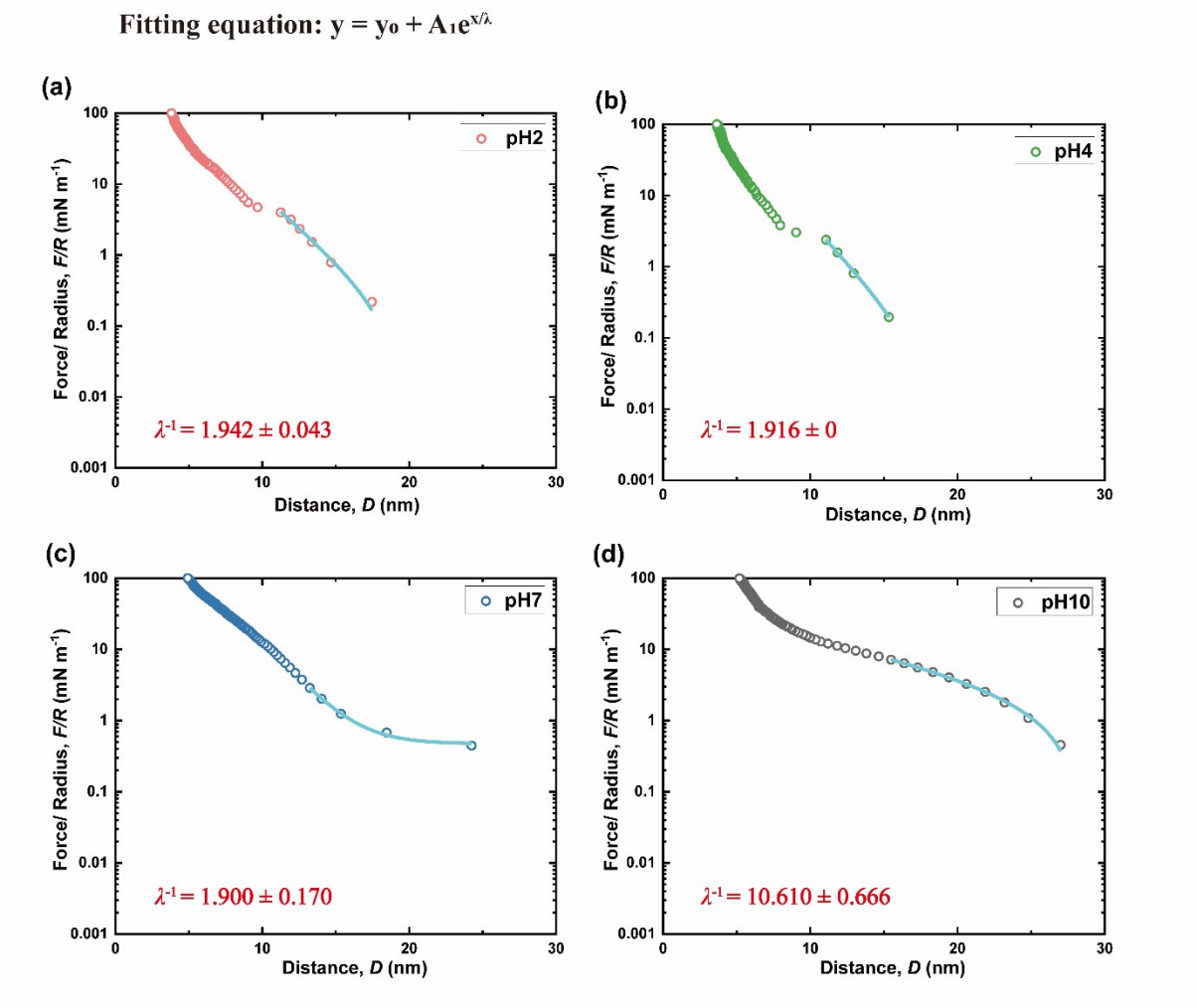

Fig. S4 Decay length fitting at SFA force−distance profiles of lignin−lignin interactions in (a) pH 2, (b) pH 4, (c) pH 7, and (d) pH 10.

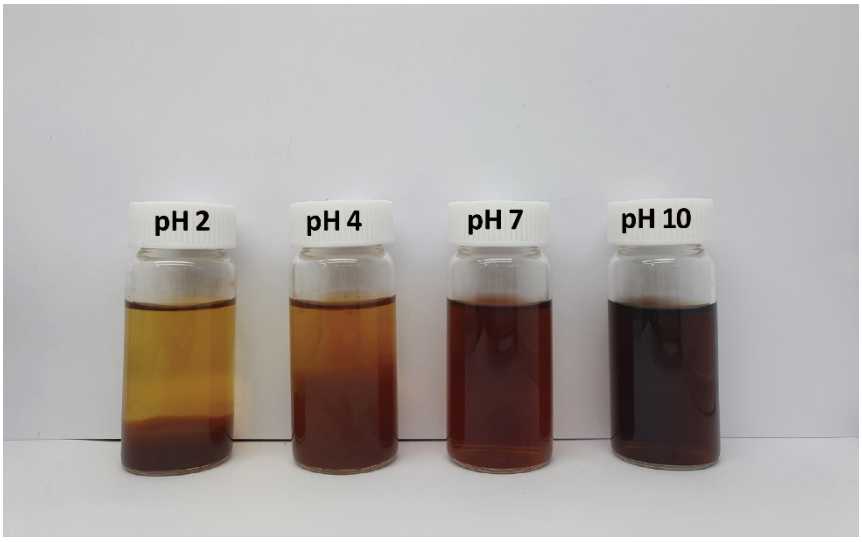

Fig. S5 Photograph of prepared lignin solution (5 mg/ml) in water with different pH conditions: 2.0, 4.0, 7.0, 10.0.

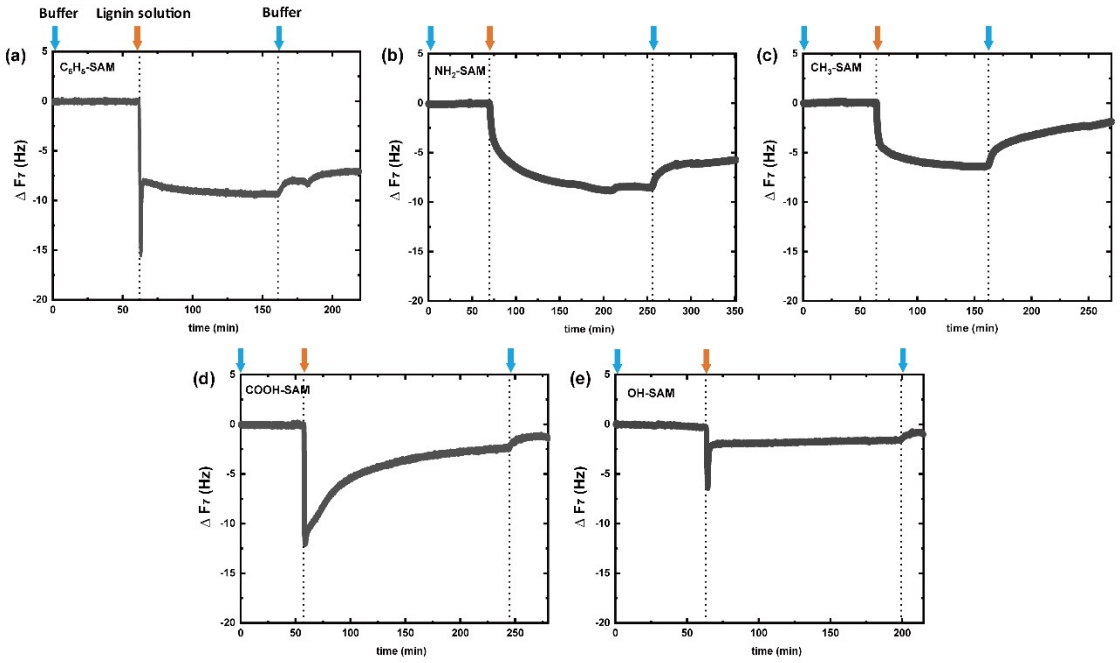

Fig. S6 QCM-D measurements of lignin adsorption on each SAM coated gold surface; (a) C_6H_5, (b) NH_2 (c) CH_3, (d) COOH, and (e) OH. Blue and orange arrows indicate injection of buffer and lignin solution, respectively.

Table S1 Amount of sodium hydroxide, nitric acid, and sodium nitrate in total volume of solution at each pH buffer solution.

pH	**2**	**4**	**7**	**10**
Amount of NaOH [mg]	48.6	99.5	100.0	100.5
Amount of 10 M HNO_3 solution [ml]	0.25	0.25	0.25	0.25
Additional amount of $NaNO_3$ [mg]	267.6	213	212.5	212.0

Table S2 pK_a values of each SAM tested in this research.

Functionalized SAM	**pK_a[1-3]**
11-hydroxy-1-undecanethiol **(OH-SAM)**	**16-18**
10-carboxy-1-decanethiol **(COOH-SAM)**	**5.5**
2-phenylethanethiol **(C_6H_5-SAM)**	**-**
1-undecanethiol **(CH_3-SAM)**	**-**
11-amino-1-undecanethiol hydrochloride **(NH_2-SAM)**	**7.5**

Table S3 The theoretical and fitted Debye-length value from decay length fitting

pH	**2**	**4**	**7**	**10**
Debye length, κ^{-1}; nm (Theoretical)	1.923	1.923	1.923	1.923
Decay length, λ^{-1}; nm (Fitted)	1.942 ± 0.043	1.916 ± 0	1.900 ± 0.170	10.610 ± 0.666

Supplementary References

1. G. C. Shields, *Computational approaches for the prediction of pKa values*, CRC Press, 2019.

2. H. Zhang, H.-X. He, J. Wang, T. Mu and Z.-F. Liu, *Applied Physics A*, 1998, **66**, S269-S271.

3. D. V. Vezenov, A. Noy, L. F. Rozsnyai and C. M. Lieber, *Journal of the American Chemical Society*, 1997, **119**, 2006-2015.