Supporting Information

Title: Cellulose and Wood Derived Activated Carbons from Pinus sylvestris: Comparative Performance in Methylene Blue Adsorption

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Table of Contents

1. Cellulose extraction

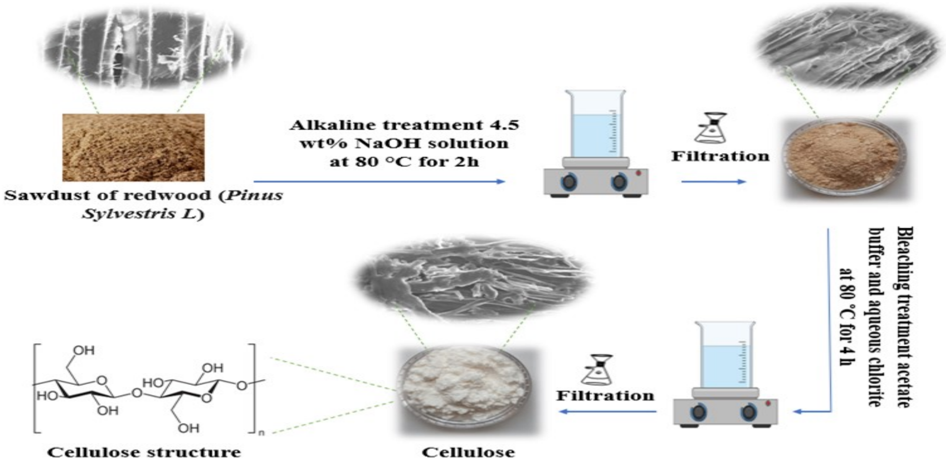

Fig. 1 Schematic representation of the cellulose isolation process from redwood (*Pinus sylvestris* L.) sawdust (*RW)*.

2. Preparation of activated carbon

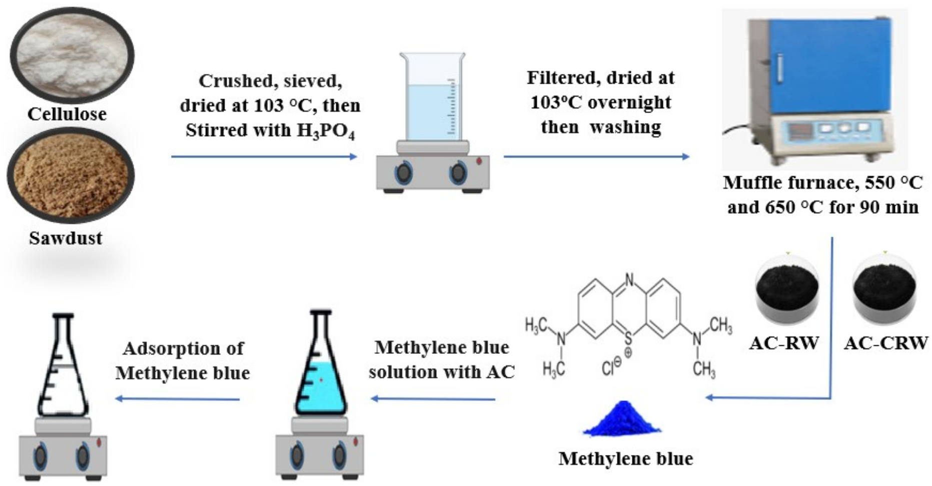

Fig. 2 Schematic illustration of the used process for activated carbons preparation and valorization as methylene blue adsorbents.

3. Comparison of the preparation conditions of AC-RW and AC-CRW

Table 1 Preparation conditions of AC-RW and AC-CRW: Type, Impregnation Time, Ratio, and Temperature.

Type of AC	Impregnation time (h)	Ratio	Temperature (°C)
AC-RW	16	1:1	550
AC-CRW	24	1:2	650

4. Adsorption isotherms

Table 2 Non-linear adsorption isotherm models and their characteristic parameters.

Isotherm Model	Non-linear Equation	Parameters	Ref.
Langmuir	$q_e = \frac{q_m K_L C_e}{1 + K_L C_e}$	q_{max}: maximum adsorption (mg/g) capacity; K_L: Langmuir constant	19,36
Freundlich	$q_e = K_F C_e^{1/n}$	K_F: Freundlich constant; n: adsorption intensity	37,38
Temkin	$q_e = Bln(K_T C_e)$	B: Temkin constant related to heat of adsorption; K_T: Temkin isotherm constant	39
Sips	$q_e = \frac{q_m (K_s C_e)^n}{1 + (K_L C_e)^n}$	K_s : (L/mg) is the Sips equilibrium constant; n : Heterogeneity index	39,40

5. Comparison of adsorption efficiency (%) of MB between AC-RW and AC-CRW

Table 3 Adsorption efficiency (%) of MB on AC-RW and AC-CRW under different preparation conditions.

Runs	Type of AC	Impregnation time (h)	Ratio	Temperature (°C)	Adsorption Efficiency (%)
1	AC-RW	16	1:1	550	78
2	AC-RW	16	1:2	550	82
3	AC-RW	24	1:1	550	81
4	AC-RW	24	1:2	550	86
5	AC-RW	16	1:1	650	84
6	AC-RW	16	1:2	650	88
7	AC-RW	24	1:1	650	87
8	AC-RW	24	1:2	650	92
9	AC-CRW	16	1:1	550	83
10	AC-CRW	16	1:2	550	87
11	AC-CRW	24	1:1	550	86
12	AC-CRW	24	1:2	550	91
13	AC-CRW	16	1:1	650	89
14	AC-CRW	16	1:2	650	93
15	AC-CRW	24	1:1	650	91
16	AC-CRW	24	1:2	650	96

6. Thermogravimetric analysis (TG-DTG).

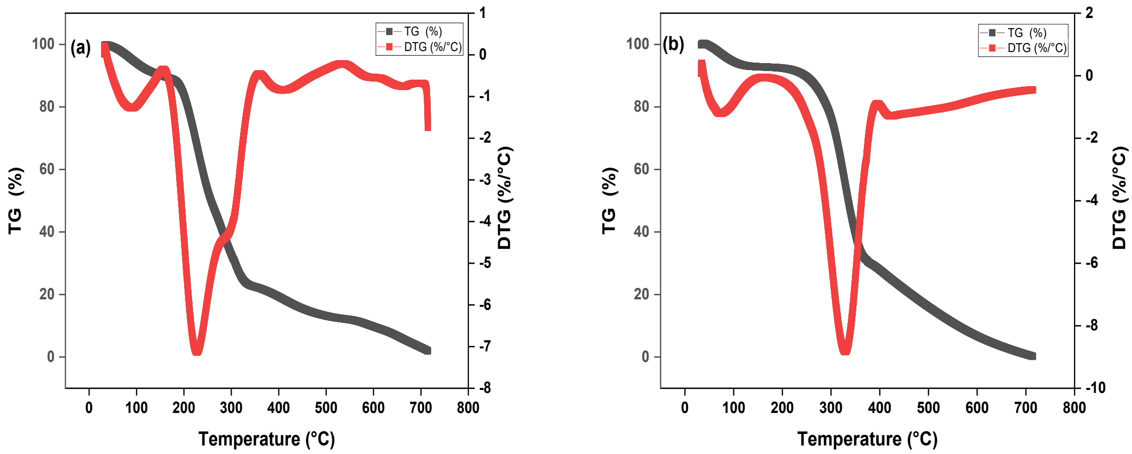

Fig. 3 TG/DTG of (a) RW (*Pinus Sylvestris L.*) and (b) cellulose.

7. SEM and BET of prepared AC-RW and AC- CRW

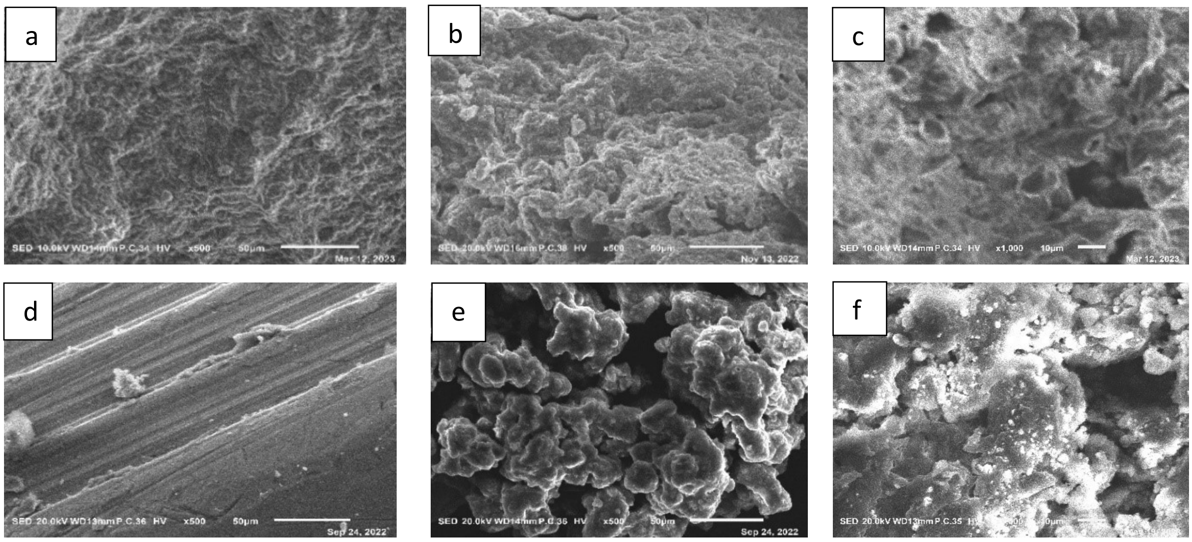

Fig. 4 SEM of optimal AC-RW (a) before activation and (b,c) after activation and AC-CRW (d) before activation and (e,f) after activation .

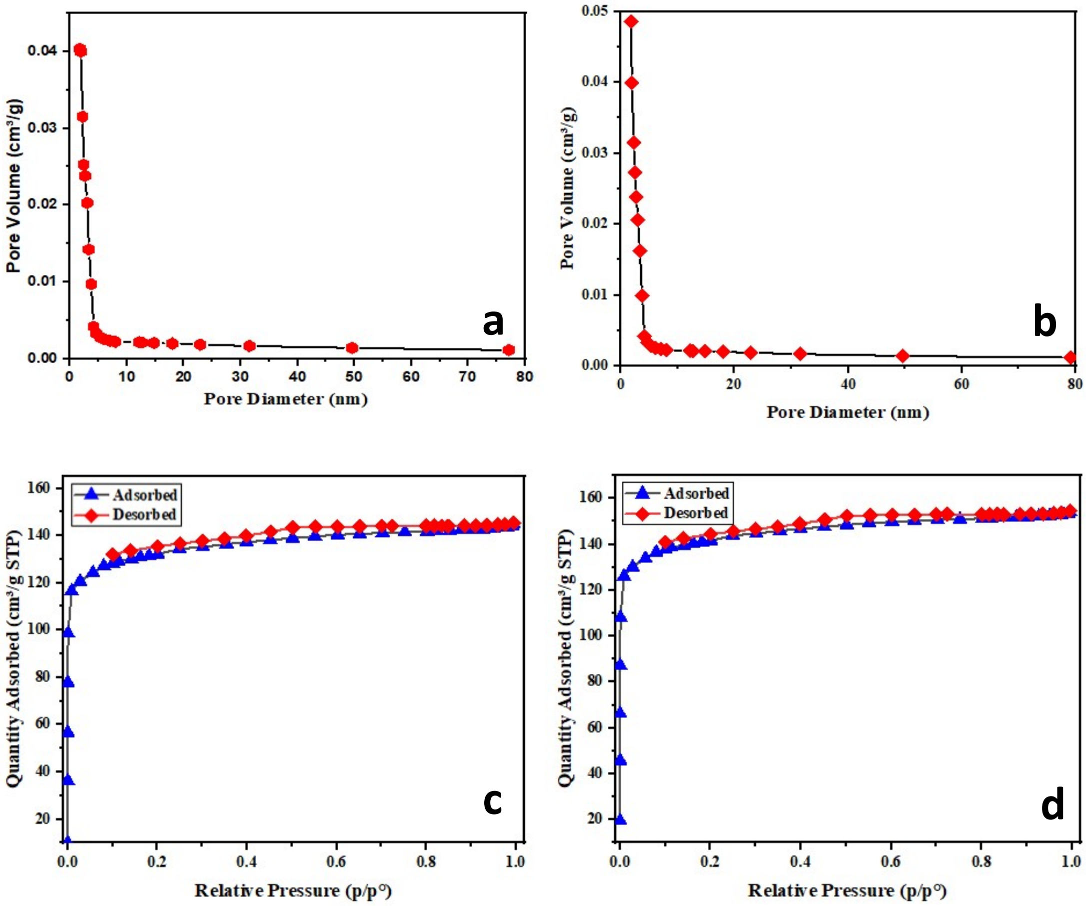

Fig. 5 N_2 adsorption-desorption isotherms and pore size distributions of AC-RW (a,c) and AC-CRW (b,d).

8. Characterization by FTIR spectrophotometry

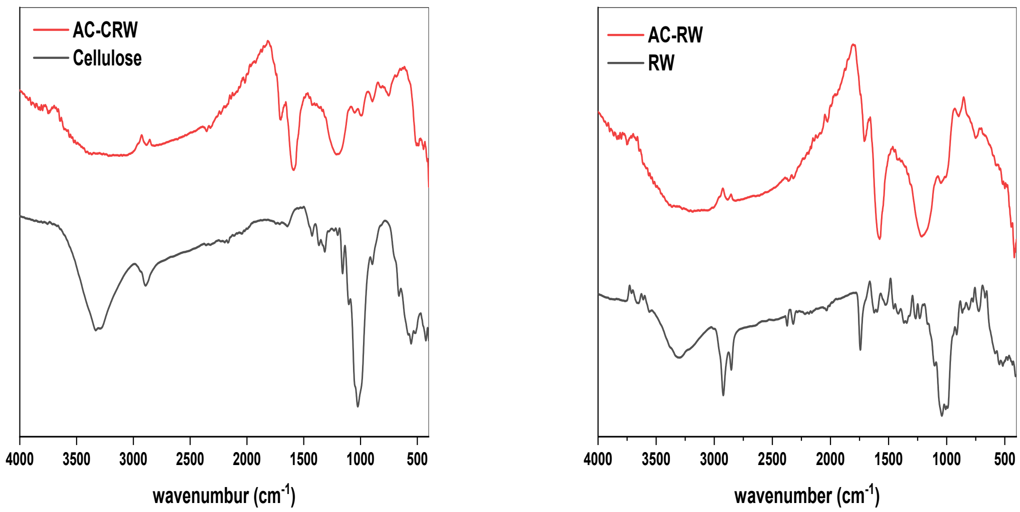

Fig. 6 FTIR spectra of RW, AC-RW, Cellulose and AC- CRW.

9. X-ray diffraction analysis.

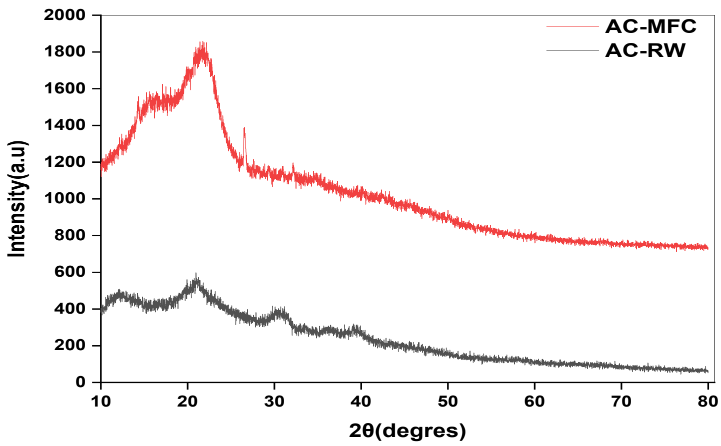

Fig. 7 XRD spectra of optimal *AC-RW and AC*-CRW.

10. pH point zero charge of AC-RW and AC-CR

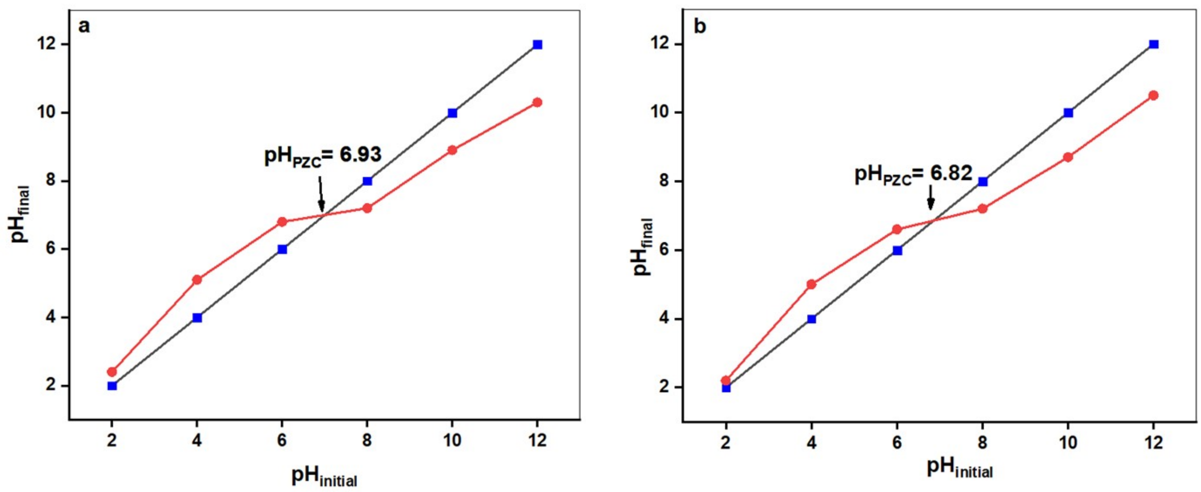

Fig.8 Determination of the point of zero charge pH_{PZC} for the prepared activated carbons: (a) cellulose-derived carbon (AC-CRW) and (b) wood-derived carbon (AC-RW).

11. Effect of pH on adsorption

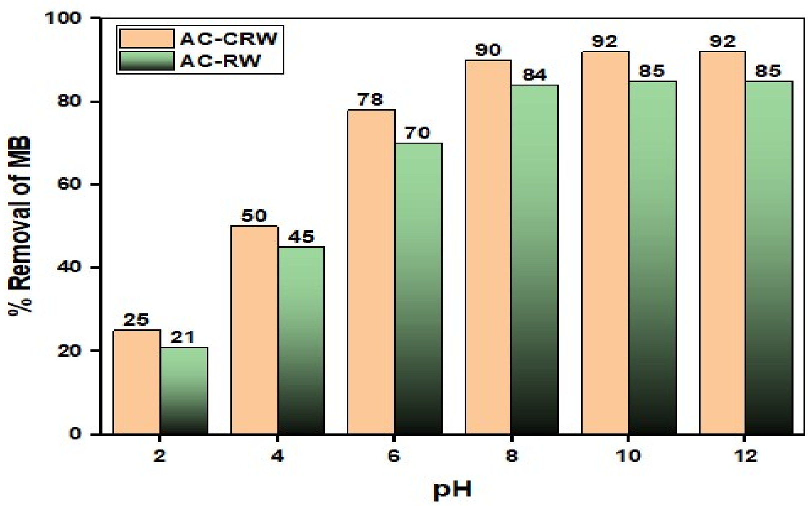

Fig. 9 Effect of solution pH on MB removal (%) by AC-CRW and AC-RW.

12. Effect of temperature on MB adsorption performance

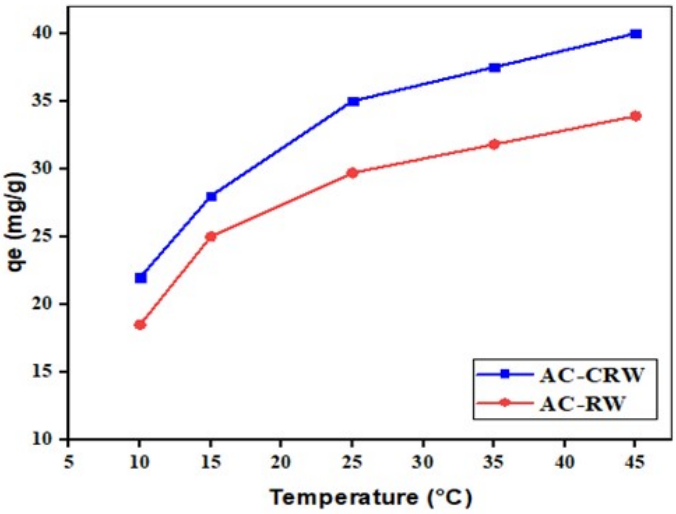

Fig. 10 Temperature effect on the adsorption capacity (qe) of MB onto AC-CRW and AC-RW (10–45 °C).

13. Effect of contact time and dye concentration on adsorption

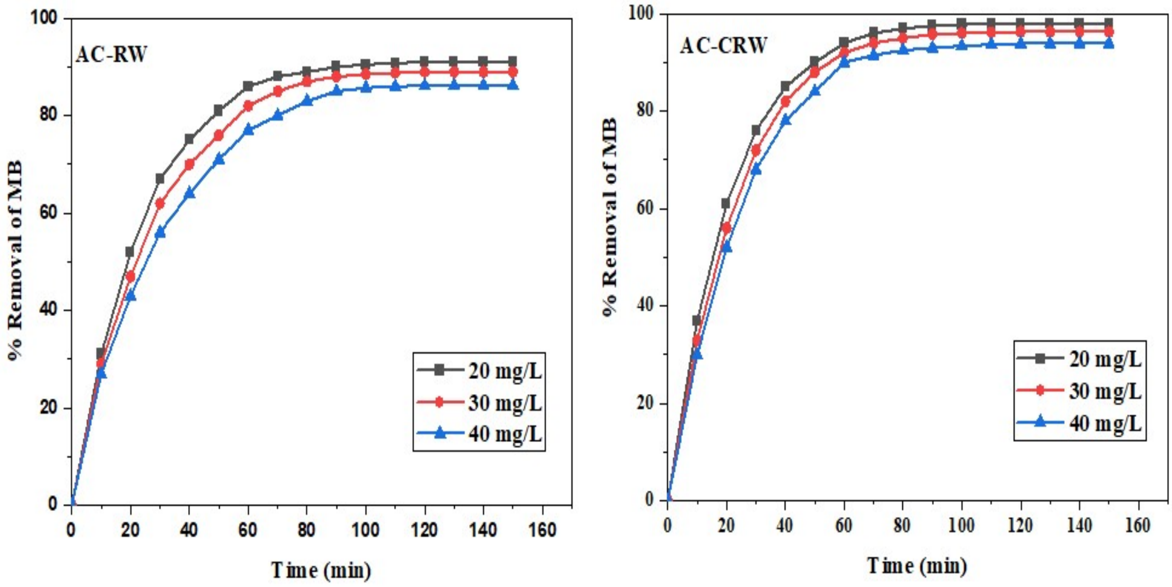

Fig. 11 Effect of contact time and initial concentration on the adsorption of methylene blue a) AC-RW and b) AC-CRW (conditions:100 mg of AC at.25 °C).

14. Adsorption kinetic

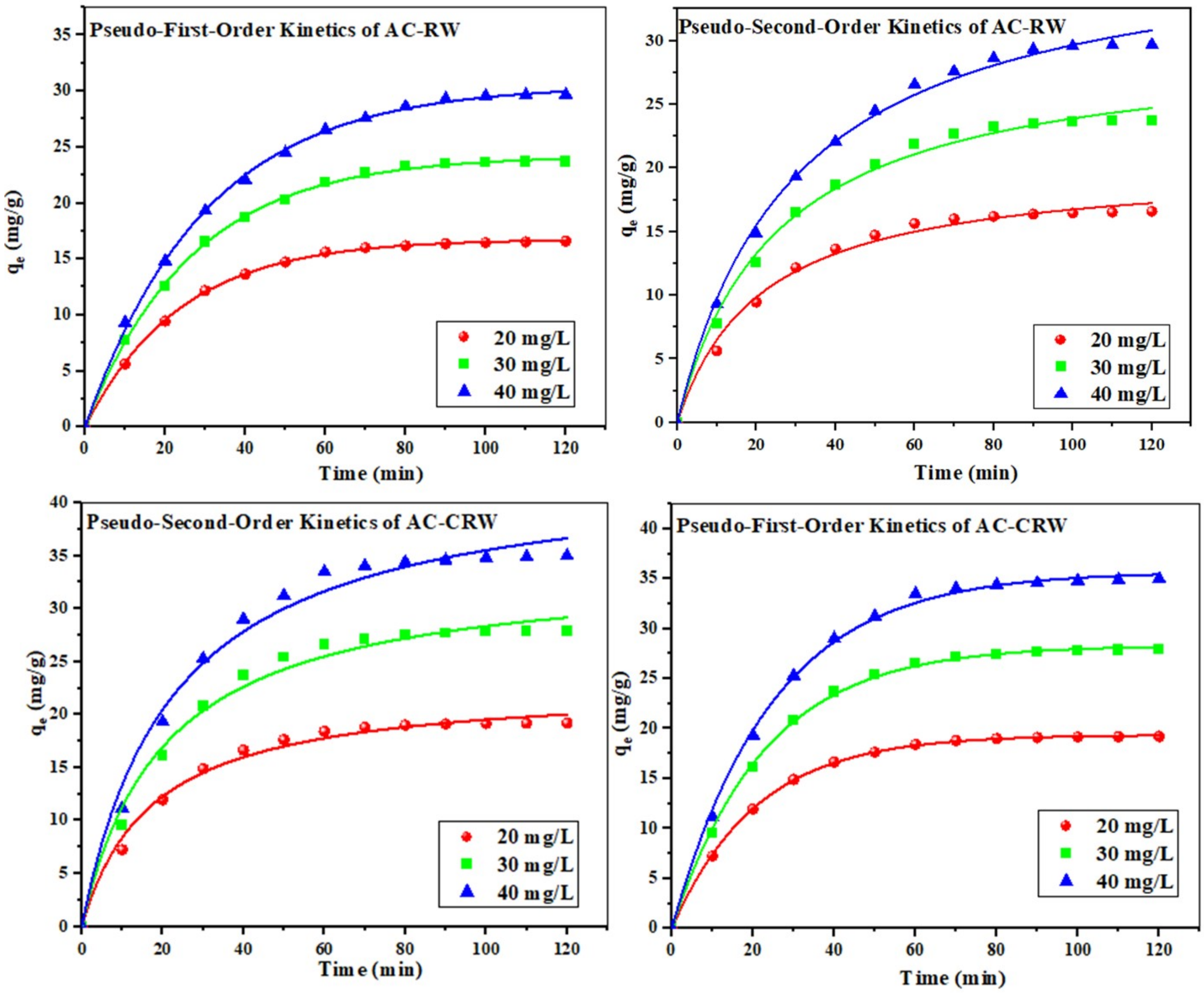

Fig. 12 Adsorption kinetics of MB onto AC-RW and AC-CRW at 25 °C fitted with pseudo-first-order and pseudo-second-order models.

Table 4 Kinetic parameters of pseudo-first-order and pseudo-second-order for MB using optimal AC-RW and AC-CRW.

Kinetics model	**Initial concentration (mg/L)**	**Kinetics constant**	**Type of adsorbent**	
			AC-CRW	**AC-RW**
First-order kinetic model40	20	$q_{e,exp}$ (mg/g)	19.21	16.6
		k_1 (min^{-1})	0.042	0.042
		$q_{e,cal}$ (mg/g)	19.37	16.77
		R^2	0.99978	0.99957
	30	$q_{e,exp}$ (mg/g)	27.9	23.7
		k_1 (min^{-1})	0.0126	0.037
		$q_{e,cal}$	28.31	24.21
		R^2	0.99909	0.99927
	40	$q_{e,exp}$ (mg/g)	35	29.7
		k_1 (min^{-1})	0.043	0.033
		$q_{e,cal}$	35.69	30.52
		R^2	0.99829	0.99899
Second-order kinetic model	20	k_2[g(mg^{-1}.min^{-1})]	0.0025	0.0023
		$q_{e,cal}$ (mg/g)	20.90	20.28
		R^2	0.98898	0.99045
	30	k_2[g(mg^{-1}.min^{-1})]	0.00145	0.00134
		$q_{e,cal}$ (mg/g)	34.07	29.85
		R^2	0.9869	0.99271
	40	k_2[g(mg^{-1}.min^{-1})]	0.0012	0.0088
		$q_{e,cal}$ (mg/g)	43.52	38.37
		R^2	0.98578	0.99587

15. Adsorption isotherms

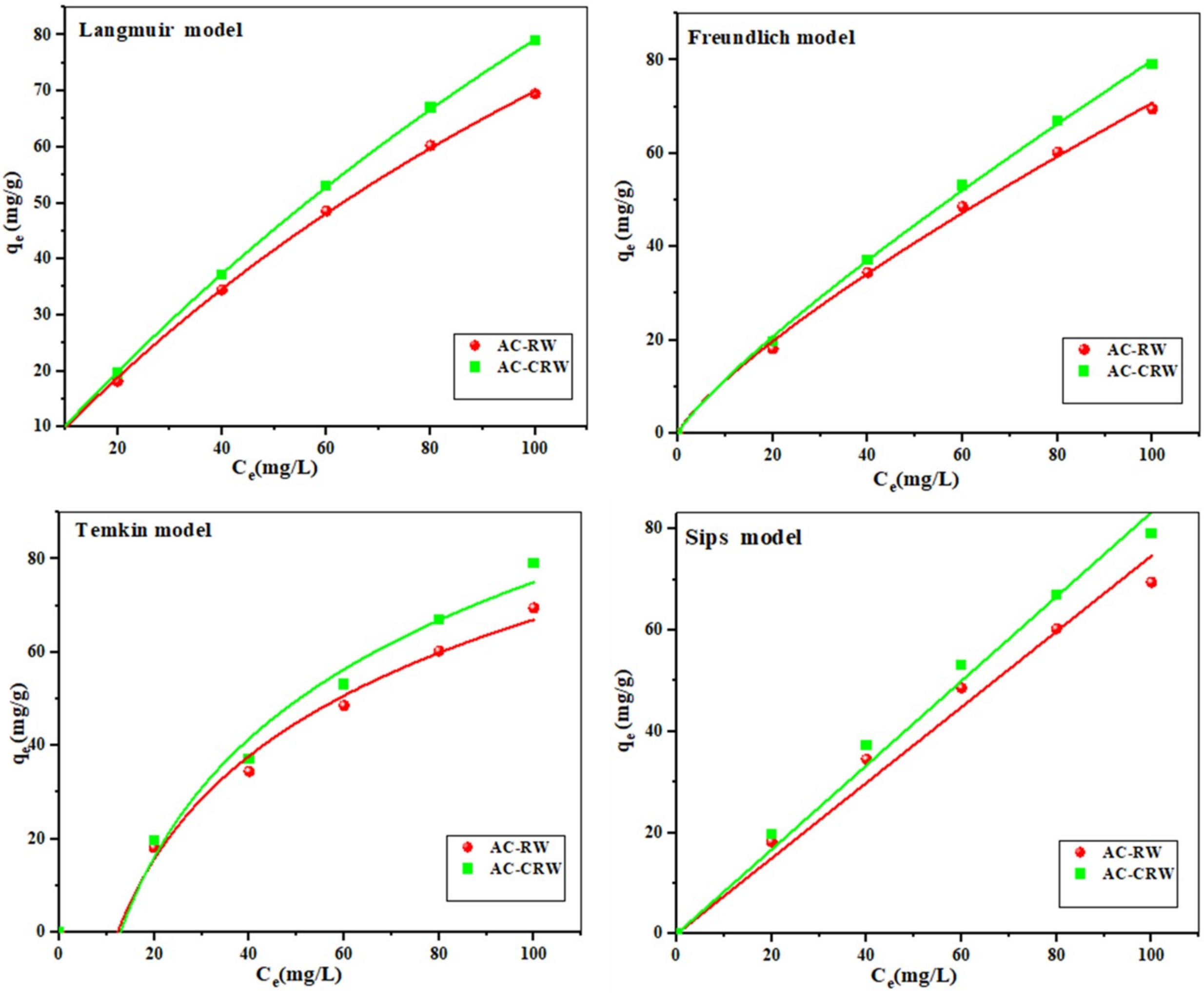

Fig. 13 Adsorption isotherm models *of MB by optimal AC-RW and AC-CRW.*

Table 5 Non-linear isotherm model parameters and statistical indicators for MB adsorption on optimal *AC-RW and AC-CRW at* 25°C.

Isotherm	**Parameters**	**Type of adsorbent**	
		AC-RW	**AC-CRW**
Langmuir	q_m (mg/g)	218.779	314.067
	K_L (L/mg)	0.0047	0.0033
	R^2 adjusted	0.99961	0.99993
	χ^2	0.266	0.062
Freundlich	K_F ((mg/g) (L/ mg)$^{1/n}$)	1.8477	1.69
	n	1.262	1.1941
	1/n	0.792	0.837
	R^2 adjusted	0.99744	0.99895
	χ^2	1.763	0.923
Temkin	B	31.865	36.598
	K_T(L.mg^{-1})	0.0818	0.0777
	R^2 adjusted	0.98985	0.98389
	χ^2	7.000	14.187
Spis	$K_{rp\ (mg/g)}$	0.747	0.833
	a	3×10^{-6}	0.0963
	g	-263.50	-221.88
	R^2 adjusted	0.96443	0.9803
	χ2	24.530	17.350

Table 6 Comparative study of the adsorption capacity of optimal AC-CRW and AC-RW for MB dye against different activated carbons.

Adsorbent	Time (min)	pH	T (°C)	qmax (mg/g)	Isotherm Model	Ref.
AC-CRW	70	7	25	314.06	Langmuir	This study
AC-RW	90	7	25	218.77	Langmuir	This study
Biomass-derived AC (general; chem-activation)	-	-	-	178.41	Langmuir	50
Spent coffee-ground carbon (acid/base-treated)	-	-	-	171.6 – 270.64	Langmuir	51
Raw olive-waste adsorbent (no high-T activation)	-	8	-	232.6 – 252.1	Langmuir	52
Hydrothermal carbon from palm kernel shell (KCS-2)	-	-	-	54.01	Langmuir	53
Cellulose-based activated carbon (CsCl)				176	Langmuir	54
Coffee-waste AC	60	6	25	176	Langmuir	55

16. Thermodynamic study

Table 7 Thermodynamic parameters for MB dye adsorption on AC-CRW and AC-RW adsorbent.

Type of AC	**ΔG° (KJ mol^{-1})**	**ΔH° (KJ mol^{-1})**	**ΔS° (Kj^{-1} mol^{-1})**

	283.15 K	288.15 K	298.15 K	308.5 K		
AC-CRW	−0.64	−1.94	−4.54	−7.14	72.63	259.94
AC-RW	−0.08	−0.82	−2.30	−3.77	41.75	147.80

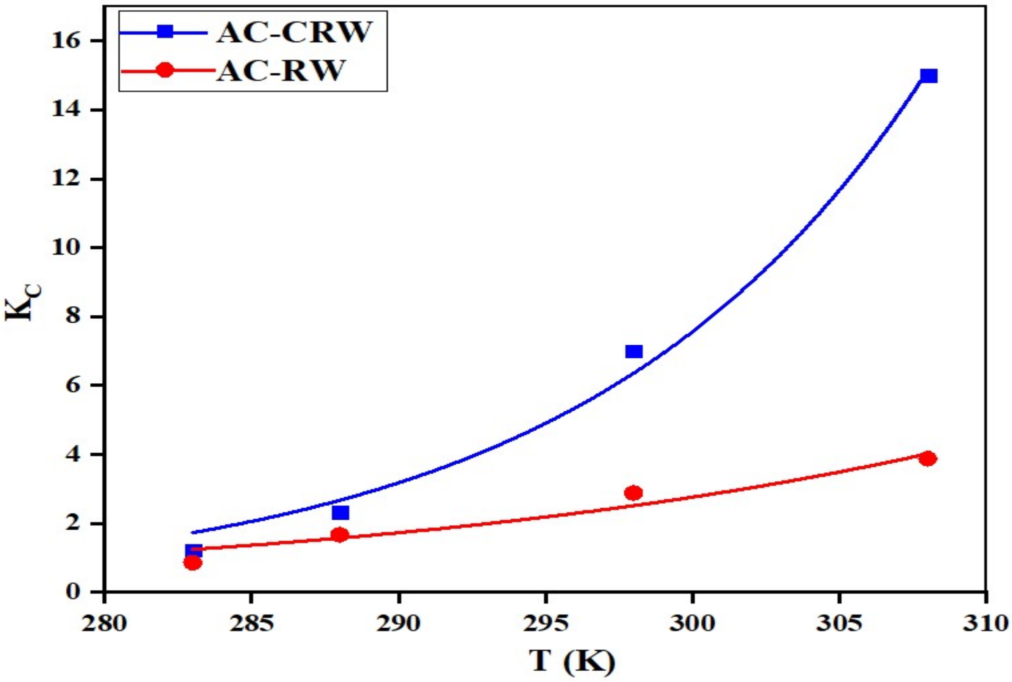

Fig. 14 Van't Hoff thermodynamic plot.

17. Adsorption Mechanism of MB on AC-RW and AC-CRW

Fig. 15 Proposed adsorption mechanism of MB onto AC-RW and AC-CRW.

18. Cyclic Stability and Regeneration Efficiency of Activated Carbons

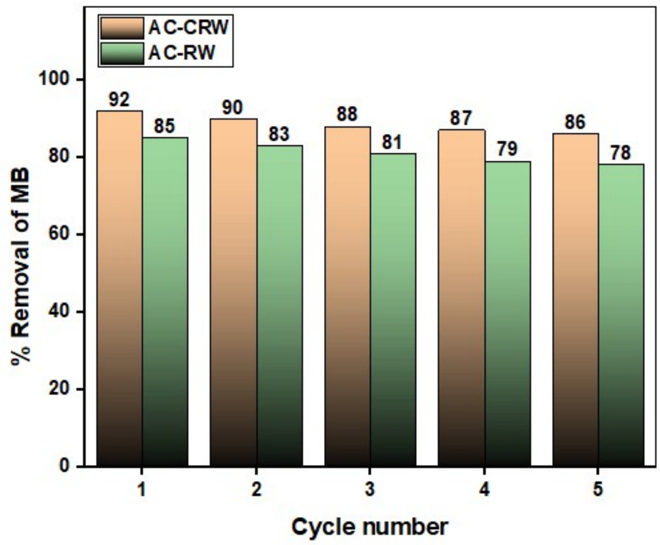

Fig. 16 Regeneration over five cycles: MB removal (%) by AC-CRW and AC-RW.