

Supplementary Information

Waste-Derived-Lignin Based Hydrogel as Reusable Adsorbent for Efficient Removal of Dyes from Aqueous Solution

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Table S1: Composition of Lignin/PVA hydrogel samples prepared with varying Lignin content.

Sample No.	Lignin (g)	PVA (g)	ECH (mL)	2 M NaOH (mL)	H ₂ O (mL)	Total volume (mL)
1	0				3.8	
2	0.1				3.7	
3	0.2				3.6	
4	0.3	0.8	0.4	5.0	3.5	10.0
5	0.4				3.4	
6	0.45				3.35	

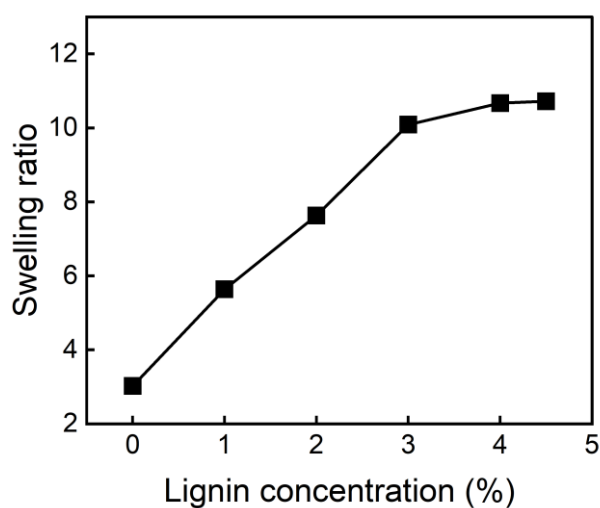


Fig. S1: Swelling ratio of the swollen Lignin/PVA gels in DI water at various Lignin concentrations

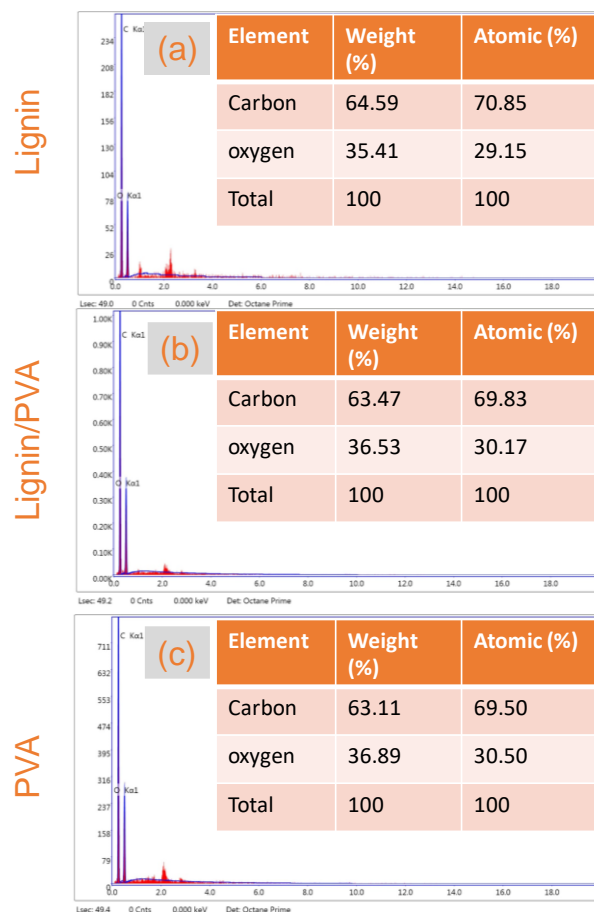


Fig. S2. EDS analysis of (b) extracted Lignin, (d) Lignin/PVA gel, (f) PVA gel.

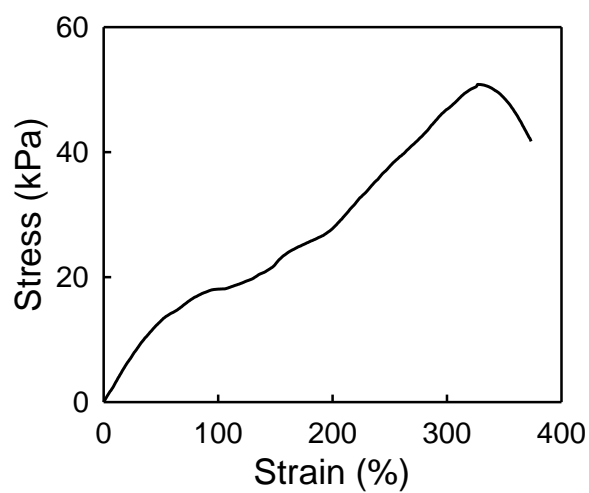


Fig. S3. Stress-strain curve of as-prepared Lignin/PVA hydrogel of 3% Lignin concentration.

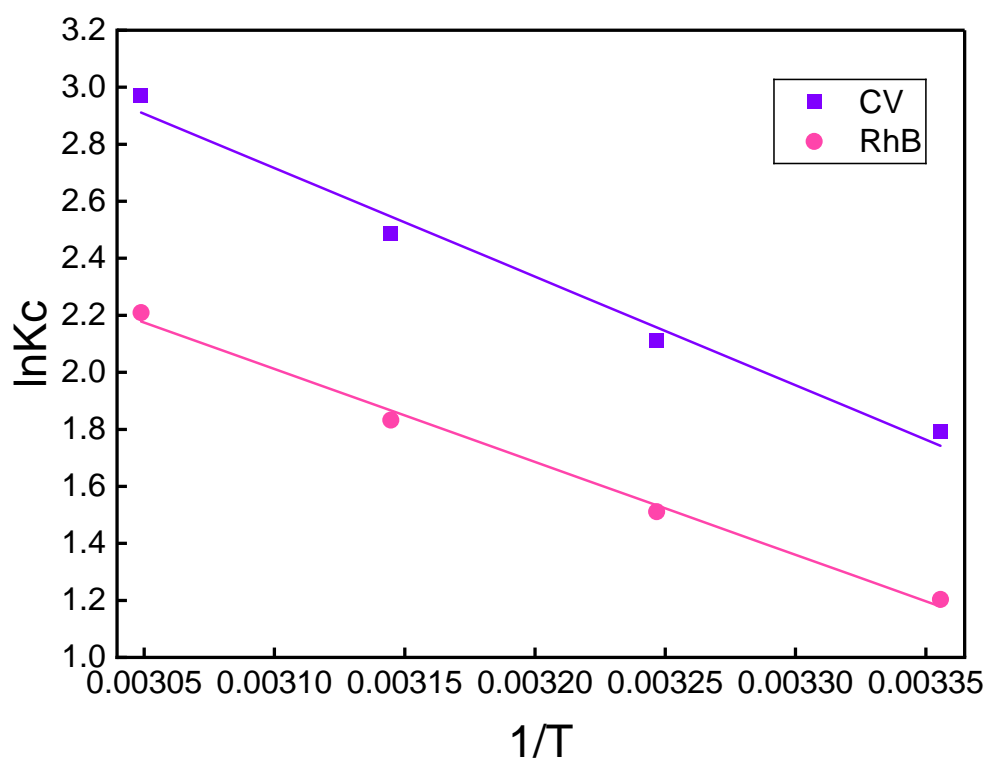


Fig. S4. Van't Hoff plots ($\ln Kc$ vs $1/T$) for the adsorption of crystal violet (CV) and rhodamine B (RhB) onto Lignin/PVA hydrogel. The linear fitting was used to determine the thermodynamic parameters, where the slope ($-\Delta H^\circ/R$) and intercept ($\Delta S^\circ/R$) correspond to the enthalpy (ΔH°) and entropy (ΔS°) changes, respectively.

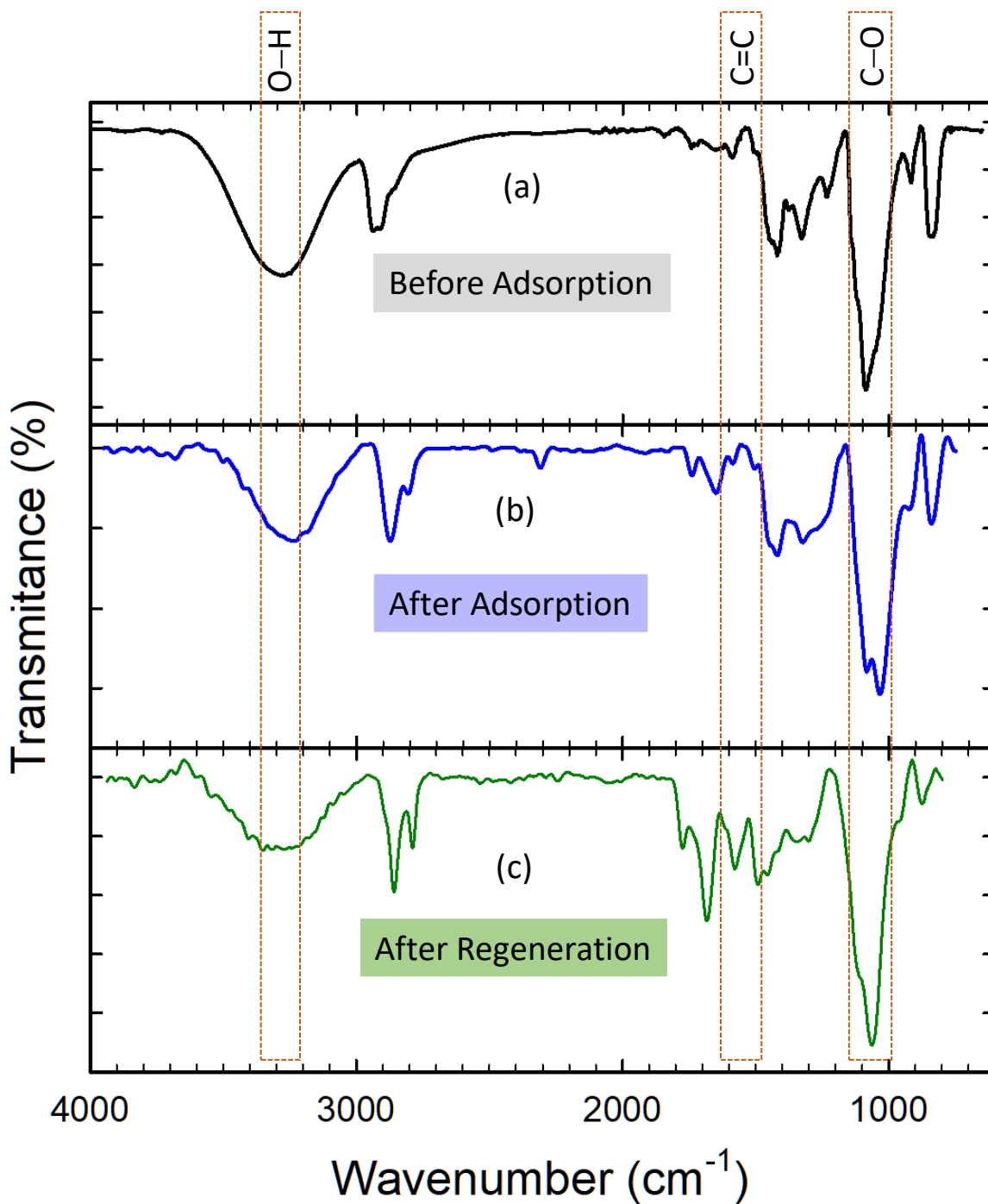


Fig. S5: FT-IR spectra of Lignin/PVA gel (a) before adsorption, (b) after adsorption, and (c) after regeneration.

The FTIR spectra of the hydrogel before and after adsorption were compared to elucidate the interaction mechanism [Fig. S5 (a)-(b)]. Although noticeable shift in peak position was not observed, the broad O–H stretching band ($\sim 3200\text{--}3400\text{ cm}^{-1}$) shifted toward lower wavenumbers, suggesting the formation of hydrogen bonding between hydroxyl groups of the

Lignin/PVA network and dye molecules. The aromatic C=C stretching peaks ($\sim 1600\text{--}1510\text{ cm}^{-1}$) also showed slight shifts, indicating $\pi\text{--}\pi$ interactions between the aromatic rings of Lignin and the conjugated structure of the dyes. Furthermore, the C–O stretching band ($\sim 1050\text{--}1100\text{ cm}^{-1}$) exhibited shifts and intensity changes, implying the participation of oxygen-containing functional groups in the adsorption process. These observations indicate that the adsorption process is governed by a combination of electrostatic attraction, hydrogen bonding, and $\pi\text{--}\pi$ interactions. The results are consistent with the kinetic and thermodynamic analyses, confirming the contribution of both physical and chemical interactions.

The FT-IR spectra of the Lignin/PVA hydrogel after adsorption (Fig. S5b) and after regeneration (Fig. S5c) were compared with that of the pristine hydrogel (Fig. S5a) to elucidate the regeneration and gradual decline of adsorption performance. After adsorption, the O–H stretching band exhibited a shift toward lower wavenumbers, indicating the formation of hydrogen bonding interactions between hydroxyl groups of the adsorbent and dye molecules. In addition, changes in the aromatic C=C stretching peaks were observed, suggesting the involvement of $\pi\text{--}\pi$ interactions between Lignin aromatic rings and the conjugated structures of CV and RhB. After regeneration, the characteristic bands (O–H, C=C, and C–O) partially return toward their original positions, indicating partial recovery of the hydrogel structure. However, slight deviations remain, suggesting incomplete desorption and gradual modification of active sites, which may contribute to the observed decline in adsorption performance over repeated cycles.