# Supplementary Information

**High-strength, Self-healable, Transparent Castor-Oil-Based Waterborne Polyurethane Barrier Coatings Enabled by Dynamic Acylhydrazone Co-monomer**

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Table S1. Overview of Biomass-based coating for paper substrates

| Biopolymers | Synthesis route | Method of coating | Barrier Properties | | Ref |
|---|---|---|---|---|---|
| | | | WVTR (g.mm/m²/day) | Cobb (g/m²) | |
| ***Nanocellulose/ Chitosan*** | CNF stabilized Pickering emulsion with chitosan | Rod coating | 3.25 | 13.5 | [1] |
| ***Cellulose*** | Cellulose stearoyl esters | Rod coating (Solvent: Toluene) | 0.87 | Not described | [2] |
| ***Lignin*** | Fatty acid chloride modification | Rod coating (Solvent: Acetone) | 2.54 | Not described | [3] |
| ***Starch*** | Starch gelatinization and montmorillonite | Rod coating | 34.5 | Not described | [4] |
| ***Chitosan*** | Chitosan-based cardanol glycidyl ether | Rod coating | 17.78-19.46 | 7.9-15.6 | [5] |
| ***Hemicellulose*** | Esterification Hemicellulose-graft-lauric acid | Spray coating | 5.05 | Not described | [6] |
| ***Tung oil*** | Tung oil and photoinitiator | Rod coating (UV-curing) | Not described | 17.0 | [7] |
| ***Castor oil*** | Anionic waterborne polyurethane | Dip coating, roller coating | 0.98 and 2.21 | 0.95~2.53 | This work |

| | | |
|---|---|---|
| dispersion | (Water) | |

Table S1 lists the biomaterials used for paper-based coatings in recent years, such as cellulose nanofibrils, chitosan, starch, lignin, hemicellulose and plant oil. Compared with some chemically modified bio-based coating, our castor-oil-based waterborne polyurethane coating uses water as the primary solvent, avoiding the use of organic reagents as ingredients during the coating process, which is more in line with the principle of green chemistry. This waterborne coating has excellent storage stability at room temperature, beneficial for continuous large-scale production. Moreover, the coated paper exhibits competitive barrier properties, with a significant reduction in WVTR and the lowest Cobb value, indicates that our coating strategy has significant advantages in developing environmental and high-performance paper-based barrier coatings.

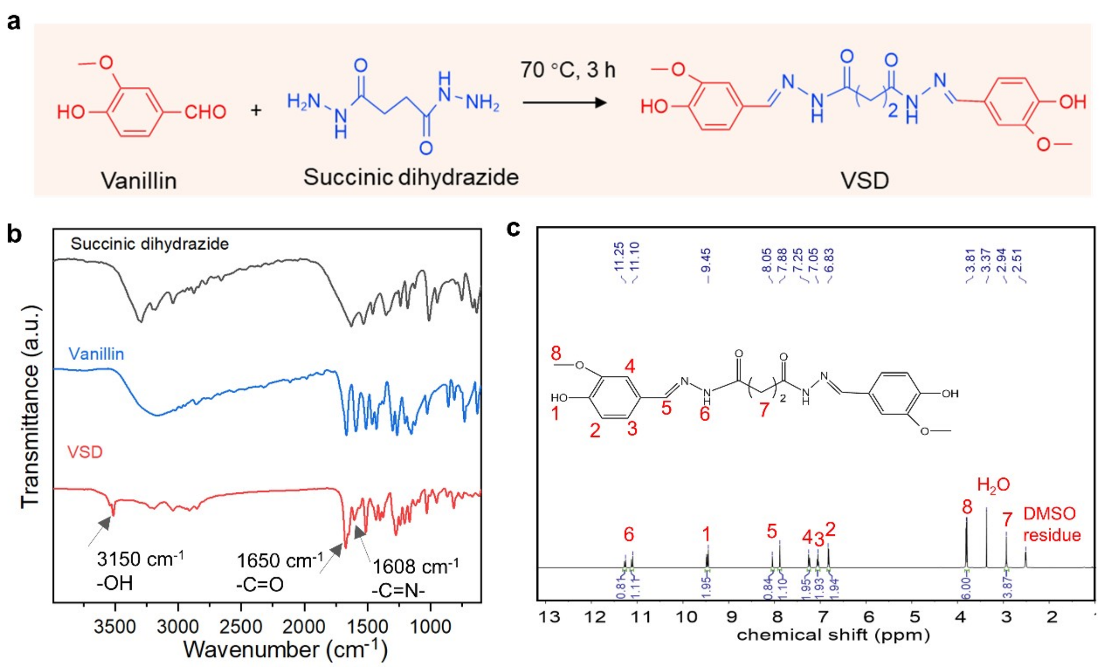


**Fig. S1** a) Synthetic routes for VSD monomer. b) FT-IR spectra of succinic dihydrazide, vanillin and acylhydrazones diol. c) $^{1}$H NMR spectrum of VSD.

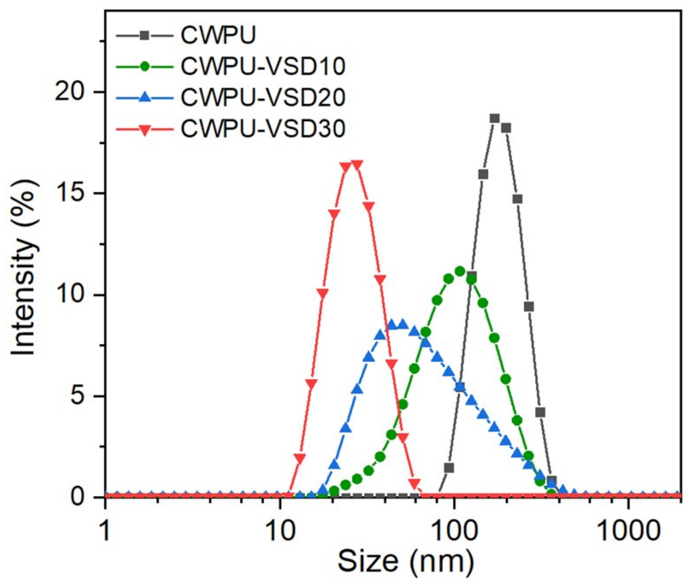


**Fig. S2** The particle size distribution profiles of dispersion samples.

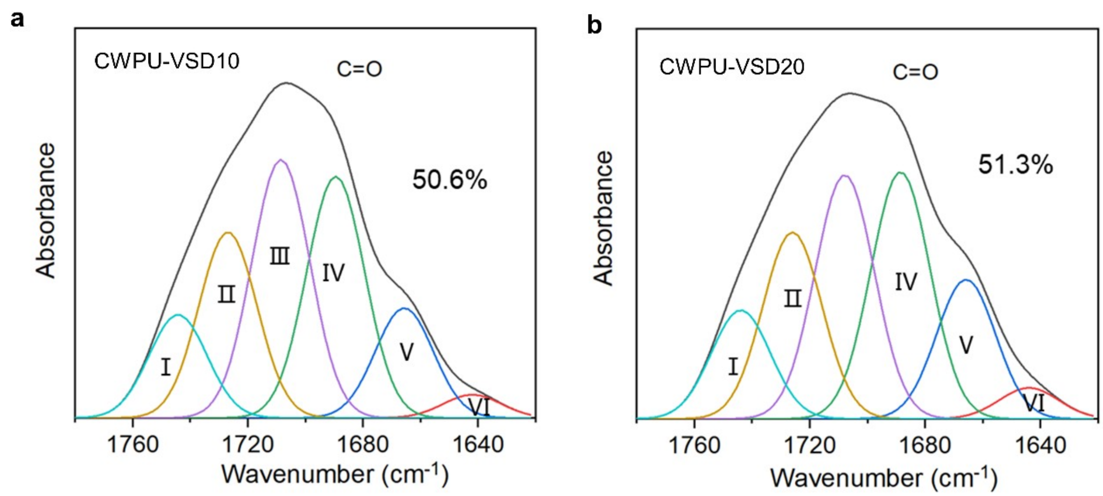


**Fig. S3** Curve-fitting results of carbonyl region in the FTIR spectra for a) CWPU-VSD10 and b) CWPU-VSD20.

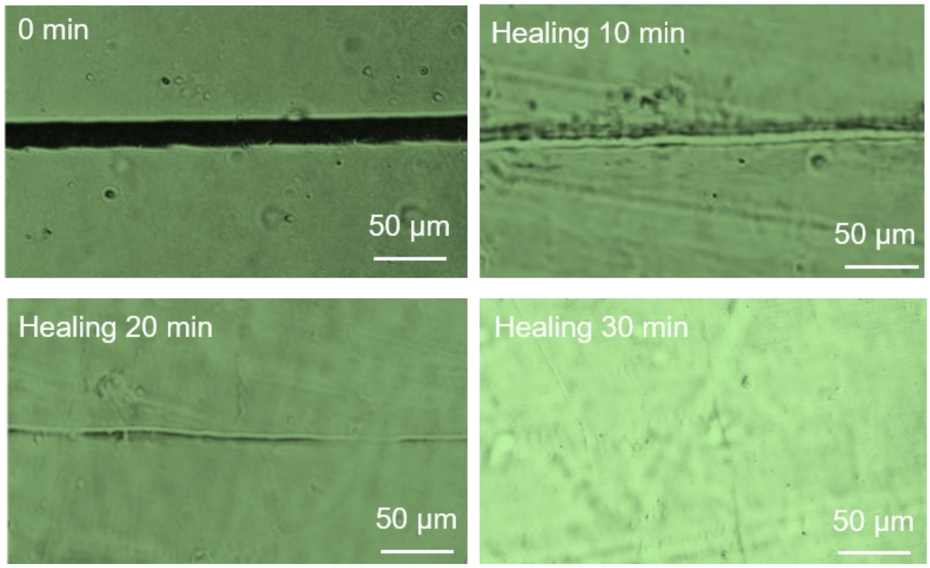


**Fig. S4** Optical microscopy images showing the scratch repair process

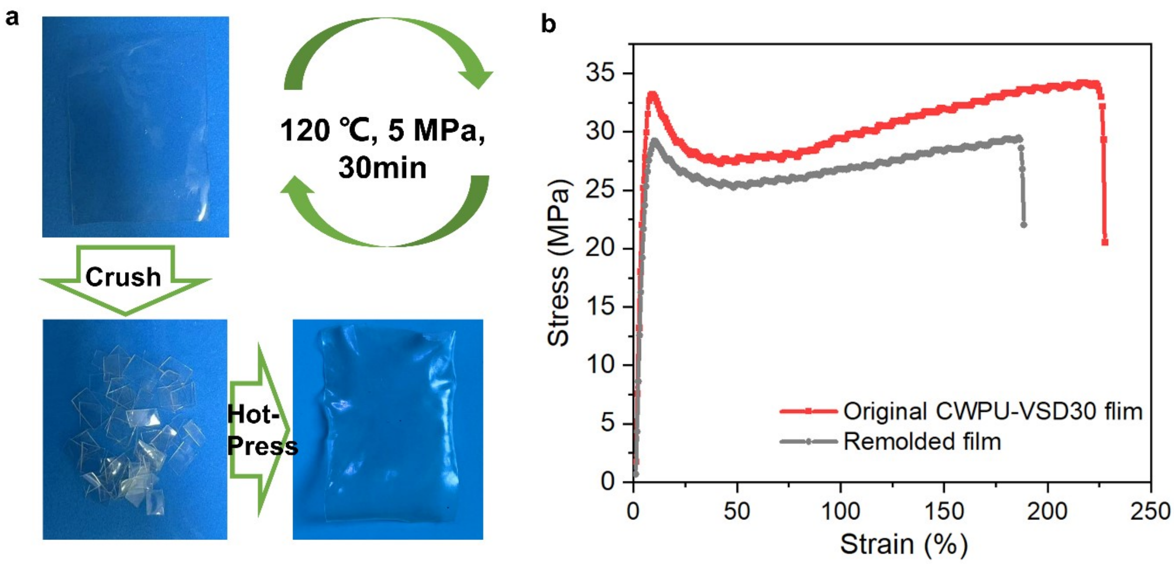


**Fig. S5** a) Photos showing the hot-pressing process. b) Stress–strain curve of CWPU-VSD30 before and after remodeling.

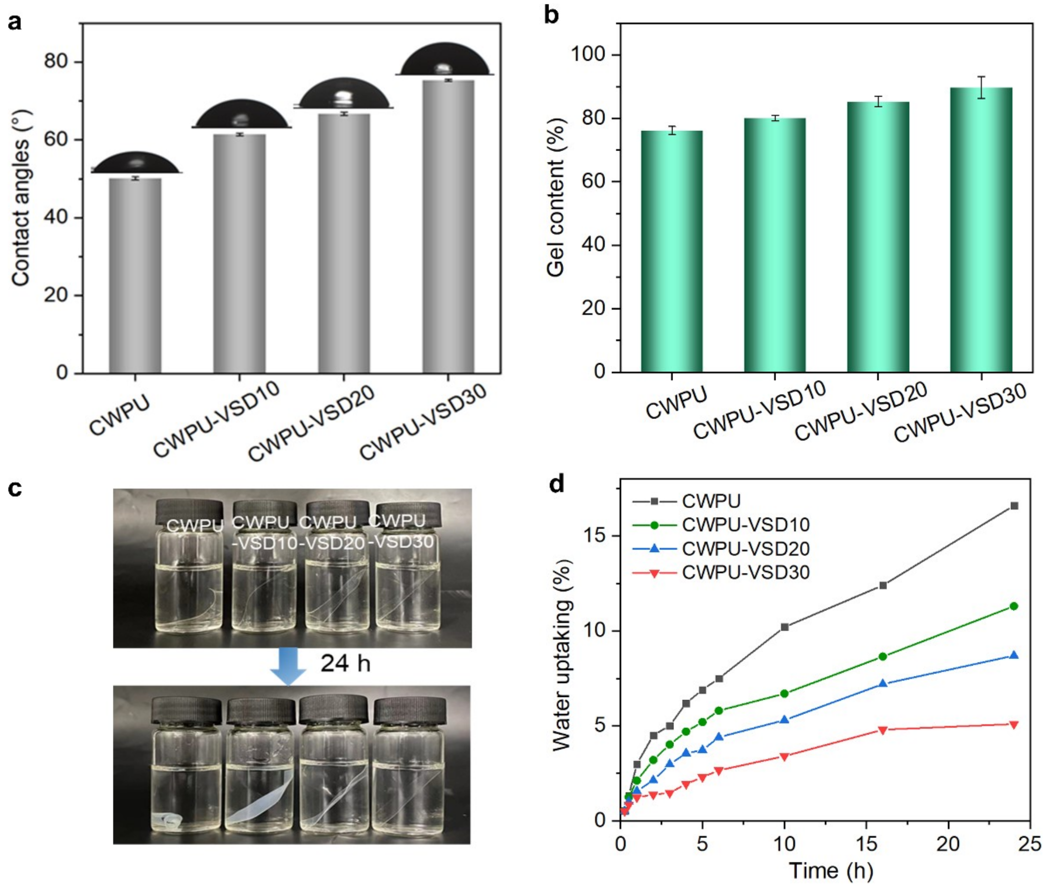


**Fig. S6** a) Water contact angle of films. b) The gel fraction of sample films. c) Photos showing the water-resistance of films. d) Water uptake curves of sample films in 24 h.

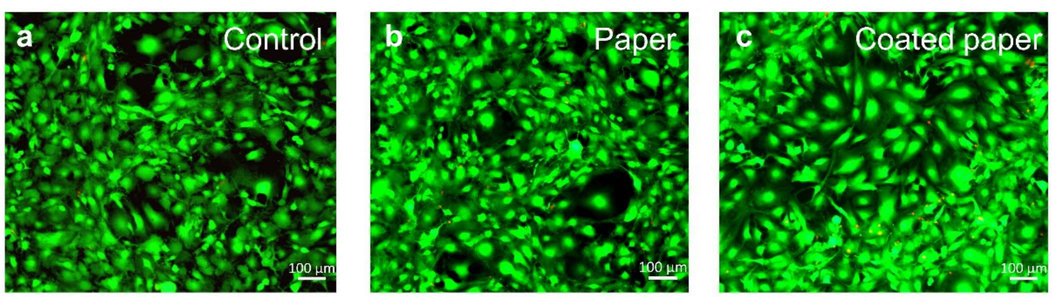


**Fig. S7** Images of a live/dead stain of a) control cells, b) paper and c) Coated paper (all cells are green, dead cells are red).

**Reference**

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