

## Electronic Supplementary Information (ESI)

### **Eco-friendly dyeing of raw cotton fibres in an ethanol-water mixture without scouring and bleaching pretreatments**

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## Conventional dyeing of RCFs

For comparison, RCFs were dyed using the conventional aqueous dyeing method, including the pretreatments and dyeing processes. In detail, RCFs were firstly scoured and bleached in the water solution containing 2 g/L of NaOH, 2.5 g/L of pectinase, and 25 g/L of 30 wt% H<sub>2</sub>O<sub>2</sub> at 95 °C for 45 min using a one bath method. Then the scoured and bleached RCFs were dyed in the aqueous dye solutions at the temperature of 60°C for 60 min. The Na<sub>2</sub>SO<sub>4</sub> and Na<sub>2</sub>CO<sub>3</sub> added in dyeing process were at the concentration of 40 g/L and 15 g/L, respectively.

## Fourier transform infrared (FTIR) spectroscopy

FTIR spectra of the dye solutions were determined using a Vertex 70 spectrometer (Bruker Corporation, Karlsruhe, Germany) via reflection–absorption spectroscopy.

## X-ray diffraction (XRD)

XRD data of the samples were recorded using a wide-angle XRD analysis system (X'Pert PRO, PANalytical B.V., Holland). Samples were scanned from 10° to 60° with a step size of 0.02°.

## X-ray photoelectron spectroscopy (XPS)

XPS spectra of the samples were examined using a dual anode XSAM800 spectrometer from KRATOS using non-monochromatic Al K $\alpha$  X-radiation ( $h\nu$ = 1486.6 eV).

## Thermogravimetric analysis (TGA)

TGA was performed on a Netzsch TG 209 F1 thermal analyzer under nitrogen flow over a temperature range of 30 °C to 800 °C at a heating rate of 10 °C·min<sup>-1</sup>.

## Calculation of exhaustion and total fixation

The dye exhaustion ( $E$ ) of reactive dyes was calculated using Eqn (1):<sup>1</sup>

$$E(\%) = \left[ 1 - \left( \frac{A_b}{A_a} \right) \right] \times 100 \quad (1)$$

where  $A_a$  and  $A_b$  are the absorbance of the dye solution at maximum absorption wavelength before and after dyeing, respectively.

The total fixation ( $T$ ) was calculated using Eqn (2):<sup>2</sup>

$$T(\%) = E \times \frac{(K/S)_b}{(K/S)_a} \quad (3)$$

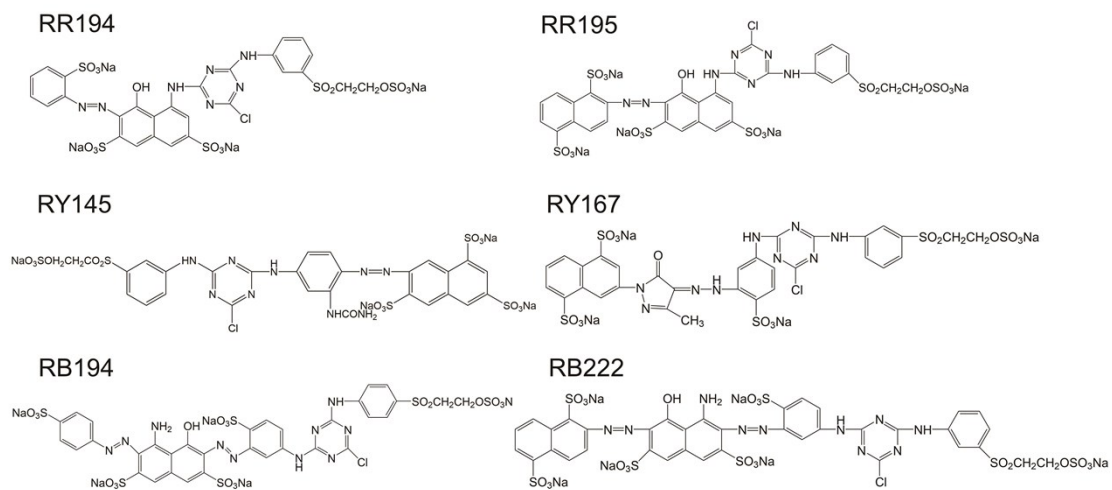
where the  $(K/S)_a$  and  $(K/S)_b$  indicate the  $K/S$  values obtained before and after soaping, respectively.

### **Colour fastness**

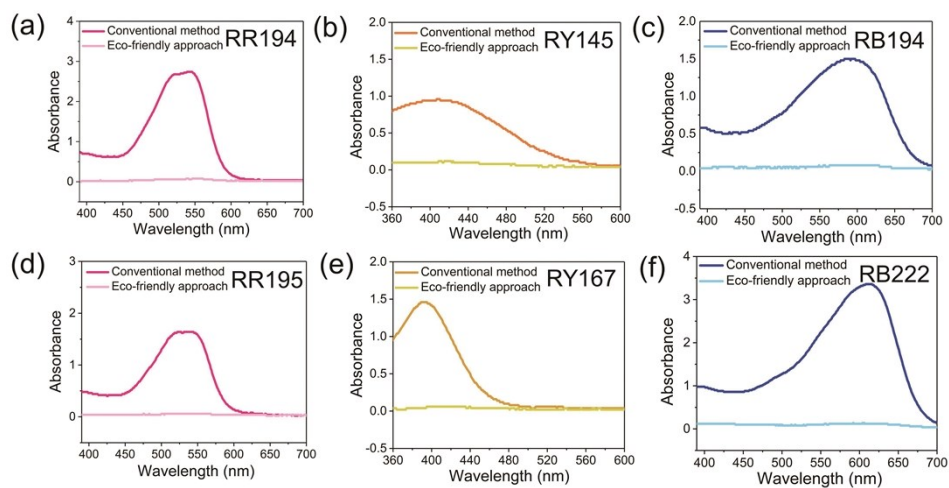
The wash fastness of the dyed samples was examined using an M228 washing fastness tester (SDL Atlas Co., Ltd., China) according to ISO 105-C10:2006. Colour degradation was determined according to the appropriate grey scale. The rubbing fastness test was conducted on a 571-II crockmeter (Wenzhou Darong Textile Instrument Co., Ltd., China) according to ISO 105-X12.

### **References**

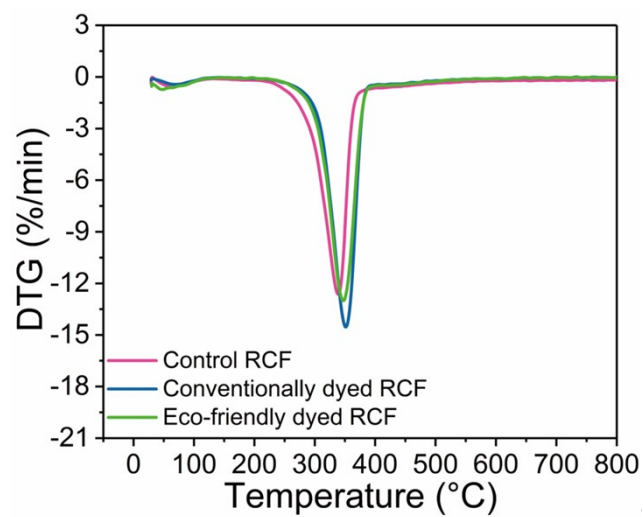
1. K. Xie, F. Cheng, W. Zhao and L. Xu, *Journal of Cleaner Production*, 2011, **19**, 332-336.
2. M. Montazer, R. M. Malek and A. Rahimi, *Fibers and Polymers*, 2007, **8**, 608-612.



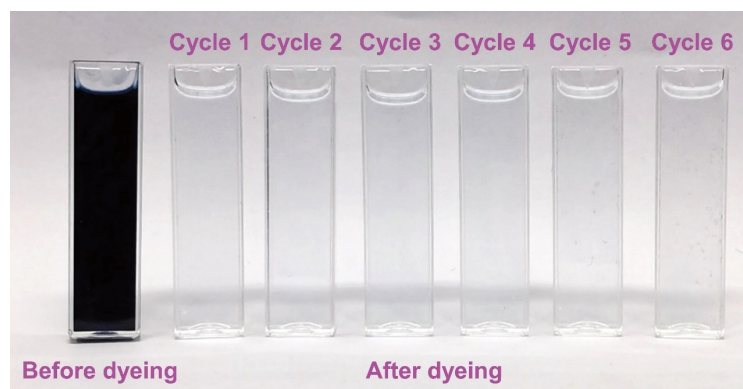
**Fig. S1** Chemical structure of the dyes employed in the experiment.



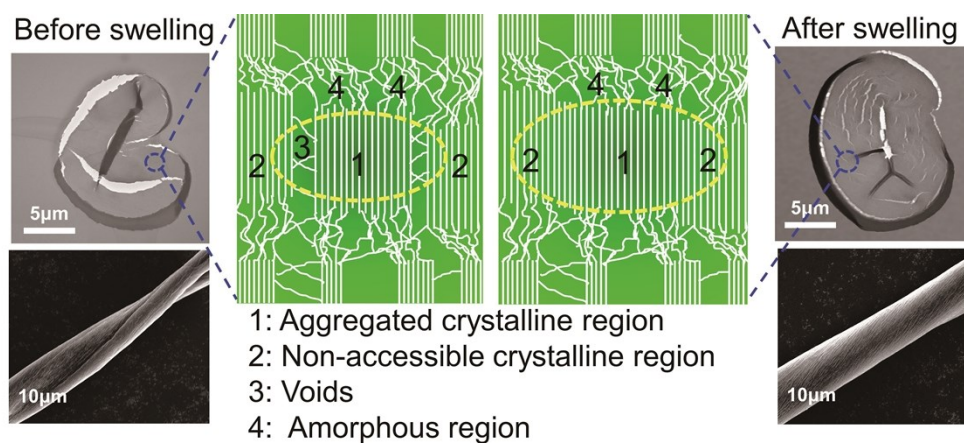
**Fig. S2** Absorption spectra for the dye solutions after dyeing of RCFs with (a) RR194; (b) RY145; (c) RB194; (d) RR195; (e) RY167 and (f) RB222 using the conventional method and eco-friendly approach.



**Fig. S3** DTG curves of the RCFs dyed using the conventionally method and eco-friendly approach.

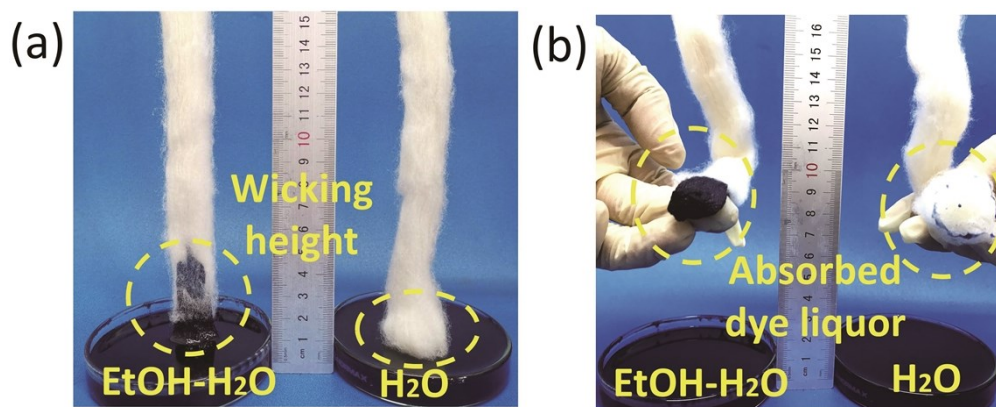


**Fig. S4** Dyeing effluents of RCFs using the fresh and waste swelling solutions.

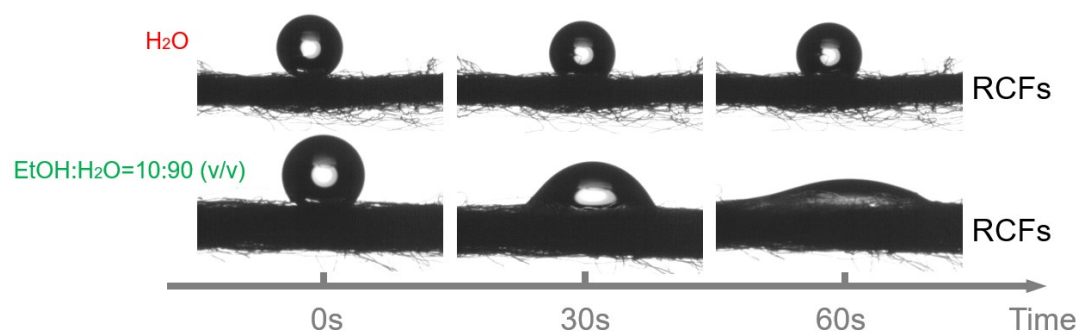


**Fig. S5** TEM and SEM images of RCF before and after swelling in the alkalic EtOH-H<sub>2</sub>O mixture.

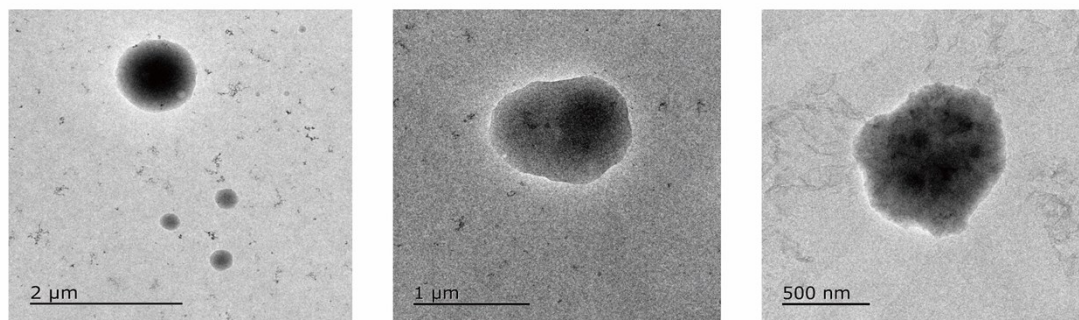




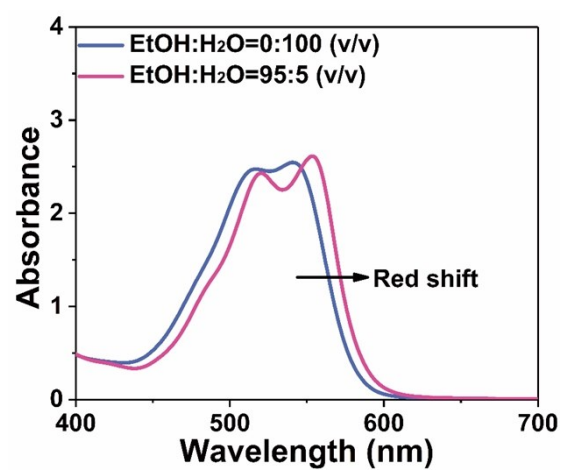
**Fig. S6** Capillary effect of RCFs with the EtOH-H<sub>2</sub>O mixture and the aqueous solution, RB222 was added to make the solutions to be observed easily.



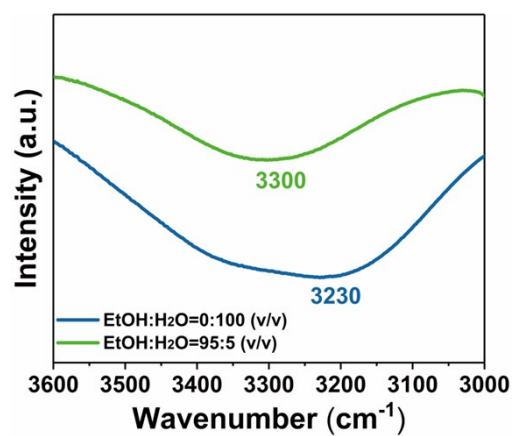
**Fig. S7** Contact angles of H<sub>2</sub>O and EtOH-H<sub>2</sub>O mixture on RCFs.



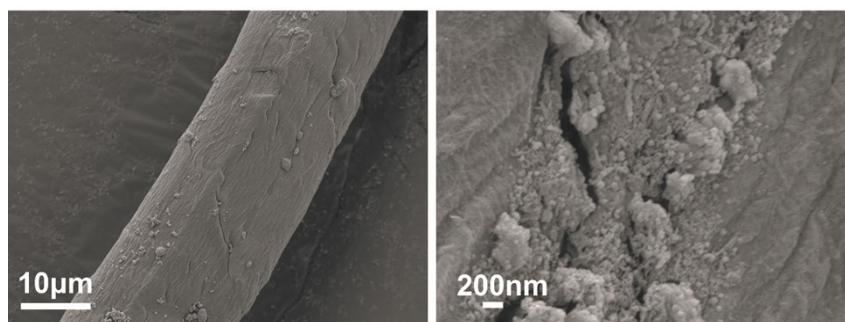
**Fig. S8** Dye aggregates of RR195 in the EtOH-H<sub>2</sub>O mixture with the EtOH volume ratio of 95%.



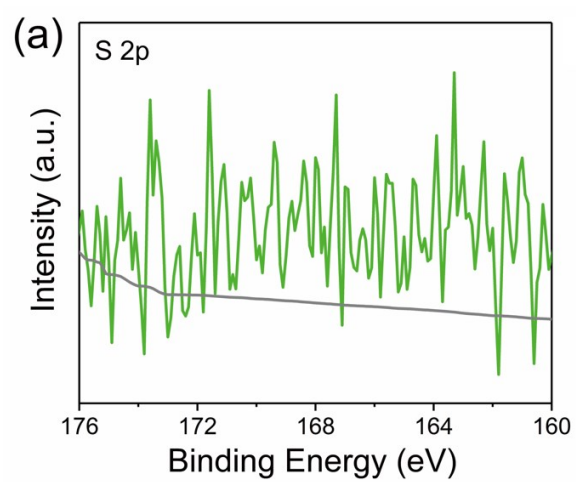
**Fig. S9** Absorption spectra of RR195 in aqueous solution and EtOH-H<sub>2</sub>O mixture.



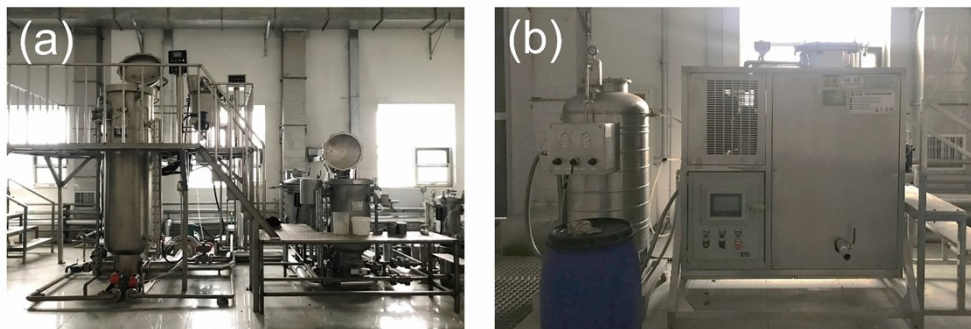
**Fig. S10** FTIR spectra of RR195 in aqueous solution and EtOH-H<sub>2</sub>O mixture.



**Fig. S11** SEM images of the RCF dyed using the eco-friendly approach at the half-time of dyeing.



**Fig. S12** High-resolution XPS spectra of S2p for the control RCF.



**Fig. S13** (a) Industrial-scale dyeing system for RCFs using the developed eco-friendly approach; (b) EtOH distillation equipment for the swelling solution.



**Table S1** The atomic percentage of carbon, nitrogen, oxygen and sulfur in RCF and eco-friendly dyed RCF.

Samples	Atomic (%)			
	C1s	N1s	O1s	S2p
Control RCF	87.50	1.54	10.67	0.30
Eco-friendly dyed RCFs	65.98	2.84	30.64	0.54

**Table S2** Physical properties of the RCFs dyed using the conventional method and eco-friendly approach.

Samples	Breaking tenacity (cN/dtex)	Length (mm)	Principal length (mm)	Short fibre content (%)	Impurities (Grain/g)	Moisture regain (%)
RCF <sub>5</sub>	2.31	28	28.9	6.9	28/34	8.5
Conventionally dyed RCFs	2.17	28	28.6	7.9	30/34	12.7
Eco-friendly dyed RCFs	2.27	28	28.8	7.1	10/28	13.3

**Table S3** Comparison of the colour fastness between the cotton fabrics fabricated using the conventionally dyed RCFs and eco-friendly dyed RCFs.

Samples	Dry rubbing fastness	Wet rubbing fastness	Fading fastness	Staining fastness	Perspiration fastness
Conventional	4	4	4	4	4
Eco-friendly	4	4	4	4	4