Supplementary Information (SI) for Green Chemistry.
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Supplementary Information

Dynamic Covalent Design of Keratin Sizes for Sustainable and Smart Yarn Coating

Jing Meng[12], Ruiyan Ni[12], Ziyu Zhou[12], Jiawen Zhang[12], Narendra Reddy[3], Xiaohui Mao[4], Wenfeng Hu[5], Xiaoyun Xu[6], Xian Li[12], Jinlian Hu [6*], Yi Zhao [12*]

[1] Shanghai Frontiers Science Center of Advanced Textiles, Donghua University, Shanghai 201620, China

[2] Engineering Research Center of Technical Textiles, Ministry of Education, Donghua University, Shanghai 201620, China

[3] Center for Incubation, Innovation, Research and Consultancy, Jyothy Institute of Technology, Bengaluru, India

[4] College of Materials Science and Engineering, Donghua University, 2999 North Renmin Road, Shanghai 201620, PR China

[5] School of Fashion Engineering Central Laboratory, Shanghai University of Engineering Science, Shanghai 201620, China

[6] Department of Biomedical Engineering, City University of Hong Kong, Hong Kong S.A.R, 999077, China.

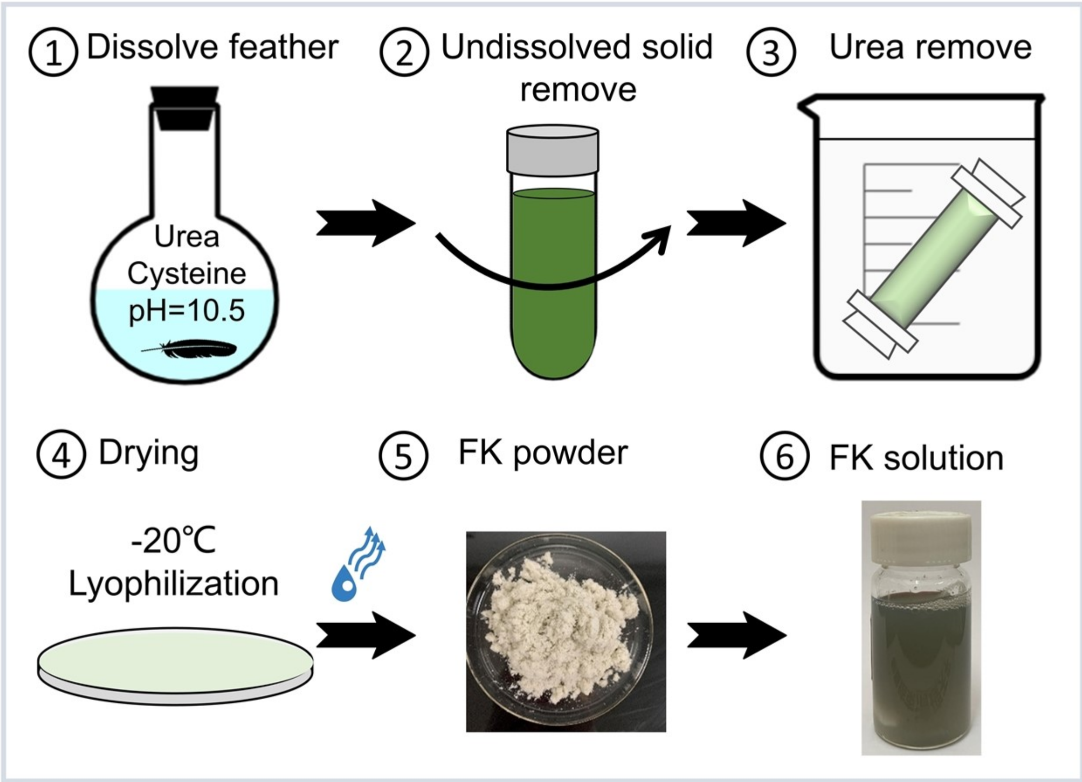


Fig. S1 Extraction protocol of keratin from duck feather.

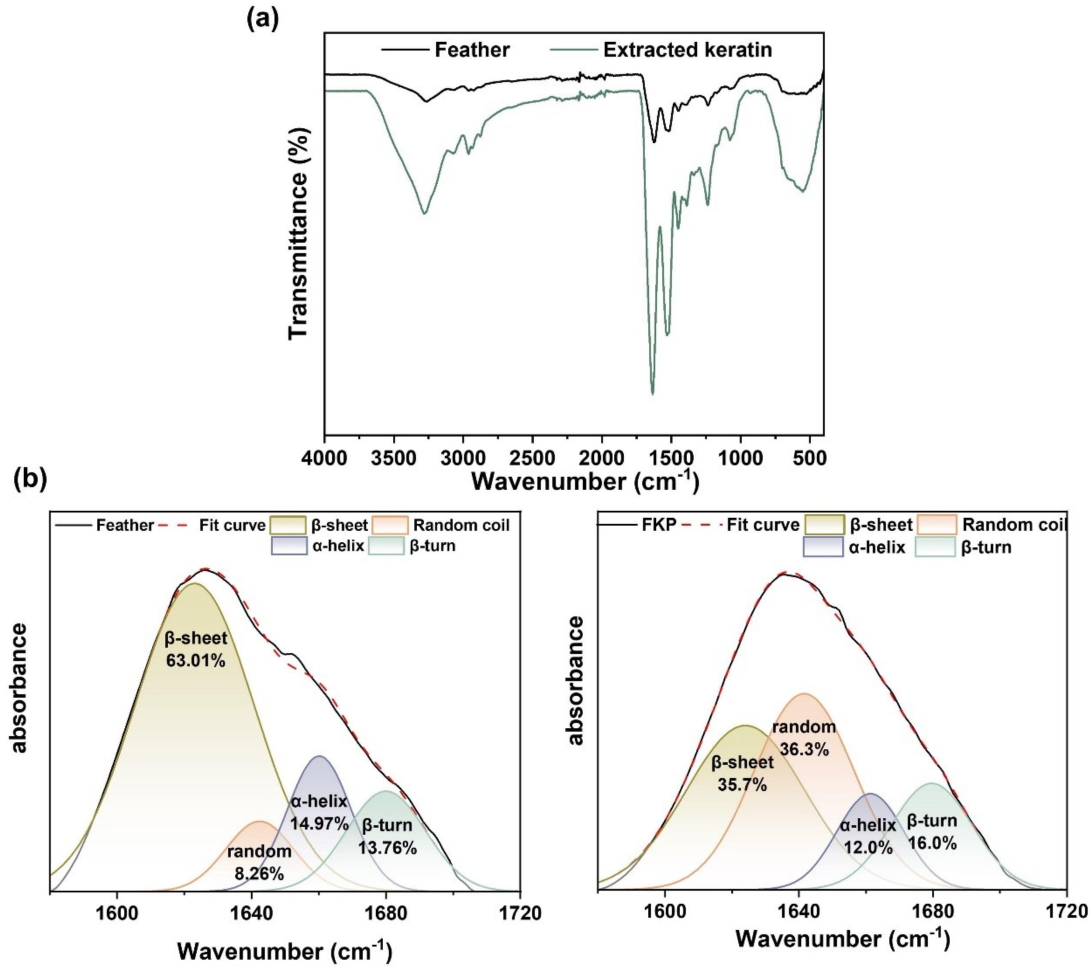


Fig. S2 FTIR and deconvolution analysis of amide Ⅰ region of the feather and FK powder, showing most of the secondary structure of feathers in β-sheet, while the regenerated keratin in random coil. This indicates that the extraction process disrupts the ordered structure of the feathers.

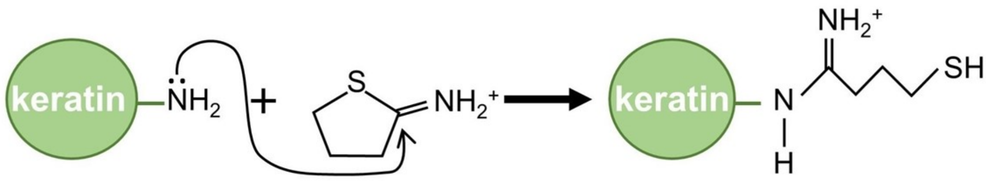


Fig. S3 Reaction mechanism of Traut's reagent modification. The TR reagent provides a thiol group after reacting with a primary amine.

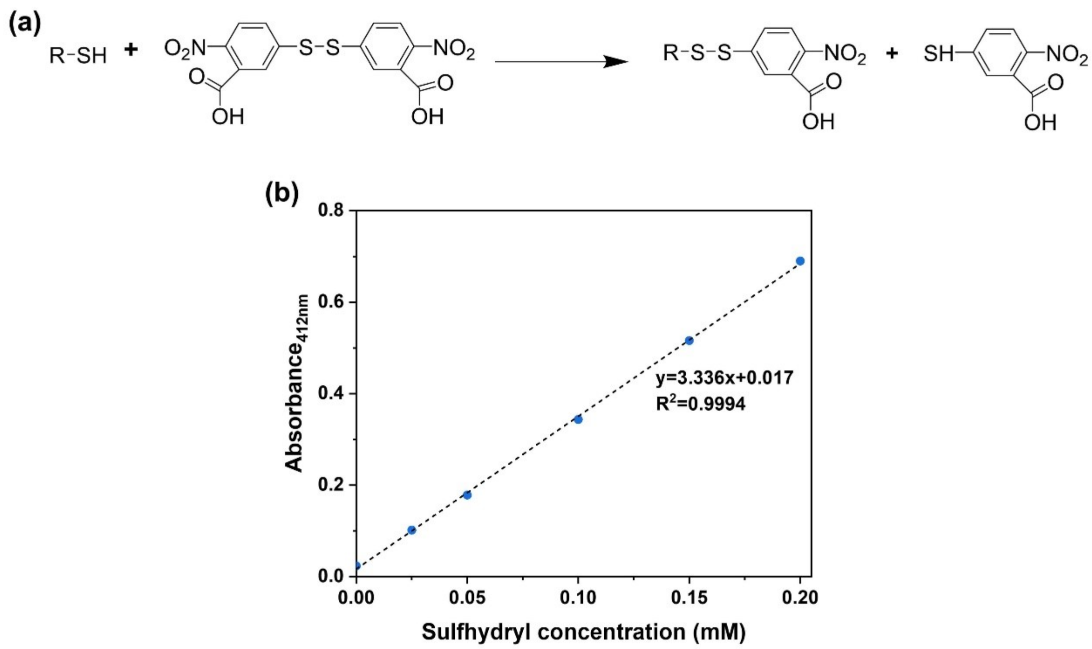
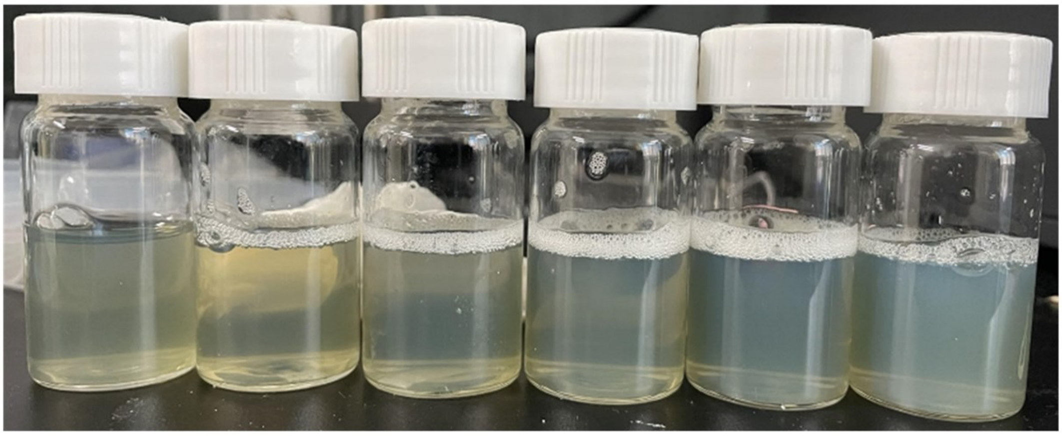


Fig. S4 (a) Schematic diagram of the reaction principle of DTNB to detect the amount of sulfhydryl groups. (b) Standard curve of the amount of sulfhydryl groups contained in cysteine. According to the absorbance of the known concentration of sulfhydryl group at 412nm, the function relationship between sulfhydryl group amount and absorbance is obtained after fitting.



FK T2-FK T3-FK T4-FK T5-FK T6-FK

Fig. S5 The actual picture of slurry modified with different dose of TR. The transparency of the slurry progressively decreases.

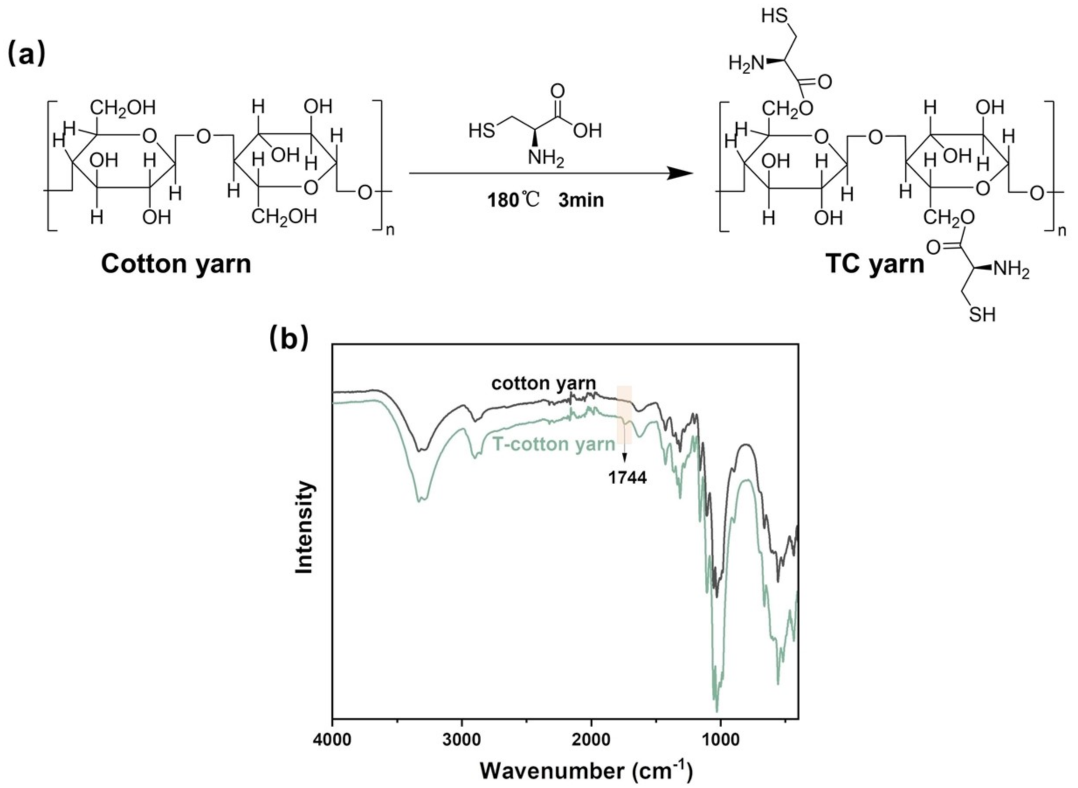


Fig. S6 The thiolation of cellulose yarns. (a) Reaction mechanism between cellulose and L-cysteine and (b) FTIR spectroscopy analysis of cotton yarn with and without L-cysteine treatment. The new peak at 1744 cm^{-1} represents the carbon-oxygen double bond, indicating that the esterification reaction was successfully conducted and the sulfhydryl group was grafted onto the cotton fiber surface. TC stands for yarns that are thiolated with L-cysteine.

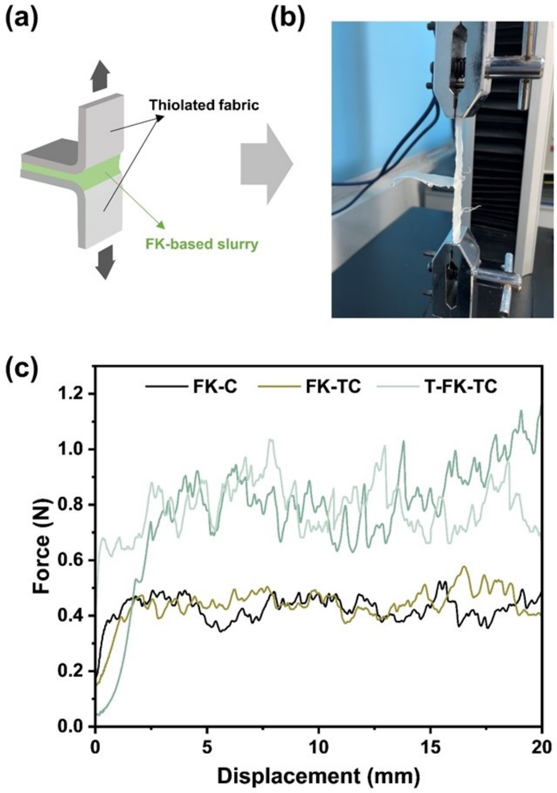


Fig. S7 T-peel test for slurry and yarn adhesion. Schematic(a) and physical(b) diagram of T-peeling test. (c) Representative displacement-force curves of TR-FK sizing agent with thiolated yarn, demonstrating a significant increase in the adhesion between two pieces of fabric after both FK slurry and fabric undergo thiolation.

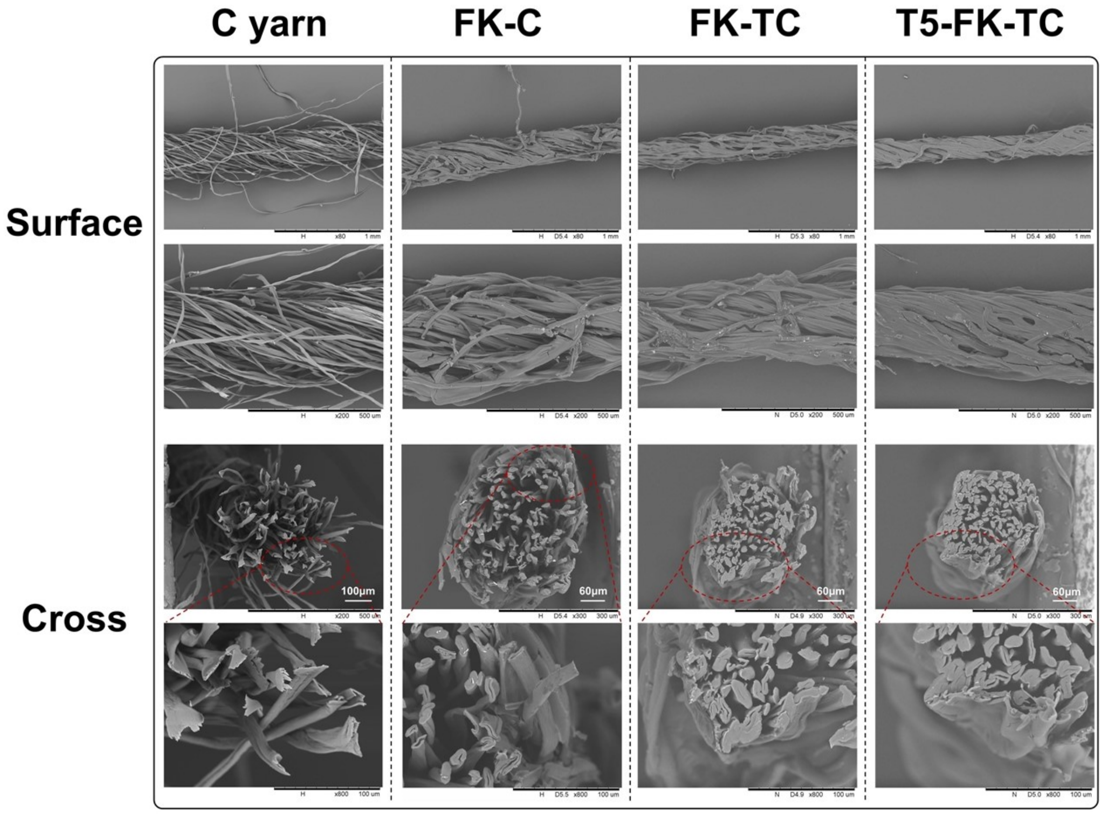


Fig. S8 Scanning electron microscope images of original cotton yarn and FK-based slurry sized yarns. Surface and cross-section morphologies of four kinds of yarns, showing that sizing films became smoother and more continuous.

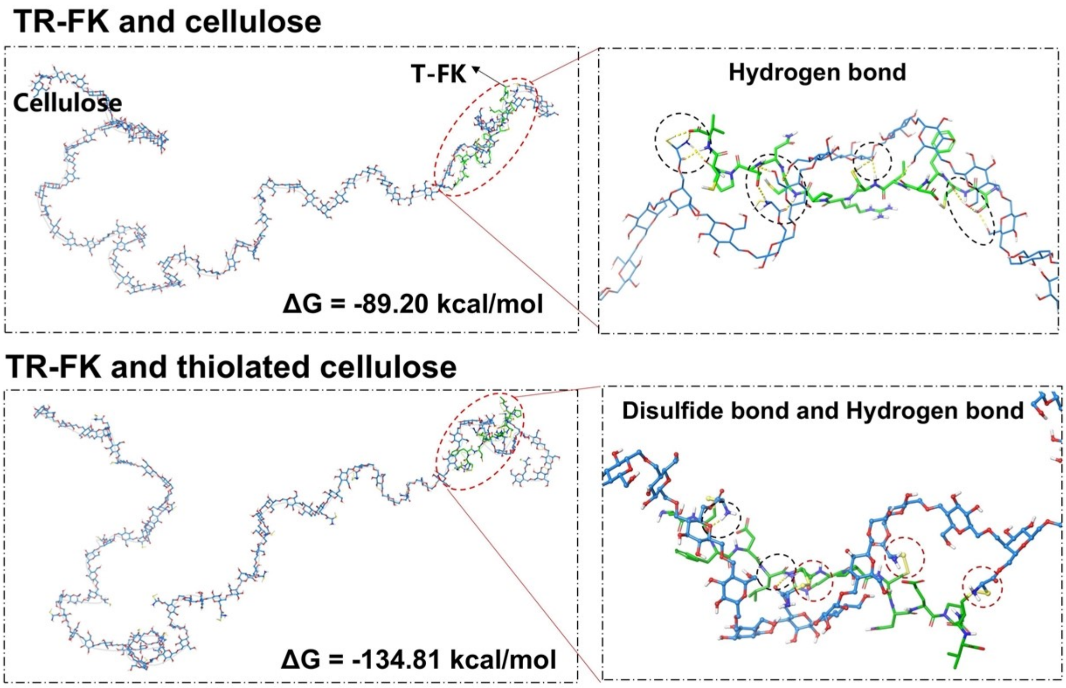


Fig. S9 Interface binding simulation of T-FK and cellulose, thiolated cellulose molecule.

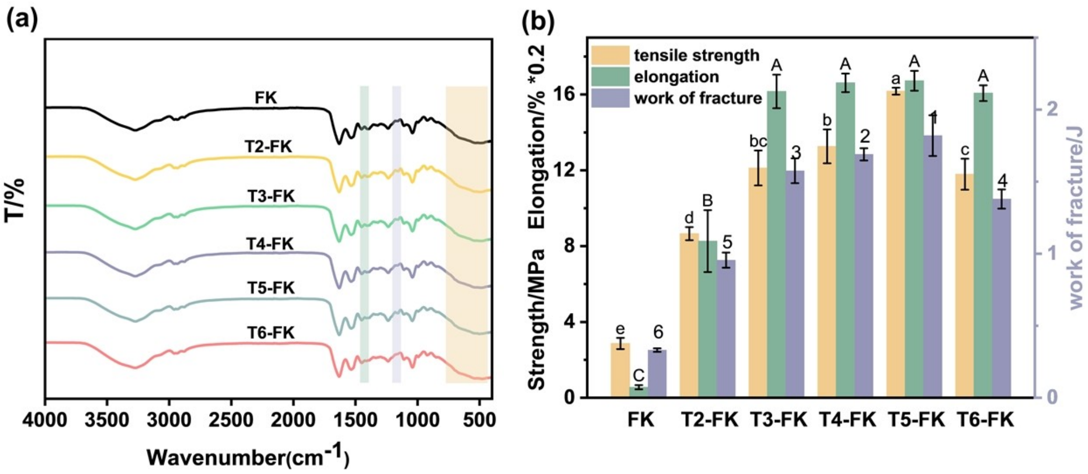


Fig. S10 FTIR spectrum and representative tensile curve of FK-based sizing films.

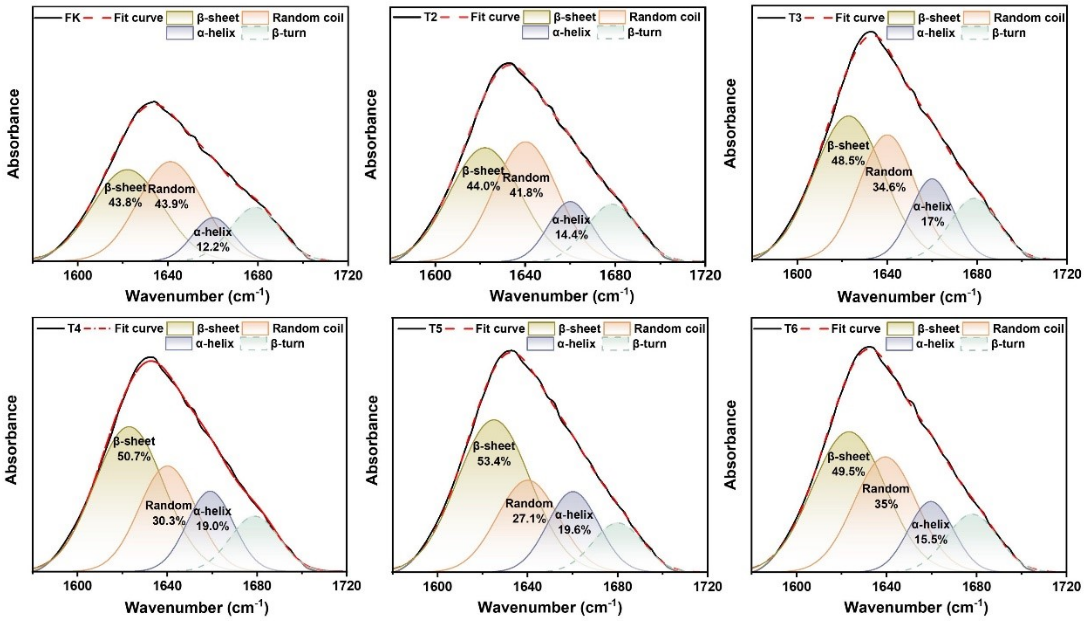


Fig. S11 FTIR deconvolution analysis of sizing films. As the amount of thiol increases, β-sheet and α-helix exhibit a trend of initially rising and then falling.

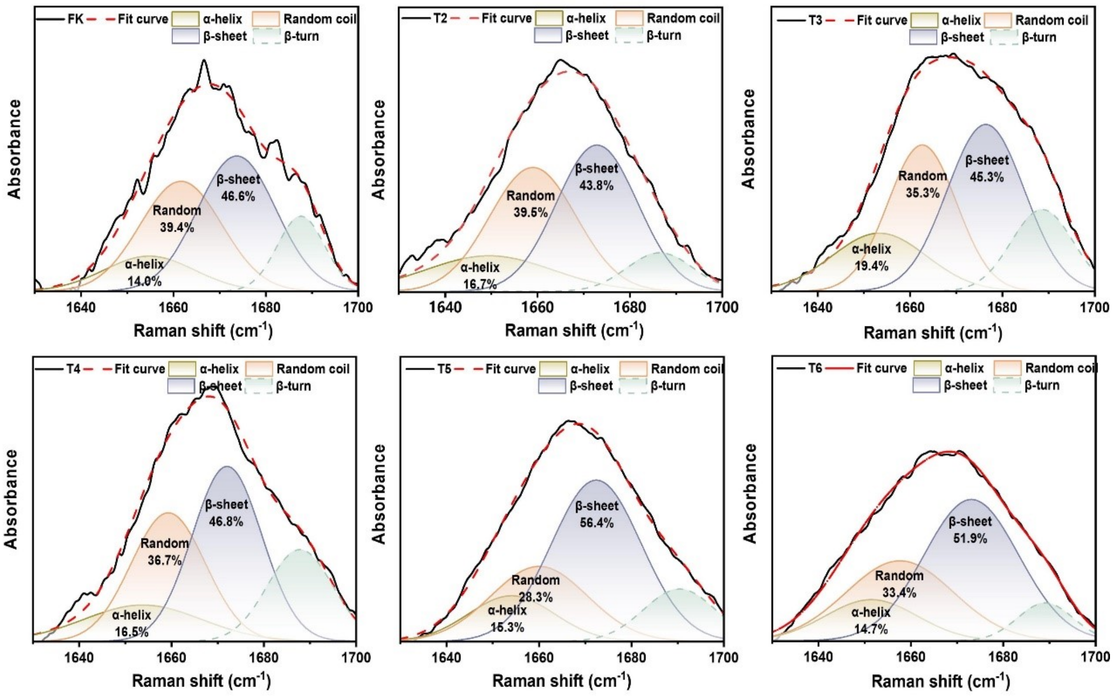


Fig. S12 Raman deconvolution analysis of sizing films. The trend of the secondary structure changes it reveals is consistent with the infrared spectroscopy, providing support for the analysis of the secondary structural transformation in the slurry films.

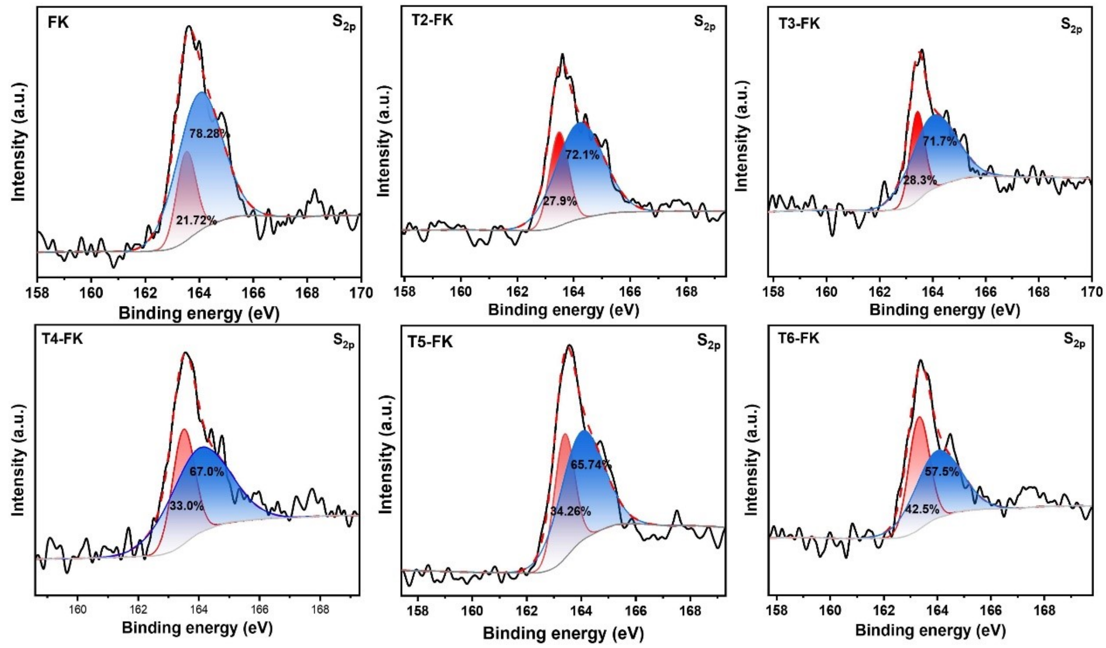


Fig. S13 Sulfur XPS spectra of sizing films. As the amount of TR reagent increases, the free sulfhydryl groups content in the keratin macromolecular chain increases, resulting in a higher proportion of sulfhydryl groups in the formed film, while the proportion of disulfide bonds decreases.

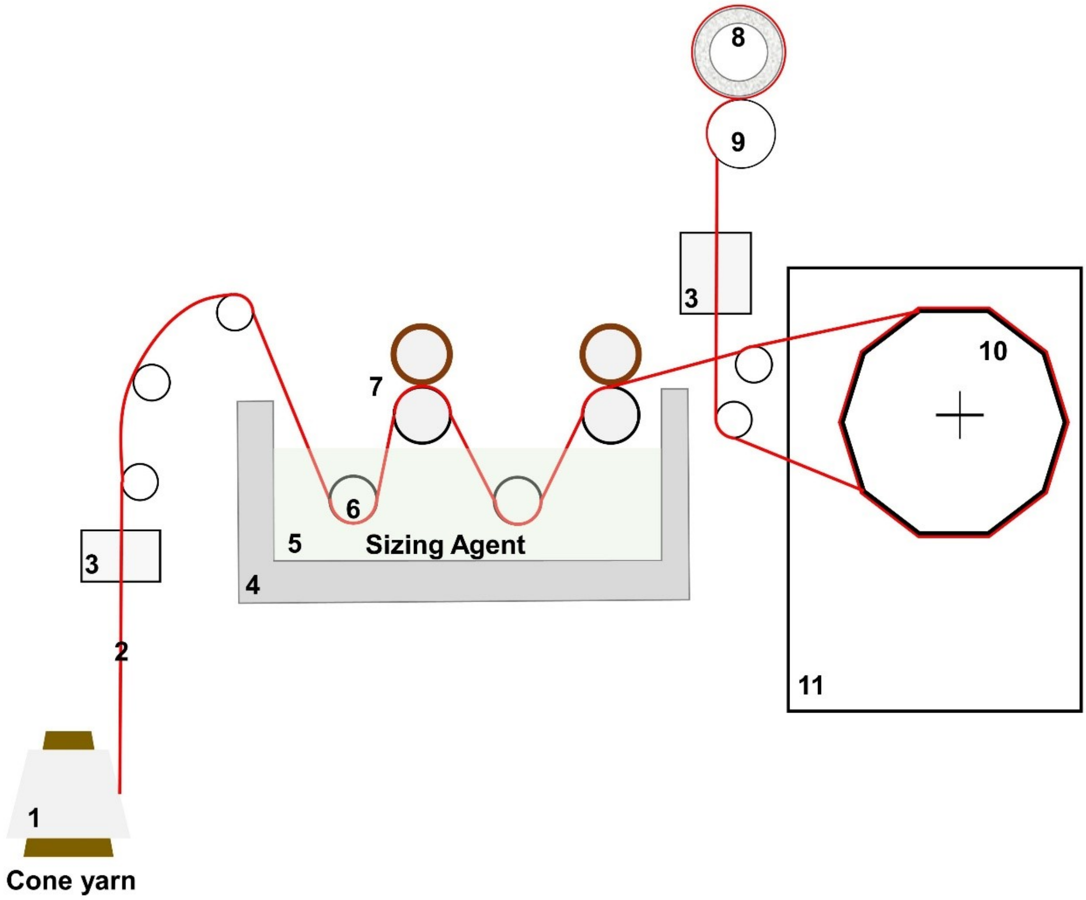


Fig. S14 Process flow chart of sizing machine: 1. Cone yarn, 2. Yarn, 3. Tenslator, 4. Slurry tank, 5. Slurry, 6. dip roll, 7. Upper and lower roller, 8. Winding drum, 9. groove drum, 10. Tumble, 11. Drying room. The red line is the path of yarn.

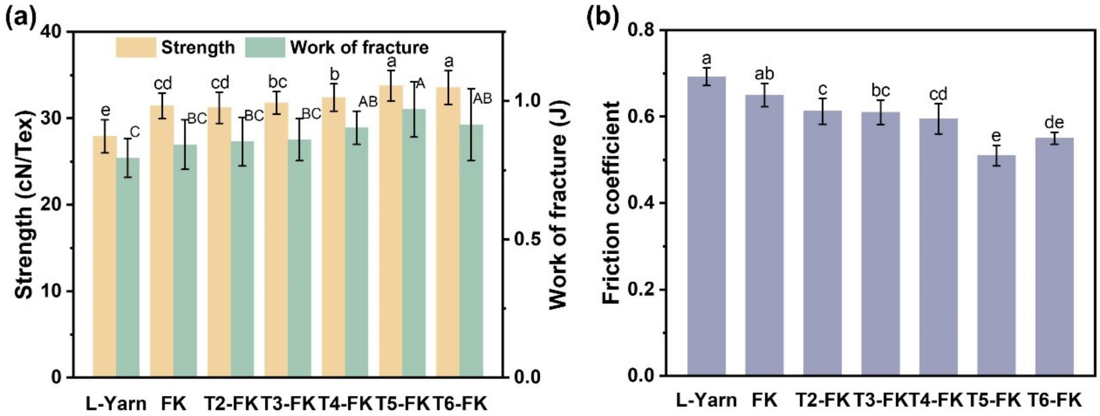


Fig. S15 Tensile strength and work of fracture of cotton yarn with and without sizing process. The yarn sized with T5-FK exhibits the optimal mechanical performance.

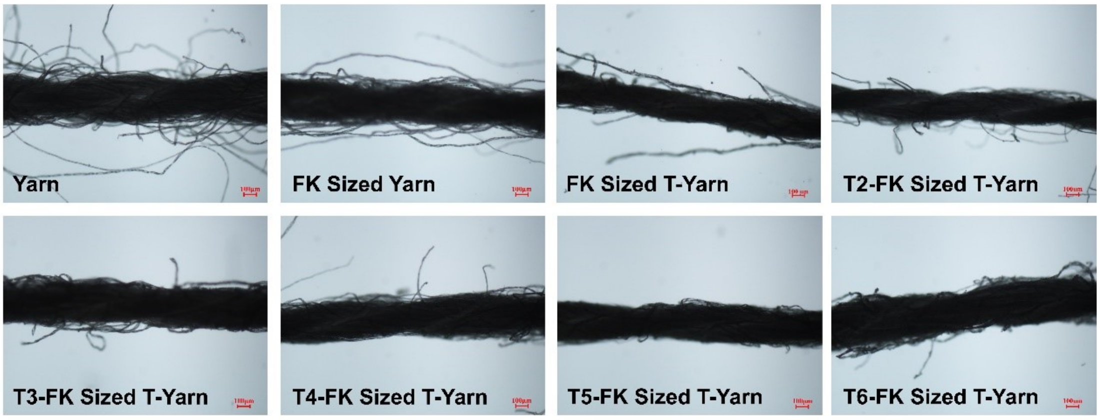


Fig. S16 Optical microscope image of greige yarn and yarn sized FK-based slurry. Benefit from the ability of hair coverage, the sized yarn possesses a significantly smoother surface. T-yarn represents yarn that has been thiolated with L-cysteine.

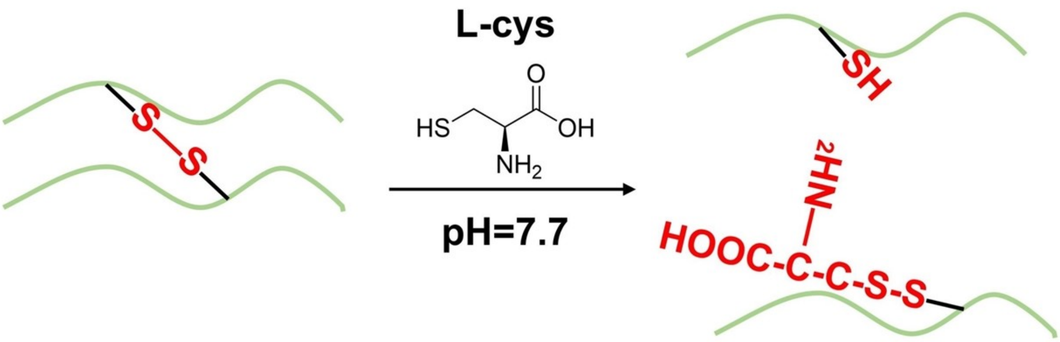


Fig. S17 Chemical reaction during the desizing process: Cysteine mediates the cleavage of disulfide bonds both within the FK-based sizing film and at the film/yarn interface.[1]

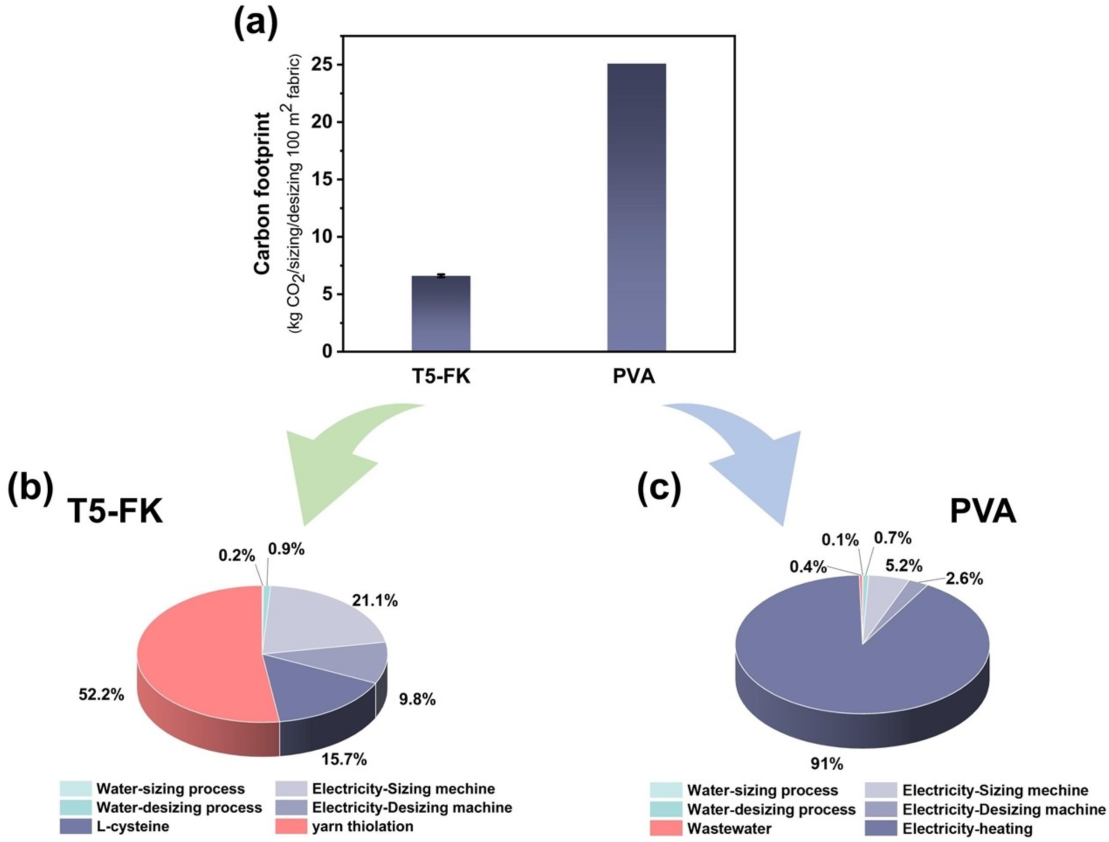


Fig. S18 Life cycle assessment (LCA) of sizing/desizing and yarn thiolation process on TR-FK and PVA slurry. (a) The carbon footprint to treat 100 m^2 (average weight 15Kg) cotton fabric by T5-FK and PVA slurry. (b) and (c) The proportion of each part of the carbon footprint to treat 100 m^2 fabric.

Table S1 Sizing properties of other commonly used textile sizing materials

Slurry	modification method	Sizing Film		Sized Yarn	Desizing Efficiency %		Biodegradability	references
		Strength /MPa	Elongation %	Strength cN/Tex	20℃	90℃	BOD_5/COD	
This work		16.17±0.18	83.62±0.52	33.76±1.77	99.53±0.3	-	-	-
Starch-based	Crosslink	32.6	4.26	32.0	-	-	0.326	[2]
	Physical plasticizing	28.5	3.53	13.3	-	-	-	
PVA		24.1±0.7	150.2±4.3		90.33±1.25	99.81±0.11	0.025	[3]
acrylic	Blending	22.09	16.53				0.0688	
		21.39	7.85				0.0948	
	-							
Soymeal	TEA	9.9±0.3	165±5.1		92.23±1.39	95.00±2.06	0.38	[4]
	CNC	11.8±0.6	50.6±4.2	25.8±0.4	-	95.5		[5]
	1,4	1.6±0.15	112.4±0.88	25±0.2	-	-	0.176	[6]
Keratin	-	-	-	-	99.8±0.28	100±0.81	0.17	[7]
	chemical crosslinking				95.51±0.82	97.12±0.9	-	[8]
	Blending	23.1±1.2	62.8±3.7	17.46±0.01	-	98.5±0.2	-	[9]

Table S2 Secondary structure content of sizing films with different TR addition presented from FTIR deconvolution analysis in this work

Sizing films \ Secondary Structure content (%)		β-sheet 1620 cm^{-1}	Random coil 1640 cm^{-1}	α-helix 1660 cm^{-1}	β-turn 1680 cm^{-1}
FK	FTIR	37	37.1	10.3	15.5
	Raman	40.27396	34.11883	12.10583	13.50138
T2-FK	FTIR	37.9	36	12.4	13.8
	Raman	40.1	36.2	15.3	8.4
T3-FK	FTIR	42.3	30.2	14.8	12.7
	Raman	38.62992	30.08928	16.53909	14.74171
T4-FK	FTIR	44.5	26.6	16.6	12.2
	Raman	38.26674	29.95678	13.48964	18.28684
T5-FK	FTIR	46.7	25.2	17.8	10.3
	Raman	49.67947	24.97339	13.48983	11.85731
T6-FK	FTIR	43.2	30.6	13.5	12.7
	Raman	48.05186	30.96226	13.63692	7.34896

Table S3 Analysis of the binding energy between keratin molecules and cellulose molecules with and without and modification

Sample	Cellulose molecules with keratin	Thiolated cellulose molecules with keratin
Total binding free energy (kcal/mol)	**-89.2**	**-134.81**
hydrogen bond (kcal/mol)	-9.12	-5.25
Van der Waals' force (kcal/mol)	-65.22	-81.43
covalent bond (kcal/mol)	0	-36.44
solvation energy (kcal/mol)	-14.86	-11.69

Table S4 Input and output (life cycle inventory) of sizing/desizing and yarn pretreatment process on T5-FK and PVA sizing agent for the life cycle carbon analysis.

Input/Output	Materials	CAS	Corresponding database	Quantity	Unit	References
Yarn pretreatment process						
Input*						
Energy			Electricity, high voltage {CN}| market group for electricity, high voltage | Cut-off, U	3.375	Kwh	
water			Water, deionised {RoW}| market for water, deionised |Cut-off, U	78.56	L	
Cys	Cysteine	52-90-4	Glycine l market for glycine | Cut-off, U	0.1875	Kg	[10]
Output*						
thiolated yarn				7.5	Kg	
Sizing process of T5-FK						
Input*						
Energy			Electricity, high voltage {CN}| market group for electricity, high voltage | Cut-off, U	1.575	Kwh	
water			Water, deionised {RoW}| market for water, deionised |Cut-off, U	28.2	L	
Output*						
Sized yarn				7.5	Kg	
Desizing process of T5-FK						
Input*						
Energy			Electricity, high voltage {CN}| market group for electricity, high voltage | Cut-off, U	0.78	Kwh	
Cys	Cysteine	52-90-4	Glycine l market for glycine | Cut-off, U	0.2181	Kg	[10]
Water			Water, deionised {RoW}| market for water, deionised | Cut-off, U	150	L	
Output*						
Desized fabric				100	m^2	

Input/Output	Materials	CAS	Corresponding database	Quantity	Unit	References
Sizing process of PVA						
Input*						
Energy			Electricity, high voltage {CN}| market group for electricity, high voltage | Cut-off, U	1.575	Kwh	
Energy (heating)			Electricity, high voltage {CN-ECGC} market for electricity, high voltage | Cut-off, U	1.575	Kwh	
water			Water, deionised {RoW}| market for water,	28.8	L	

			deionised |Cut-off, U			
Output*						
Sized yarn					m	
Desizing process of PVA						
Input*						
Energy			Electricity, high voltage {CN}| market group for electricity, high voltage | Cut-off, U	0.77	Kwh	
Energy (heating)			Electricity, high voltage {CN-ECGC} market for electricity, high voltage | Cut-off, U	28.875	Kwh	
Water			Water, deionised {RoW}| market for water, deionised |Cut-off, U	450	L	
Output*						
Desized fabric				100	m^2	
Wastewater			Wastewater from textile production {GLO} market for wastewater from textile production | Cut-off, U	450	L	

Movie S1 Sizing process of thiolated cotton ribbon with dyed T5-FK sizing agent

Movie S2 The reduction-triggered desizing process of sized cotton ribbon

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