

Supporting Information

Remarkable synergy between sawdust biochar and attapulgite/diatomite after co-ball milling to adsorb methylene blue

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1. Removal rate R (%) and adsorption capacity Q (mg·g⁻¹):

$$R = \frac{C_0 - C}{C_0} \times 100\%$$

$$Q = \frac{(C_0 - C) \times V}{M}$$

Notes:

C₀ is the initial MB concentration, mg·L⁻¹;

C is the MB concentration after adsorption, mg·L⁻¹;

V is the volume of solution containing MB, L;

M is the amount of adsorbent added, g.

2. Adsorption kinetic models

Adsorption kinetic data were fitted with pseudo-first-order and pseudo-second-order models to evaluate potential adsorption mechanisms.

Pseudo first-level model (Al-Ghouti et al., 2009):

$$Q_t = Q_e(1 - e^{-K_1 t})$$

Pseudo secondary model (Al-Ghouti et al., 2009):

$$Q_t = \frac{Q_e^2 K_2 t}{1 + Q_e K_2 t}$$

Notes:

Q_e is the heavy metal adsorption capacity at equilibrium, mg·g⁻¹;

Q_t is the heavy metal adsorption capacity at time t, mg·g⁻¹;

K₁ is the pseudo-first-order adsorption rate constant, L·min⁻¹;

K₂ is the pseudo-second-order adsorption rate constant, g·(mg·min)⁻¹.

3. Adsorption isotherm models

Adsorption isotherms were fitted with the Langmuir and Freundlich models, which can be described as the following equations, respectively (Tayibi et al., 2021):

$$Q_e = K_F C_e^{\frac{1}{n}}$$

$$Q_e = Q_m \frac{K_L C_e}{1 + K_L C_e}$$

Notes:

C_e is the equilibrium concentration, $\text{mg}\cdot\text{L}^{-1}$;

Q_e is the equilibrium adsorption capacity, $\text{mg}\cdot\text{g}^{-1}$;

Q_m is the maximum theoretical adsorption capacity, $\text{mg}\cdot\text{g}^{-1}$;

K_L is the Langmuir equilibrium constant, $\text{L}\cdot\text{mg}^{-1}$;

K_F and n are the Freundlich equilibrium constants, $(\text{mg}\cdot\text{g}^{-1}) (\text{L}\cdot\text{min}^{-1})^{1/n}$.

4. Table S1. Adsorption capacity and removal rate of MB by various adsorbents

Samples	Adsorption capacity (mg·g ⁻¹)	Removal rate (%)
MBC	69.49	44.19
BC	1.10	0.46
MATP	31.55	21.72
ATP	74.34	47.47
MDE	13.79	13.34
DE	0	0.38
MABC10%	158.10	98.95
MABC20%	156.59	98.02
MABC30%	150.18	93.43
MABC40%	141.33	88.74
MABC10%-CK	85.14	52.87
MDBC10%	145.66	91.02
MDBC20%	145.78	91.17
MDBC30%	140.20	87.74
MDBC40%	143.19	90.39
MDBC10%-CK	68.69	42.17

5. Table S2. Properties of selected ball milled adsorbents.

Samples	C (%)	H (%)	O (%)	N (%)	H/C	O/C	CEC (cmol ⁺ ·kg ⁻¹)	Surface Area (m ² ·g ⁻¹)	Pore Volume (cm ³ ·g ⁻¹)	Mean Pore Diameter (nm)
MBC	80.8	2.6	13.0	0.2	0.39	0.12	10.8	400	0.19	0.96
MABC10%	70.6	2.6	17.3	0.2	0.45	0.18	82.9	313	0.16	0.99
MDBC10%	70.7	2.4	14.3	0.1	0.41	0.15	25.3	330	0.16	0.97

6. Table S3. Fitting parameters of adsorption isotherms and adsorption kinetics of selected ball-milled adsorbents.

Samples	Langmuir		Freundlich			Pseudo-first-order			Pseudo-second-order			
	Q_m ($\text{mg}\cdot\text{g}^{-1}$)	k_L ($\text{L}\cdot\text{mg}^{-1}$)	R^2	$1/n$	k_F ($\text{mg}^{1-1/n}\cdot\text{L}^{1/n}\cdot\text{g}^{-1}$)	R^2	Q_e ($\text{mg}\cdot\text{g}^{-1}$)	k_1 (h^{-1})	R^2	Q_e ($\text{mg}\cdot\text{g}^{-1}$)	k_2 ($\text{g}\cdot\text{mg}^{-1}\cdot\text{h}^{-1}$)	R^2
MBC	68.2	5.93	0.998	0.043	57.4	0.969	64.5	0.247	0.319	67.2	0.006	0.725
MABC10%	183	22.7	0.842	0.0512	0.0512	0.804	145	0.110	0.795	154	0.001	0.969
MDBC10%	155	1.19	0.993	0.0568	0.0568	0.813	124	0.112	0.635	133	0.001	0.877

7. Table S4. Figure 1 and Figure 6 Analysis Data Sharing.

Samples	Absorbance (a)	Q_e (a)	Absorbance (b)	Q_e (b)	Mean standard deviation
MBC	21.80	71.92	22.40	69.49	1.71
BC	43.50	0.37	43.30	1.10	0.52
MATP	32.30	37.97	34.00	31.55	4.54
ATP	20.40	77.58	21.20	74.34	2.29
MDE	34.70	28.90	38.70	13.79	10.68
DE	39.30	1.21	39.60	0	0.85
MABC10%	0.36	158.55	0.47	158.10	0.31
MABC20%	0.72	157.07	0.84	156.59	0.34
MABC30%	2.77	148.81	2.43	150.18	0.97
MABC40%	4.30	142.63	4.62	141.33	0.91
MABC10%-CK	20.7	84.04	20.40	85.13	0.77
MDBC10%	3.56	145.62	3.55	145.66	0.03
MDBC20%	3.47	145.98	3.52	145.78	0.14
MDBC30%	4.81	140.57	4.90	140.20	0.25
MDBC40%	3.45	146.06	4.16	143.19	2.03
MDBC10%-CK	23.20	66.26	22.60	68.69	1.71

Samples		Absorbance (a)	Qe (a)	Absorbance (b)	Qe (b)	Mean standard deviation
MBC	2	27.30	50.11	27.25	50.31	0.18
	3	26.70	53.20	26.95	52.2	0.88
	5	23.55	66.62	22.70	69.98	2.97
	7	21.20	73.24	20.50	76.11	2.53
	9	17.40	89.96	16.70	92.77	2.49
	10	15.40	93.04	15.25	93.69	0.57
MABC10%	2	14.57	101.33	13.57	105.35	3.55
	3	10.55	117.80	10.62	117.5	0.26
	5	2.20	151.25	2.50	150.06	1.05
	7	2.06	151.57	2.52	149.68	1.66
	9	1.07	155.69	0.74	157.00	1.15
	10	1.12	155.10	2.36	149.71	4.76
MDBC10%	2	8.65	125.18	8.70	124.98	0.17
	3	8.35	126.60	8.62	125.5	0.97
	5	3.32	146.81	3.27	147.01	0.17
	7	2.05	151.61	2.14	151.24	0.32
	9	0.30	158.77	0.28	158.87	0.09
	10	0.16	159.26	0.15	159.34	0.06

Samples		Absorbance (a)	Qe (a)	Absorbance (b)	Qe (b)	Mean standard deviation
MBC	0	23.70	73.02	23.60	73.39	0.32
	0.001	22.20	69.38	21.80	71.02	1.44
	0.01	21.15	74.97	21.35	74.17	0.71
	0.1	18.30	85.77	16.35	93.68	6.99
MABC10%	0	2.74	149.94	2.55	150.64	0.61
	0.001	2.37	150.30	2.12	151.32	0.90
	0.01	1.22	155.09	1.42	154.29	0.71
	0.1	1.09	155.55	0.87	156.45	0.78
MDBC10%	0	6.77	135.13	6.82	134.95	0.16
	0.001	11.17	114.38	11.10	114.69	0.27
	0.01	9.82	120.50	10.90	116.18	3.82
	0.1	4.65	141.14	4.92	140.02	0.98

References

- Al-Ghouti, M.A., Khraisheh, M.A., Ahmad, M.N., Allen, S. 2009. Adsorption behaviour of methylene blue onto Jordanian diatomite: a kinetic study. *J Hazard Mater*, **165**(1-3), 589-98.
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