Supporting Information

Preparation of a double-network hydrogel based on wastepaper and its application

in the treatment of wastewater containing copper (II) and methylene blue

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Text S1: Cost analysis.

The price paraments were derived from Sinopharm Chemical Reagent Co, Ltd. (China). The chemicals used in the experiment were shown below: urea (AR, £ 32.84/t), sodium hydroxide (NaOH, AR, £ 26.14/t), acrylamide (AM, AR, £ 88.07/t), epichlorohydrin (ECH, AR, £ 35.39/L), potassium persulfate (KPS, AR, £ 982.03/t), tetramethylethylenediamine (TEMED, AR, £ 1201.74/L), N, N-methylenebisacrylamide (MBA, AR, £ 1118.55/t), etc. According to the synthetic ratio, the preparation of 1 t WP/PAM required 0.3 L ECH, 1.75 t NaOH, 3 t urea, 1 t AM, 0.0484 t MBA, 0.0272 t KPS, and 0.1 L TEMED. Without considering other factors (e.q., electricity, labor and machine maintenance costs) the production cost of the material was about £ 444/t.

Text S2: Measurement of isoelectric point (pH_{PZC}) by the ΔpH drift method:

The WP/PAM were added to a series of 0.01 mol/L NaCl solutions of different initial pH (1-10). After shaking at 170 rpm for 24 h at 25°C, the pH of the supernatant was measured and the pH change before and after the reaction (ΔpH) was calculated. The initial pH corresponding to when $\Delta pH = 0$ was defined as the zero charge point of the material (pH_{PZC}).

Text S3: Specific surface area analysis.

As could be seen from Table S1, the specific surface area of the WP/PAM was 52.1380 m^2/g . It was probably due to the fact that the specific surface area data was obtained by measuring the adsorption and desorption process of N₂ by a BET instrument. N₂ is a neutral molecule, and the adsorption process of N₂ belongs to physical adsorption. The relatively small specific surface area of the hydrogel was due to the small amount of N₂ adsorbed by the WP/PAM, indicating a small physical adsorption occupancy in the hydrogel adsorption process.¹



Fig. S1. The preparation process of the WP/PAM.



Fig. S2. Sampling site of Xiangjiang river water (Changsha, Hunan province, China,

August 19th, 2020. North latitude: 28°17'4", East longitude: 112°95'65".).



Fig. S3. Effect of WP content on (a) Cu (II) removal and (b) MB removal by the WP/PAM. (*** indicated a significant difference between the two groups, P value<0.001; no labeling indicated no significant difference between the two groups) (Dosage: 0.5 g/L; $C_{0(Cu (II))}$: 100 mg/L, $C_{0(MB)}$: 500 mg/L; 180 min; Temperature: 25°C).



Fig. S4. Photos of (a) 1 wt%, (b) 2 wt%, (c) 3 wt%, (d) 4 wt% WP/PAM and SEM

images of (e) 1 wt%, (f) 2 wt%, (g) 3 wt%, (h) 4 wt% WP/PAM.



Fig. S5. Effect of adsorbent dosage. ($C_{0(Cu (II))}$: 100 mg/L, $C_{0(MB)}$: 500 mg/L; 180 min; Temperature: 25°C).



Fig. S6. Regeneration and readsorption of the WP/PAM within 6 cycles. (Dosage_{(Cu (II))}: 0.7 g/L, Dosage_(MB): 0.4 g/L; $C_{0(Cu (II))}$: 100 mg/L, $C_{0(MB)}$: 600 mg/L; 180 min; Temperature: 25°C).

	1 wt% WP/PAM	2 wt% WP/PAM	3 wt% WP/PAM	4 wt% WP/PAM
Porosity	07.5	00.1	02.7	00.5
(%)	97.5	98.1	92.7	88.5

Table S1 the porosity of the WP/PAM with different WP content

	WP/PAM
Specific surface Area (m ² /g)	52.1380
porous ratio (cm ³ /g)	0.02179

 Table S2 Specific surface area and porous ratio of the WP/PAM

Adsorbent	$\frac{1}{Q_e (mg/g)}$	
	Cu (II)	MB
WP/PAM	132.2	1653.6
WP/PAM stored after 16 months	131.0	1630.8

Table S3 the comparison of the WP/PAM performance before and after storage

	Cu (II)		MB	
Cycles	Q_d	Desorption Rate	Q_d	Desorption Rate
	(mg/g)	(%)	(mg/g)	(%)
0	$Q_e = 139.5$	-	$Q_e = 1705.5$	-
1	130.3	93.4	1626.9	95.4
2	119.0	91.3	1473.4	90.6
3	110.1	92.5	1398.0	94.9
4	101.0	91.7	1298.2	92.9
5	93.1	92.2	1222.9	94.2
6	87.5	94.00	1130.8	92.5

Table S4 Desorption data of Cu (II) and MB by WP/PAM in 6 cycles

References

1 S. Tian, Q. Liu, J. Sun, M. Zhu, S. Wu and X. Zhao, *J. Colloid Interface Sci.*, 2019, **534**, 389-398.