

Article

Mechanical Performance and Dye Adsorption of Lignin/Poly(ethylene glycol) Diglycidyl Ether/Sorbitol Polyglycidyl Ether Hydrogels

Yiming Wang ¹, Hailing Liu ^{1,*}, Yu-Peng He ^{2,*} and Guoqing Guan ³

¹ College of Petrochemical Engineering, Liaoning Petrochemical University, Dandong Road West 1, Wanghua District, Fushun 113001, China

² State Key Laboratory of Fine Chemicals, Ningbo Institute of Dalian University of Technology, No. 26 Yucai Road, Jiangbei District, Ningbo 315016, China

³ Graduate School of Science and Technology, Institute of Regional Innovation (IRI), Hirosaki University, 3-Bunkyocho, Hirosaki 036-8561, Japan; guan@hirosaki-u.ac.jp

* Correspondence: hl2490053@outlook.com (H.L.); yupeng.he@dlut.edu.cn (Y.-P.H.); Tel.: +86-130-0926-8874 (H.L.)

Abstract

A bisphenol-A-free lignin hydrogel platform with programmable network density is reported. Lignin was crosslinked with poly(ethylene glycol) diglycidyl ether (PEGDGE) and sorbitol polyglycidyl ether (SPE) via epoxide ring-opening to generate hydrogel networks spanning eleven PEGDGE/SPE ratios. A single compositional lever—the SPE fraction—allowed the predictable densification of the network, translating into a monotonic shift in swelling and viscoelastic/mechanical responses. Importantly, the well-performing hydrogel (LSiP₉) coupled swelling ratio with adsorption functionality, removing 72% methylene blue from water under the tested conditions. This work positions lignin as more than a passive filler: it serves as an active phenolic macromonomer for designing sustainable, multifunctional hydrogels.

Keywords: polymers; PEGDGE; SPE; tunable mechanical property; MB adsorption; crosslinking

1. Introduction

Hydrogels are hydrophilic polymers in a network structure [1–3]. Owing to their high water uptake and ability to absorb and/or adsorb solutes such as dyes [4,5], hydrogels have been widely explored for applications including wastewater treatment and drug delivery [6–9]. During the COVID-19 pandemic, an inhalable bioadhesive hydrogel was developed to form a protective airway barrier and prevent viral infection in animal models [10]. Polymer-nanoparticle hydrogel systems have also been used for controlled vaccine release [11]. Common hydrogel components include poly(ethylene glycol) diglycidyl ether (PEGDGE) [3,12], sorbitol polyglycidyl ether (SPE) [13], bisphenol A [14], chitosan [15], and polyacrylamide [16]. Among these, PEGDGE is low-cost, non-toxic, and highly water-soluble [17]. SPE, in addition to its use as a plasticizer [18], has recently been employed as a crosslinker for hydrogel formation [13,19,20]. Bisphenol A is widely used because it improves processability (processability means how easily a material can be manufactured, for example, how well it flows, molds, extrudes, or forms during processing)

Academic Editor: Fei Han

Received: 23 January 2026

Revised: 11 February 2026

Accepted: 14 February 2026

Published: 15 February 2026

Copyright: © 2026 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the [Creative Commons Attribution \(CC BY\) license](https://creativecommons.org/licenses/by/4.0/).

[21]. However, the U.S. FDA has raised concerns about bisphenol A as an endocrine disruptor that may adversely affect reproductive, metabolic, and neurological systems, even at low exposure levels [22]. Therefore, replacing bisphenol A with greener alternatives is increasingly desirable.

Lignin has emerged as a promising bio-based substitute for bisphenol A in polymer networks because it is non-toxic, sustainable, and rich in hydroxyl groups [23,24]. Nikafshar et al. [25] demonstrated that organosolv corn stover lignin can completely replace bisphenol A in solubilized epoxy resins while maintaining thermomechanical performance. Xue et al. [26] partially replaced bisphenol A with lignin in a polyether amine (D-400) crosslinked network, resulting a 239% increase in elongation. Oveissi et al. [27] reported that incorporating 2.5 wt% lignin into polyether-based polyurethane hydrogels increased the fracture energy from 1540 to 2050 J m⁻² and increased the Young's modulus from 1.29 to 2.62 MPa.

Lignin-based hydrogels can also adsorb dyes for wastewater remediation because lignin contains abundant hydroxyl and phenolic groups [28–30]. Meng et al. [31] demonstrated that aminated lignin alone removed 90% of Direct Blue 1 from aqueous solution. Zhao et al. [32] developed a magnetic lignin- γ -Fe₂O₃ nanoparticle composite adsorbent that achieved up to 98% methylene blue in water. In this study, adsorbent hydrogels were prepared from lignin, poly(ethylene glycol) diglycidyl ether (PEGDGE), and sorbitol polyglycidyl ether (SPE). Eleven formulations with varying PEGDGE/SPE ratios were synthesized and characterized using swelling measurements, rheology, and compression tests. The results reveal clear structure–property relationships linking crosslinking density to hydrogel performance. In addition, the hydrogels show dye adsorption capability, indicating potential for wastewater treatment applications.

2. Materials and Methods

2.1. Materials

Lignin (Dealkaline) was purchased from TCI. Potassium hydroxide was purchased from Aladdin. Sorbitol polyglycidyl ether (SPE, purity: 99%, epoxy value: 0.52–0.63 mol 100 g⁻¹) was obtained from the Jingmen Dongxin Biotechnology Company (Jingmen, China). Poly(ethylene glycol) diglycidyl ether (PEGDGE, purity: 99%, epoxy value: 0.35 mol 100 g⁻¹) was purchased from Guangdong Qianjin Chemical Reagent Company (Foshan, China). The reagents are directly used without further purification unless stated.

2.2. Synthesis of LSnPn Hydrogels

In a 20 mL vial, SPE (amount in Table 1) was dissolved in 0.2 mL of DMF and shaken until homogeneous. Then, 1 g of lignin, 2.8 mL of 3.3 M potassium hydroxide solution, and a stir bar were added, and the mixture was stirred for 1 h to achieve homogeneity at room temperature. Next, PEGDGE (amount in Table 1) was added. The reaction was stirred to a homogeneous state and then placed in an oven at 40 °C for 12 h.

2.3. FTIR

FTIR spectra in transmission mode were acquired using an FTIR-600 spectrometer (Agilent, Santa Clara, CA, USA) with a resolution of 4 cm⁻¹ over a wavenumber range of 4000 to 400 cm⁻¹. The hydrogel samples were freeze-dried for 24 h prior to testing.

2.4. Rheological Characterization Subsection

The rheological properties of the samples were evaluated using a Netzsch Kinexus Lab+ rotational rheometer. All experiments were conducted with a 20 mm parallel-plate geometry with a gap of 1 mm. Prior to the frequency sweep, an amplitude sweep was

performed to define the linear viscoelastic region (LVR). Here, a stress sweep (0.1–100 Pa) was conducted, and both the storage modulus (G') and loss modulus (G'') remained essentially constant over this range, indicating that the measurements were within the LVR. Therefore, 5 Pa, which lies well within the identified LVR, was selected for the subsequent frequency sweep. Therefore, dynamic shear tests were performed over a frequency range of 0.1–20 Hz at 25 °C, and the G' and G'' were recorded.

2.5. Swelling Tests

Swelling tests of hydrogels were carried out by completely immersing samples in deionized water at room temperature for 24 h. The swollen hydrogels were weighed at specific time points. The water content was calculated by the formula:

$$\text{water content (\%)} = \frac{w_s - w_d}{w_d} \times 100 \quad (1)$$

where w_s and w_d are the weights of swollen sample and dry sample, respectively. Five swelling tests were carried out for each sample, and the results of the five repetitions were consistent.

2.6. Mechanical Properties

Mechanical properties were measured using a universal testing machine (AL-7000M, Gotech, Zhuhai, China) equipped with a 10 kN load cell. The samples were prepared in cylindrical shapes with a diameter of 12 mm and a height of 2 mm, which were then subjected to compression tests. The samples were compressed at a rate of 0.4 mm min^{−1} until they reached 80% strain. Each condition was tested five times. The cyclic compression was measured at 40% strain with five consecutive tests.

2.7. Dye Removal Experiments

The methylene blue (MB) dye adsorption ability of the synthesized hydrogels was measured. Dry hydrogels were finely cut into small pieces (1.6242 g) and put into 500 mL of aqueous solution of MB (30 mg L^{−1}). The adsorption was carried out on an orbital shaker (90 rpm) at room temperature for 48 h. The absorbance of MB in the solution was measured at 665 nm using a UV–VIS spectrometer (Cary 60 UV-Vis, Agilent, Santa Clara, CA, USA) at different time intervals. A calibration curve was plotted for the absorbance of MB solutions at 665 nm against the known MB concentration. The removal efficiencies (R) and adsorption capacities (q_e) of MB were calculated using the following equations:

$$R(\%) = \frac{C_0 - C_e}{C_0} \times 100 \quad (2)$$

$$q_e(\text{mg g}^{-1}) = \frac{(C_0 - C_e) \times V}{m} \quad (3)$$

where C_0 and C_e are the initial and final dye concentrations in the solution (mg L^{−1}), respectively; V is the volume (L) of the dye solution; and m is the mass (g) of the adsorbent.

2.8. Adsorption Kinetics

The adsorption behavior of LSIP₉ for MB was studied using the pseudo-first-order and pseudo-second-order adsorption kinetic models. The adsorption process was carried out in a MB solution (30 mg L^{−1}), and the adsorption capacity was measured at regular time intervals over a period of 48 h. The pseudo-first-order kinetic model was fitted using Equation (4), and the pseudo-second-order kinetic model was fitted using Equation (5).

$$q_t = q_e (1 - e^{-k_1 t}) \quad (4)$$

$$q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t} \quad (5)$$

where q_e (mg g⁻¹) is the equilibrium adsorption capacity; q_t (mg g⁻¹) is the adsorption capacity at time t ; and k_1 (min⁻¹) and k_2 (g mg⁻¹ min⁻¹) are the rate constants for the pseudo-first-order and pseudo-second-order models. The kinetic adsorption capacities q_t are measured in the same condition as those in Section 2.7.

2.9. Adsorption Isotherm

The adsorption behavior of MB onto LS₁P₉ was investigated using the Langmuir and Freundlich adsorption isotherm models. The prepared hydrogels were immersed in MB solutions with different concentrations for 48 h, and the equilibrium adsorption capacity (q_e) was calculated using Equation (3). The experimental data were fitted using the Langmuir isotherm model Equation (6) and the Freundlich isotherm model Equation (7).

$$q_e = \frac{q_{\max} K_L C_e}{1 + K_L C_e} \quad (6)$$

$$q_e = K_F C_e^{\frac{1}{n}} \quad (7)$$

where $q_{\max}$ is the theoretical maximum adsorption amount (mmol g⁻¹), and K_L and K_F are the Langmuir constant and the Freundlich constant (mmol¹⁻ⁿ·g⁻¹·L⁻ⁿ).

3. Results and Discussion

3.1. Synthesis of Hydrogels

The hydrogels were synthesized from lignin, poly(ethylene glycol) diglycidyl ether (PEGDGE), and sorbitol polyglycidyl ether (SPE) via an epoxide ring-opening in alkaline aqueous solutions at 40 °C. Network formation yields ether linkages between lignin hydroxyl groups and the epoxide groups of PEGDGE and SPE. The stoichiometry was controlled by fixing the hydroxyl-to-epoxide equivalent ratio (OH:epoxide) at 1:1.5, where epoxide equivalents include contributions from both PEGDGE and SPE. For formulation calculations, lignin was assigned an equivalent molecular weight of 178 g mol⁻¹ based on an average of three lignin monomeric units (Ref. [33]). Figure 1 schematically summarizes the synthesis strategy, and the mechanism is shown in Figure S1. Because SPE contains four epoxide groups per molecule and PEGDGE contains two, the feed compositions were calculated accordingly and are listed in Table 1.

Table 1. Compositions and corresponding sample names of the hydrogels.

Sample	SPE:PEGDGE (Epoxy Ratio) ¹	Lignin Mole (mmol) ²	SPE Mole (mmol) ³	SPE Amount (g)	PEGDGE Mole (mmol) ⁴	PEGDGE Amount (g)
LP	0:10	5.62	0	0	4.215	2.409
LS ₁ P ₉	1:9	5.62	0.211	0.162	3.794	2.168
LS ₂ P ₈	2:8	5.62	0.422	0.324	3.372	1.927
LS ₃ P ₇	3:7	5.62	0.632	0.486	2.951	1.686
LS ₄ P ₆	4:6	5.62	0.843	0.648	2.529	1.445
LS ₅ P ₅	5:5	5.62	1.054	0.811	2.108	1.204
LS ₆ P ₄	6:4	5.62	1.265	0.973	1.686	0.963

LS7P3	7:3	5.62	1.475	1.135	1.265	0.723
LS8P2	8:2	5.62	1.686	1.297	0.843	0.482
LS9P1	9:1	5.62	1.897	1.459	0.422	0.241
LS	10:0	5.62	2.108	1.621	0.000	0

¹ The molar ratio of hydroxyls from lignin to epoxy groups from PEGDGE and SPE was set at 1:1.5.

² 1 g lignin corresponds to 5.62 mmol hydroxyls according to the 178 g mol⁻¹ molecular weight from reference [33]. ³ Each SPE molecule contains 4 moles of epoxy groups; the total epoxy amount from PEGDGE and SPE is constant and 1.5 times 5.62 mmol. ⁴ Each PEGDGE molecule contains 2 moles of epoxy groups; the total epoxy amount from PEGDGE and SPE is constant and 1.5 times 5.62 mmol.

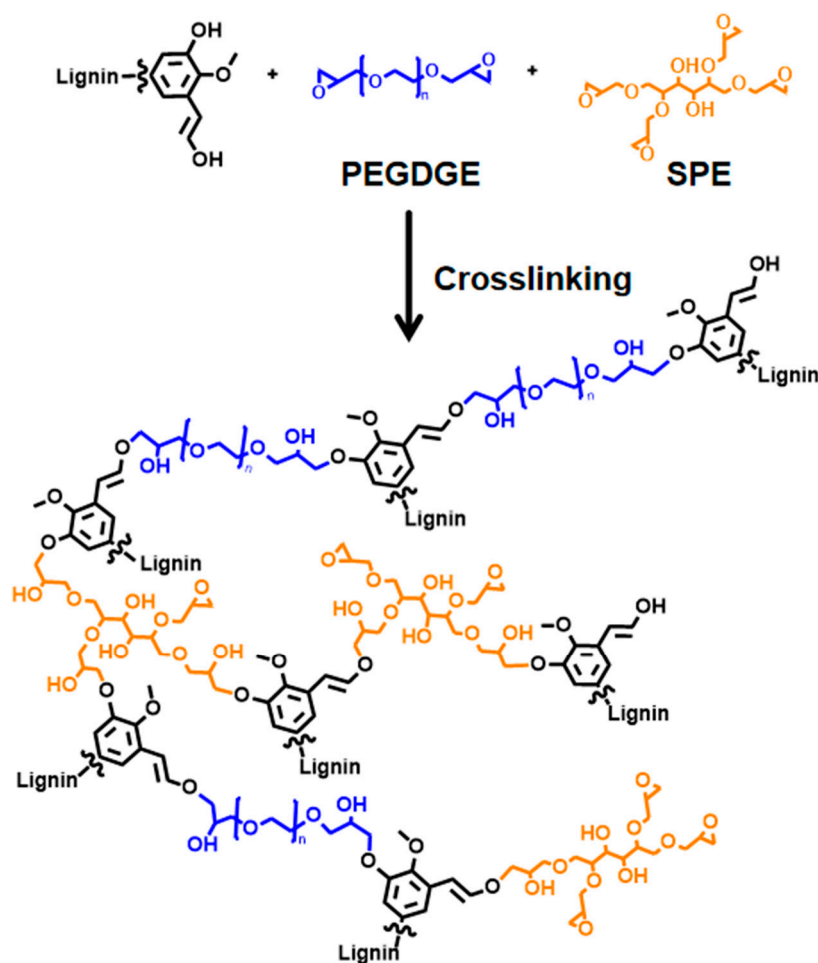


Figure 1. Schematic representation of hydrogel synthesis. SPE is drawn in orange, and PEGDGE is drawn in blue.

Lignin, PEGDGE, and the lignin–PEGDGE hydrogel (LP) were characterized by FTIR in Figure 2a, and the enlarged spectra are shown in Figure S2. PEGDGE exhibited characteristic absorption bands at 755 cm⁻¹ and 910 cm⁻¹, which were assigned to the epoxy groups. After crosslinking, these bands disappeared in LP, indicating consumption of the epoxide groups. In addition, a band at 1100 cm⁻¹ appeared, consistent with the formation of ether linkages. These results confirm successful crosslinking between PEGDGE and lignin. Similarly, lignin, SPE, and the lignin–SPE hydrogel (LS) were analyzed by FTIR in Figure 2b. SPE showed characteristic epoxide bands at 847 cm⁻¹ and 910 cm⁻¹. After crosslinking, these bands were absent in LS, suggesting that the epoxide groups were consumed. Overall, the FTIR spectra confirmed that SPE can successfully crosslink with lignin. Hydrogels containing both PEGDGE and SPE also showed successful crosslinking with lignin.

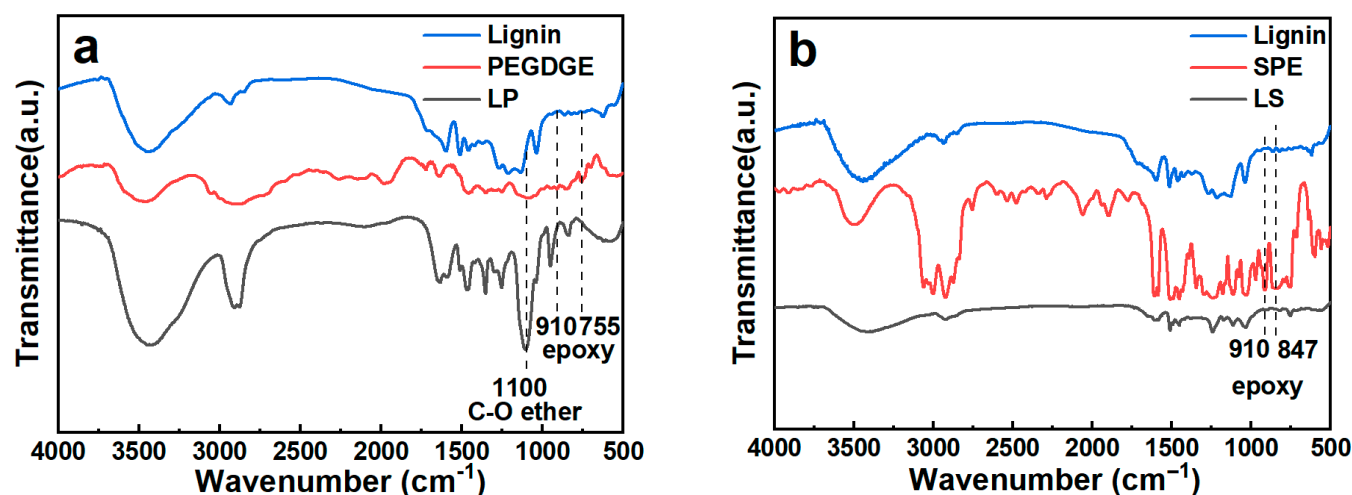


Figure 2. FTIR spectra of (a) lignin, PEGDGE, LP; (b) lignin, SPE, LS.

3.2. Swelling Studies

The swelling ratios of the hydrogels were systematically investigated, as shown in Figure 3. The swelling ratio decreased with increasing SPE content, because the higher epoxide functionality of SPE produced a denser crosslinked network that restricted network expansion upon hydration. These results indicate a clear relationship between cross-linker composition and the equilibrium swelling ratio [28,34]. Among all samples, LP exhibited the highest swelling ratio of 429.17%. The numerical values of swelling ratio and gel fractions are shown in Table S1.

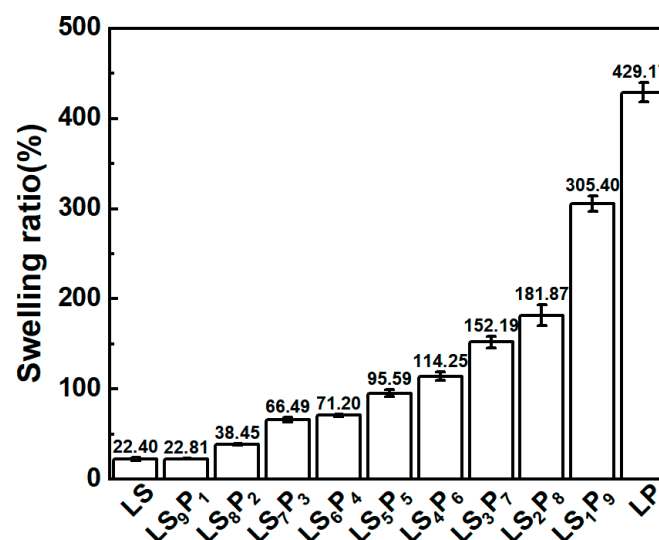


Figure 3. Swelling ratio of the hydrogels.

3.3. Rheological Properties

The storage modulus (G') and loss modulus (G'') were measured by oscillatory rheology. The comparison of G' and G'' for each sample is shown in Figure S3. For all samples, G' remained higher than G'' across the entire frequency range (0.1–20 Hz). This behavior indicates that the hydrogels were solid-like and predominantly elastic. Moreover, the $\tan\delta$ (G''/G') of eleven samples are shown in Figure S4, and all the numbers were lower than 1, indicating elastic characteristics. In addition, no crossover between G' and G'' was observed, suggesting that the networks were fully gelled and maintained elastic-dominated behavior throughout the frequency sweep. The G' of hydrogels are shown in Figure 4.

Among the eleven samples, LS exhibited the highest G' , indicating the most compact network structure and the highest crosslinking density. With the introduction of PEGDGE, flexible chain segments were incorporated into the network, and G' generally decreased with PEGDGE increasing. This trend suggests that increasing PEGDGE reduces network rigidity and strength, despite several formulations deviating to certain extents. The effect of PEGDGE is also reported elsewhere [35].

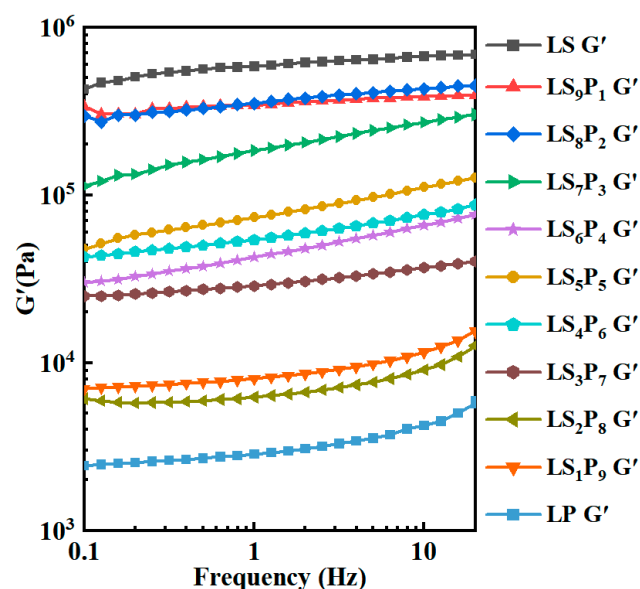


Figure 4. Frequency dependence of G' for hydrogels.

3.4. Mechanical Properties

The mechanical properties were evaluated by compression tests. From the stress–strain curves in Figure 5, the compressive strength at 40% strain, as well as toughness over 0–40% strain (calculated as the area under the stress–strain curve), were determined. Both metrics increased with increasing SPE content. As SPE is a four-functional epoxide crosslinker, higher SPE levels are expected to generate a denser network structure, thereby enhancing mechanical performance. At 40% strain, LS9P1 exhibited the best overall performance, with a compressive strength of 3.66 MPa and a toughness $416.84 \text{ kJ}\cdot\text{m}^{-3}$. The LS sample ranked second, followed by LS8P2. The numerical values are shown in Table 2.

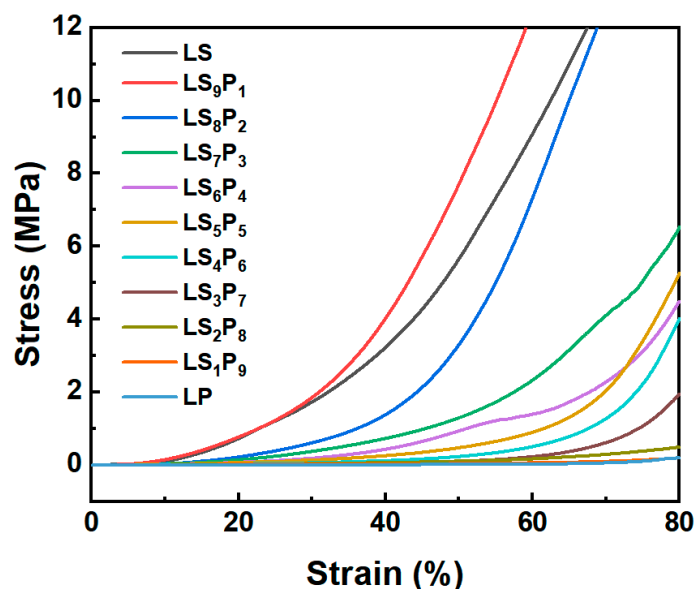


Figure 5. Compressive stress–strain curves of the hydrogels.**Table 2.** Compressive strength at 40% strain, and toughness over 0–40% strain.

Sample	Strength (MPa)	Toughness ($\text{kJ}\cdot\text{m}^{-3}$)
LP	0.0059 ± 0.0054	0.39 ± 0.45
LS ₁ P ₉	0.018 ± 0.011	2.66 ± 1.63
LS ₂ P ₈	0.049 ± 0.016	6.85 ± 2.63
LS ₃ P ₇	0.047 ± 0.013	5.88 ± 1.09
LS ₄ P ₆	0.094 ± 0.049	12.53 ± 5.77
LS ₅ P ₅	0.25 ± 0.03	31.82 ± 3.36
LS ₆ P ₄	0.43 ± 0.16	45.01 ± 17.00
LS ₇ P ₃	0.62 ± 0.13	68.33 ± 13.20
LS ₈ P ₂	1.19 ± 0.45	122.74 ± 56.55
LS ₉ P ₁	3.66 ± 0.50	416.84 ± 62.85
LS	3.23 ± 1.36	403.60 ± 245.75

Cyclic compression tests were performed to further evaluate the mechanical performance of the hydrogels. As shown in Figure 6, each sample was subjected to five loading–unloading cycles. In the first cycle, the hydrogels were compressed to the 40% strain. Upon unloading, the strain did not fully return to zero. For LS, LS₉P₁, LS₈P₂, and LS₇P₃, this behavior was attributed to fracture or irreversible damage, as evidenced by the corresponding stress–strain curves. This likely resulted from the high SPE content, which increased crosslink density and led to a more brittle network. In contrast, hydrogels with lower SPE content exhibited better recovery under repeated compression. With increasing cycle numbers, the stress–strain responses gradually stabilized. By the fourth and fifth cycles, the curves nearly overlapped, indicating that the mechanical response had reached a steady state.

To facilitate comparison among the eleven formulations in this stabilized regime, the fifth cycle is presented in Figure 7. The peak stress increased with SPE content, consistent with the trend observed in the non-cyclic compression results in Figure 5.

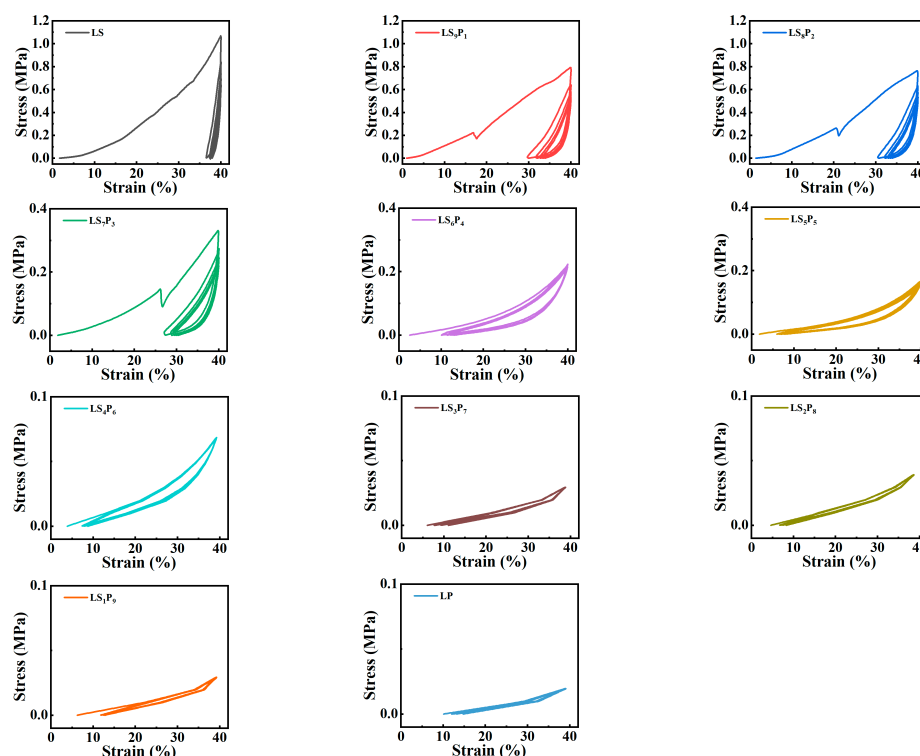
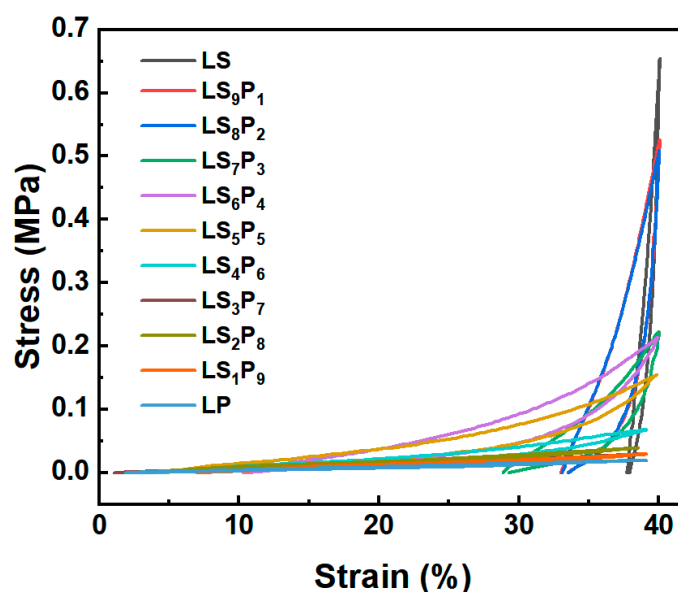


Figure 6. Cyclic compression stress–strain curves of hydrogels.**Figure 7.** The 5th cycle compression stress–strain curves of hydrogels.

3.5. Dye Adsorption Property

Since adsorption is related to swelling performance, and LS₁P₉ had a good swelling ratio of 305.40%, it was selected for the adsorption test. Adsorption was monitored over 0–2880 min. The relationship between contact time and MB removal efficiency is shown in Figure 8. The removal efficiency exceeded 50% within 120 min and reached approximately 72% after 2880 min. The adsorption reached 99% equilibrium at 30 h. In addition to removal efficiency, adsorption capacity is an important metric for evaluating adsorption performance. The equilibrium adsorption capacity (q_e) of LS₁P₉ toward MB was 6.99 mg g^{−1}. This reported q_e was not the best due to the low initial concentration and high adsorbent dosage limitation. But we observed an increase in the initial concentration, or reduced hydrogel made it harder to reach equilibrium. The adsorption isotherm is shown in Figure S5. For comparison, lignosulfonate-based ionic hydrogels have been reported to exhibit MB adsorption capacities of $q_e > 52$ mg g^{−1} [36], while starch-based hydrogels show $q_e < 2$ mg g^{−1} [37].

To enhance mass transfer, the hydrogel was cut into small pieces (~5 mm × 4 mm × 1.5 mm) prior to adsorption. In contrast, a single intact cylindrical specimen (diameter 2.5 cm, thickness 1.5 cm) was also tested. The intact specimen achieved 45% removal after 2880 min, with an equilibrium q_e of 3.53 mg g^{−1}. The cut pieces provided a 77.6-fold higher surface-area-to-volume ratio than the intact sample, enhancing external mass transfer. Moreover, based on the characteristic diffusion–time relationship ($t \propto L^2/D_{eff}$), reducing the characteristic length scale by cutting was expected to accelerate internal diffusion by approximately 100–278 times.

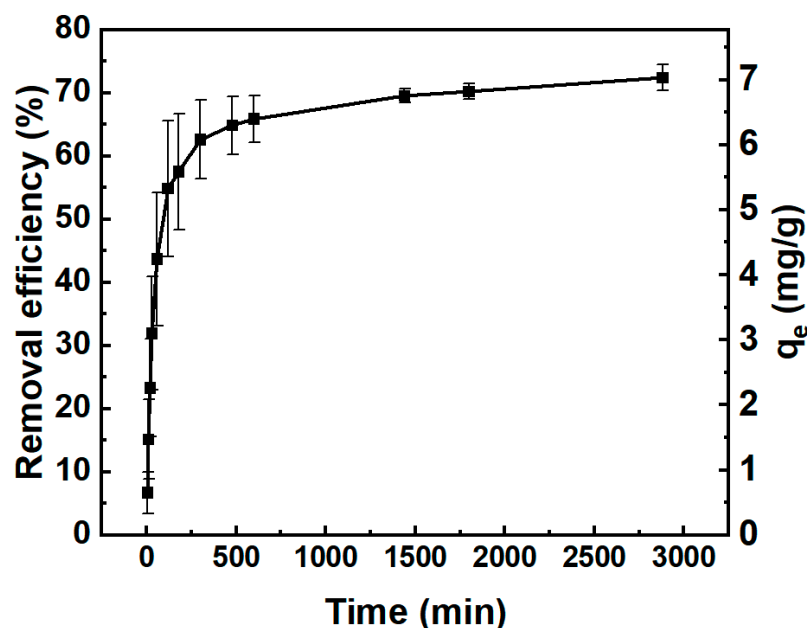


Figure 8. MB removal efficiency and adsorption capacity of LS1P9 hydrogel.

3.6. Adsorption Kinetics

Adsorption kinetics were analyzed using the adsorption time-uptake data and the equilibrium adsorption capacity q_e . The kinetic raw data was carried in the exact same condition as the experimental tests, including adsorbent mass, solution volume, initial concentration, temperature, agitation, and hydrogel geometry. The pseudo-first-order and pseudo-second-order kinetic models were applied, which are commonly associated with predominantly physisorption- and chemisorption-controlled processes, respectively. As shown in Figure 9 and Table 3, the pseudo-first-order kinetic model gave an R^2 of 0.976, whereas the pseudo-second-order model yielded a higher R^2 of 0.998, indicating a better fit. In addition, the equilibrium q_e calculated from the pseudo-second-order model was close to the experimental q_e . Therefore, MB adsorption behavior was better described by the pseudo-second-order kinetic model, suggesting that chemisorption played a dominant role in the adsorption process [38].

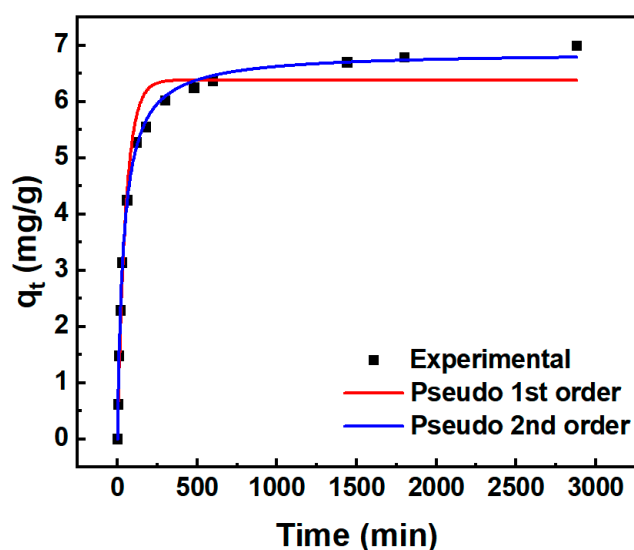


Figure 9. Pseudo-first-order and pseudo-second-order kinetic models of MB adsorption kinetics.

Table 3. Characteristic parameters of the adsorption kinetics.

Pseudo-first-order	R^2	0.976
	k_1 (min ⁻¹)	0.019
	q_e (cal) (mg g ⁻¹)	6.382
Pseudo-second-order	R^2	0.998
	k_2 (g mg ⁻¹ min ⁻¹)	0.004
	q_e (cal) (mg g ⁻¹)	6.879

4. Conclusions

Sustainable lignin-based hydrogels were prepared by crosslinking lignin with PEGDGE and SPE. Lignin and SPE functioned as crosslinking components, while PEGDGE provides introduced ductile soft segments into network. Increasing crosslinker content enhanced the mechanical properties of the hydrogels. Based on rheological and compression measurements, LS₉P₁ exhibited the best overall mechanical performance, with a compressive strength of 3.66 MPa and a toughness of 416.84 kJ·m⁻³ at 40% strain. The swelling test showed that lower crosslinker content led to higher equilibrium swelling. Accordingly, LS₁P₉ was selected for methylene blue adsorption from aqueous solution, achieving a maximum removal efficiency of approximately 72%. Kinetic analysis indicated that the adsorption process followed a pseudo-second-order model, suggesting that chemisorption was the dominant adsorption mechanism. Overall, these structure-property-tunable lignin-PEGDGE-SPE hydrogels show potential for wastewater treatment applications.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/polym18040489/s1>. Figure S1. Schematic illustration of the synthesis mechanism of LP, LS_nP_n, and LS. Figure S2. Enlarged FTIR spectra of (a) lignin, PEGDGE, and LP, and (b) lignin, SPE, and LS. Figure S3. Rheological properties of the hydrogels. Figure S4. Frequency dependence of tan δ (G''/G') for the hydrogels. Figure S5. Langmuir and Freundlich isotherm models for MB adsorption. Table S1. Gel fraction and swelling ratio of the hydrogels. Table S2. Characteristic parameters of the adsorption isotherms. Geometry-based analysis of mass-transfer limitation.

Author Contributions: Conceptualization, H.L.; methodology, Y.W.; software, Y.W.; validation, Y.W.; resources, Y.-P.H.; data curation, Y.W.; writing—original draft preparation, Y.W.; writing—review and editing, H.L.; visualization, G.G.; supervision, H.L.; project administration, H.L.; funding acquisition, H.L. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Science Foundation of China, grant number 52103005; the Liaoning Revitalization Talents Program, grant number XLYC2203072; the Fundamental Research Foundation of the Liaoning Education Department, grant number LJ212410148041; the Liaoning Province Science and Technology Joint Funding Program 2025-MSLH-449; and the Natural Science Foundation of Ningbo, grant number 2024J009.

Institutional Review Board Statement: Not applicable.

Data Availability Statement: All data generated or analyzed during this study are included in this published article.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

FDA	Food and Drug Administration
DMF	N,N-Dimethylformamide

References

1. Akhtar, M.F.; Hanif, M.; Ranjha, N.M. Methods of synthesis of hydrogels: A review. *Saudi Pharm. J.* **2016**, *24*, 554–559. <https://doi.org/10.1016/j.jsps.2015.03.022>.
2. Rico-García, D.; Ruiz-Rubio, L.; Pérez-Alvarez, L.; Hernández-Olmos, S.L.; Guerrero-Ramírez, G.L.; Vilas-Vilela, J.L. Lignin-based hydrogels: Synthesis and applications. *Polymers* **2020**, *12*, 81. <https://doi.org/10.3390/polym12010081>.
3. Kim, M.S.; Heo, J.W.; Xia, Q.; Oh, D.H.; Kim, J.W.; Kim, Y.S. Facile fabrication of lignin crosslinked hydrogel for cationic dye adsorption and antioxidant. *BioResources* **2024**, *19*, 5316–5337. <https://doi.org/10.15376/biores.19.3.5316-5337>.
4. Zhu, T.; Ni, Y.; Biesold, G.M.; Cheng, Y.; Ge, M.; Li, H.; Huang, J.; Lin, Z.; Lai, Y. Recent advances in conductive hydrogels: Classifications, properties, and applications. *Chem. Soc. Rev.* **2023**, *52*, 473–509. <https://doi.org/10.1039/D2CS00173J>.
5. Li, W.; Liu, J.; Wei, J.; Yang, Z.; Ren, C.; Li, B. Recent Progress of Conductive Hydrogel Fibers for Flexible Electronics: Fabrications, Applications, and Perspectives. *Adv. Funct. Mater.* **2023**, *33*, 2213485. <https://doi.org/10.1002/adfm.202213485>.
6. Singh, T.R.R.; Garland, M.J.; Migalska, K.; Salvador, E.C.; Shaikh, R.; McCarthy, H.O.; Woolfson, A.D.; Donnelly, R.F. Influence of a Pore-Forming Agent on Swelling, Network Parameters, and Permeability of Poly(ethylene glycol)-Crosslinked Poly(methyl Vinyl Ether-co-Maleic Acid) Hydrogels: Application in Transdermal Delivery Systems. *J. Appl. Polym. Sci.* **2012**, *125*, 2680–2694. <https://doi.org/10.1002/app.36524>.
7. Peppas, N.A.; Hilt, J.Z.; Khademhosseini, A.; Langer, R. Hydrogels in Biology and Medicine: From Molecular Principles to Bionanotechnology. *Adv. Mater.* **2006**, *18*, 1345–1360. <https://doi.org/10.1002/adma.200501612>.
8. Larrañeta, E.; Lutton, R.E.M.; Brady, A.J.; Vicente-Pérez, E.M.; Woolfson, A.D.; Thakur, R.R.S.; Donnelly, R.F. Microwave-Assisted Preparation of Hydrogel-Forming Microneedle Arrays for Transdermal Drug Delivery Applications. *Macromol. Mater. Eng.* **2015**, *300*, 586–595. <https://doi.org/10.1002/mame.201500016>.
9. Larrañeta, E.; Barturen, L.; Ervine, M.; Donnelly, R.F. Hydrogels Based on Poly(methyl Vinyl Ether-co-Maleic Acid) and Tween 85 for Sustained Delivery of Hydrophobic Drugs. *Int. J. Pharm.* **2018**, *538*, 147–158. <https://doi.org/10.1016/j.ijpharm.2018.01.025>.
10. Mei, X.; Li, J.; Wang, Z.; Zhu, D.; Huang, K.; Hu, S.; Popowski, K.D.; Cheng, K. An Inhaled Bioadhesive Hydrogel to Shield Non-Human Primates from SARS-CoV-2 Infection. *Nat. Mater.* **2023**, *22*, 903–912. <https://doi.org/10.1038/s41563-023-01475-7>.
11. Gale, E.C.; Powell, A.E.; Roth, G.A.; Meany, E.L.; Yan, J.; Ou, B.S.; Grosskopf, A.K.; Adamska, J.; Picece, V.C.T.M.; D’Aquino, A.I.; et al. Hydrogel-Based Slow Release of a Receptor-Binding Domain Subunit Vaccine Elicits Neutralizing Antibody Responses Against SARS-CoV-2. *Adv. Mater.* **2021**, *33*, 2104362. <https://doi.org/10.1002/adma.202104362>.
12. Lv, Z.; Zheng, Y.; Zhou, H.; Pan, Z.; Li, C.; Dai, L.; Zhang, M.; Si, C. Hydrothermal Method-Assisted Synthesis of Self-Crosslinked All-Lignin-Based Hydrogels. *Int. J. Biol. Macromol.* **2022**, *216*, 670–675. <https://doi.org/10.1016/j.ijbiomac.2022.07.003>.
13. Clarke, R.W.; Rognerud, E.G.; Puente-Urbina, A.; Barnes, D.; Murdy, P.; McGraw, M.L.; Newkirk, J.M.; Beach, R.; Wrubel, J.A.; Hamernik, L.J.; et al. Manufacture and Testing of Biomass-Derivable Thermosets for Wind Blade Recycling. *Science* **2024**, *385*, 854–860. <https://doi.org/10.1126/science.adp5395>.
14. Zhu, H.; Li, Z.; Yang, J. A Novel Composite Hydrogel for Adsorption and Photocatalytic Degradation of Bisphenol A by Visible Light Irradiation. *Chem. Eng. J.* **2018**, *334*, 1679–1690. <https://doi.org/10.1016/j.cej.2017.11.148>.
15. Bhattarai, N.; Gunn, J.; Zhang, M. Chitosan-Based Hydrogels for Controlled, Localized Drug Delivery. *Adv. Drug Deliv. Rev.* **2010**, *62*, 83–99. <https://doi.org/10.1016/j.addr.2009.07.019>.
16. Baker, B.A.; Murff, R.L.; Milam, V.T. Tailoring the Mechanical Properties of Polyacrylamide-Based Hydrogels. *Polymer* **2010**, *51*, 2207–2214. <https://doi.org/10.1016/j.polymer.2010.02.022>.
17. Shar, A.S.; Wang, N.; Chen, T.; Zhao, X.; Weng, Y. Development of PLA/Lignin Bio-Composites Compatibilized by Ethylene Glycol Diglycidyl Ether and Poly(ethylene Glycol) Diglycidyl Ether. *Polymers* **2023**, *15*, 4049. <https://doi.org/10.3390/polym15204049>.
18. Viretto, A.; Galy, J. Development of Biobased Epoxy Matrices for the Preparation of Green Composite Materials for Civil Engineering Applications. *Macromol. Mater. Eng.* **2018**, *303*, 1700521. <https://doi.org/10.1002/mame.201700521>.
19. Shibata, M.; Nakai, K. Preparation and Properties of Biocomposites Composed of Bio-Based Epoxy Resin, Tannic Acid, and Microfibrillated Cellulose. *J. Polym. Sci. Part B Polym. Phys.* **2010**, *48*, 425–433. <https://doi.org/10.1002/polb.21903>.

20. Shibata, M.; Yoshihara, S.; Yashiro, M.; Ohno, Y. Thermal and Mechanical Properties of Sorbitol-Based Epoxy Resin Cured with Quercetin and the Biocomposites with Wood Flour. *J. Appl. Polym. Sci.* **2013**, *128*, 2753–2758. <https://doi.org/10.1002/app.38438>.
21. Zhang, Y.; Liu, X.; Wan, M.; Zhu, Y.; Zhang, K. Recent Development of Functional Bio-Based Epoxy Resins. *Molecules* **2024**, *29*, 4428. <https://doi.org/10.3390/molecules29184428>.
22. Rochester, J.R. Bisphenol A and Human Health: A Review of the Literature. *Reprod. Toxicol.* **2013**, *42*, 132–155. <https://doi.org/10.1016/j.reprotox.2013.08.008>.
23. Meng, Y.; Lu, J.; Cheng, Y.; Li, Q.; Wang, H. Lignin-Based Hydrogels: A Review of Preparation, Properties, and Application. *Int. J. Biol. Macromol.* **2019**, *135*, 1006–1019. <https://doi.org/10.1016/j.ijbiomac.2019.05.198>.
24. Yang, W.; Ding, H.; Puglia, D.; Kenny, J.M.; Liu, T.; Guo, J.; Wang, Q.; Ou, R.; Xu, P.; Ma, P.; et al. Bio-Renewable Polymers Based on Lignin-Derived Phenol Monomers: Synthesis, Applications, and Perspectives. *SusMat* **2022**, *2*, 535–568. <https://doi.org/10.1002/sus2.87>.
25. Nikafshar, S.; Wang, J.; Dunne, K.; Sangthonganotai, P.; Nejad, M. Choosing the Right Lignin to Fully Replace Bisphenol A in Epoxy Resin Formulation. *ChemSusChem* **2021**, *14*, 1184–1195. <https://doi.org/10.1002/cssc.202002729>.
26. Xue, M.; Hu, S.; Zhang, S.; Ou, J.; Chen, H.; Liao, Y.; Yue, F. Flexible and Heat-Resisting Lignin-Based Epoxy Resins by Hardwood Kraft Low-Molecular-Weight Lignin as a Sustainable Substitute for Bisphenol A. *ACS Sustain. Chem. Eng.* **2023**, *11*, 16774–16784. <https://doi.org/10.1021/acssuschemeng.3c04898>.
27. Oveissi, F.; Naficy, S.; Le, T.Y.L.; Fletcher, D.F.; Dehghani, F. Tough and Processable Hydrogels Based on Lignin and Hydrophilic Polyurethane. *ACS Appl. Bio Mater.* **2018**, *1*, 2073–2081. <https://doi.org/10.1021/acsabm.8b00546>.
28. Domínguez-Robles, J.; Peresin, M.S.; Tamminen, T.; Rodríguez, A.; Larrañeta, E.; Jääskeläinen, A.-S. Lignin-Based Hydrogels with “Super-Swelling” Capacities for Dye Removal. *Int. J. Biol. Macromol.* **2018**, *115*, 1249–1259. <https://doi.org/10.1016/j.ijbiomac.2018.04.044>.
29. Ralph, J.; Lundquist, K.; Brunow, G.; Lu, F.; Kim, H.; Schatz, P.F.; Marita, J.M.; Hatfield, R.D.; Ralph, S.A.; Christensen, J.H.; et al. Lignins: Natural Polymers from Oxidative Coupling of 4-Hydroxyphenylpropanoids. *Phytochem. Rev.* **2004**, *3*, 29–60. <https://doi.org/10.1023/B:PHYT.0000047809.65444.a4>.
30. Heo, J.W.; Kim, M.S.; Oh, D.H.; Kim, Y.S. Innovative construction of amine-functionalized lignin-based hydrogel for ultrafast and selective dye remediation. *Chem. Eng. J.* **2024**, *498*, 155506. <https://doi.org/10.1016/j.cej.2024.155506>.
31. Meng, X.; Scheidemantle, B.; Li, M.; Wang, Y.-Y.; Zhao, X.; Toro-González, M.; Singh, P.; Pu, Y.; Wyman, C.E.; Ozcan, S.; et al. Synthesis, characterization, and utilization of a lignin-based adsorbent for effective removal of azo dye from aqueous solution. *ACS Omega* **2020**, *5*, 2865–2877. <https://doi.org/10.1021/acsomega.9b03717>.
32. Zhao, W.; Cui, Y.; Zhou, S.; Ye, J.; Sun, J.; Liu, X. Rapid adsorption of dyes from aqueous solutions by modified lignin derived superparamagnetic composites. *J. Mol. Struct.* **2022**, *1261*, 132954. <https://doi.org/10.1016/j.molstruc.2022.132954>.
33. Milotskyi, R.; Szabó, L.; Takahashi, K.; Bliard, C. Chemical Modification of Plasticized Lignins Using Reactive Extrusion. *Front. Chem.* **2019**, *7*, 633. <https://doi.org/10.3389/fchem.2019.00633>.
34. Liu, H.; Li, X.; Pan, Z.; Dai, L.; Zhang, M.; Shen, F.; Si, C. Lignin-Based Plugging Hydrogel with High-Temperature Resistance and Adjustable Gelation. *Adv. Compos. Hybrid Mater.* **2025**, *8*, 111. <https://doi.org/10.1007/s42114-024-01132-w>.
35. Teng, X.; Xu, H.; Song, W.; Shi, J.; Xin, J.; Hiscox, W.C.; Zhang, J. Preparation and Properties of Hydrogels Based on PEGylated Lignosulfonate Amine. *ACS Omega* **2017**, *2*, 251–259. <https://doi.org/10.1021/acsomega.6b00296>.
36. Mondal, A.K.; Wu, S.; Xu, D.; Zou, Q.; Chen, L.; Huang, L.; Huang, F.; Ni, Y. Preparation of Lignosulfonate Ionic Hydrogels for Supercapacitors, Sensors and Dye Adsorbent Applications. *Int. J. Biol. Macromol.* **2021**, *187*, 189–199. <https://doi.org/10.1016/j.ijbiomac.2021.07.021>.
37. Mohammadzadeh, F.; Golshan, M.; Haddadi-Asl, V.; Salami-Kalajahi, M. Adsorption Kinetics of Methylene Blue from Wastewater Using pH-Sensitive Starch-Based Hydrogels. *Sci. Rep.* **2023**, *13*, 11900. <https://doi.org/10.1038/s41598-023-39241-z>.
38. Liu, P.; Lyu, J.; Bai, P. Synthesis of Mixed Matrix Membrane Utilizing Robust Defective MOF for Size-Selective Adsorption of Dyes. *Sep. Purif. Technol.* **2024**, *354*, 128672. <https://doi.org/10.1016/j.seppur.2024.128672>.

Disclaimer/Publisher’s Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.