Supplementary information (SI)

Figure S1. The pseudo-first-order plots for the adsorption of hydRB5 at 40 mg L^{-1} (■), 60 mg L^{-1} (◆), 80 mg L^{-1} (▲), and 100 mg L^{-1} (●) on the membranes: **(a)** M0, **(b)** M1, **(c)** M2, and **(d)** M3.

Figure S2. The pseudo-second-order plots for the adsorption of hydRB5 at 40 mg L^{-1} (■), 60 mg L^{-1} (◆), 80 mg L^{-1} (▲), and 100 mg L^{-1} (●) on the membranes: **(a)** M0, **(b)** M1, **(c)** M2, and **(d)** M3.

Figure S3. The intraparticle diffusion plots for the adsorption of hydRB5 at 40 mg L^{-1} (■), 60 mg L^{-1} (◆), 80 mg L^{-1} (▲), and 100 mg L^{-1} (●) on the membranes: **(a)** M0, **(b)** M1, **(c)** M2, and **(d)** M3.

Figure S4. The isotherm models for the adsorption of hydRB5 on the membrane M2: **(a)** Langmuir, **(b)** Freundlich, and **(c)** Temkin.

Figure S5. (a) The residuals as a function of membrane adsorption, **(b)** the residual density histogram by model, **(c)** the Q-Q plot by model, and d) the location-scale plots by model for each membrane.

Figure S6. The interactive 3D-surface plots for the M0, M1, M2, and M3 membranes.

Figure S7. (a) The residuals of the log-logistic models as a function of the adsorption capacity, and (**b)** the residual histograms for each model and per membrane.

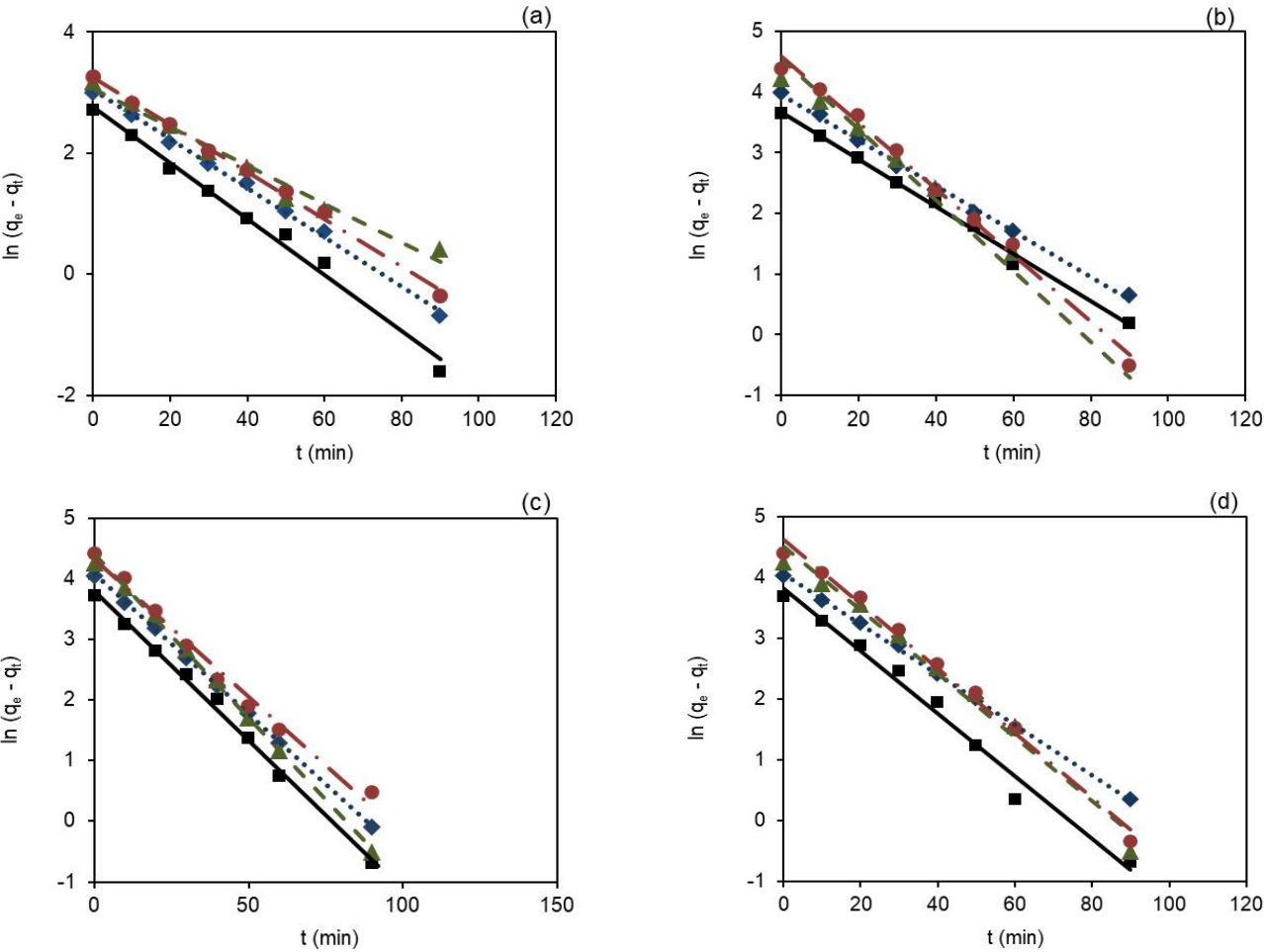**Figure S8.** The desirability functions for the adsorption capacity for each membrane.
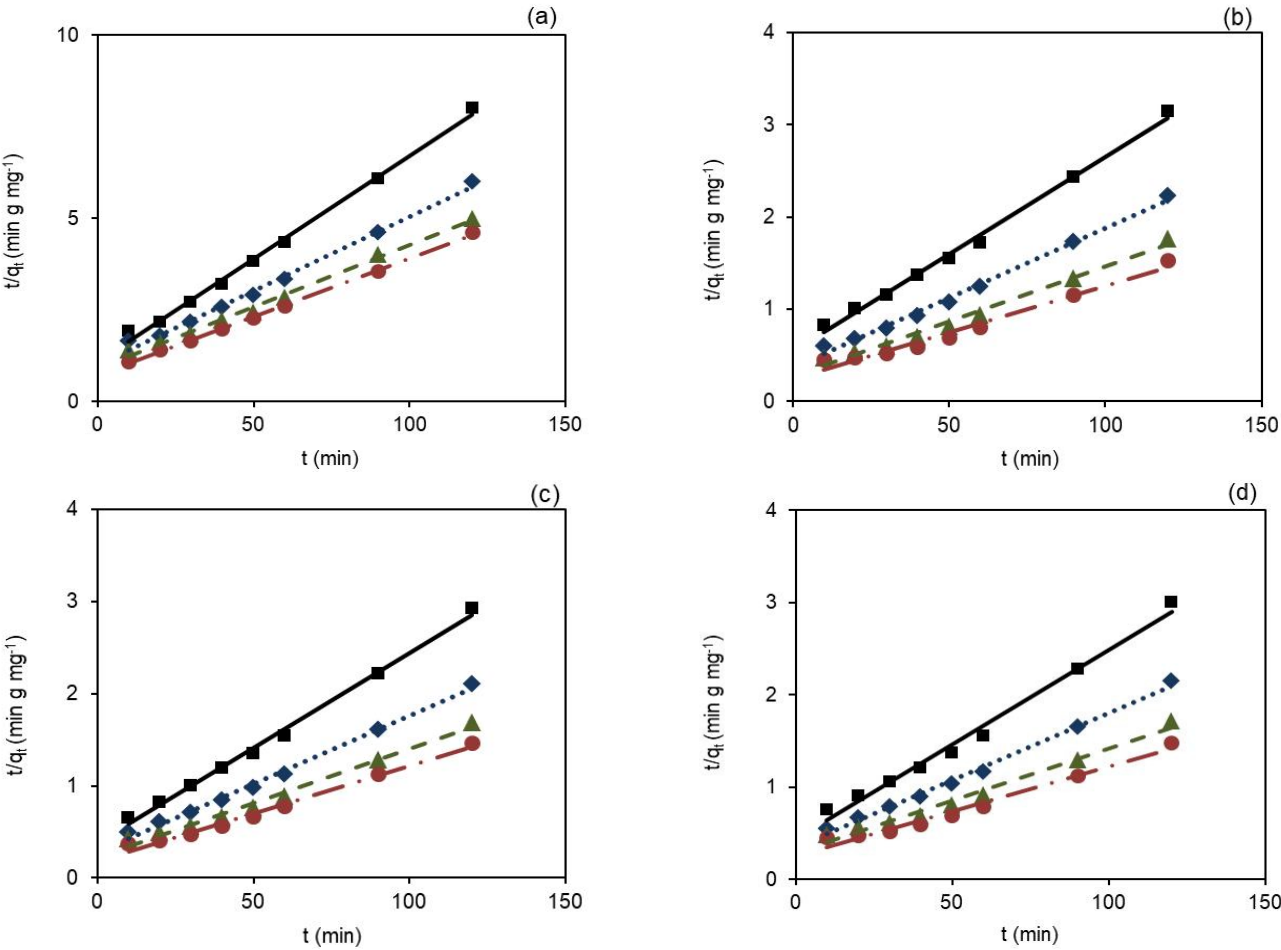
Figure S1.

Figure S2.

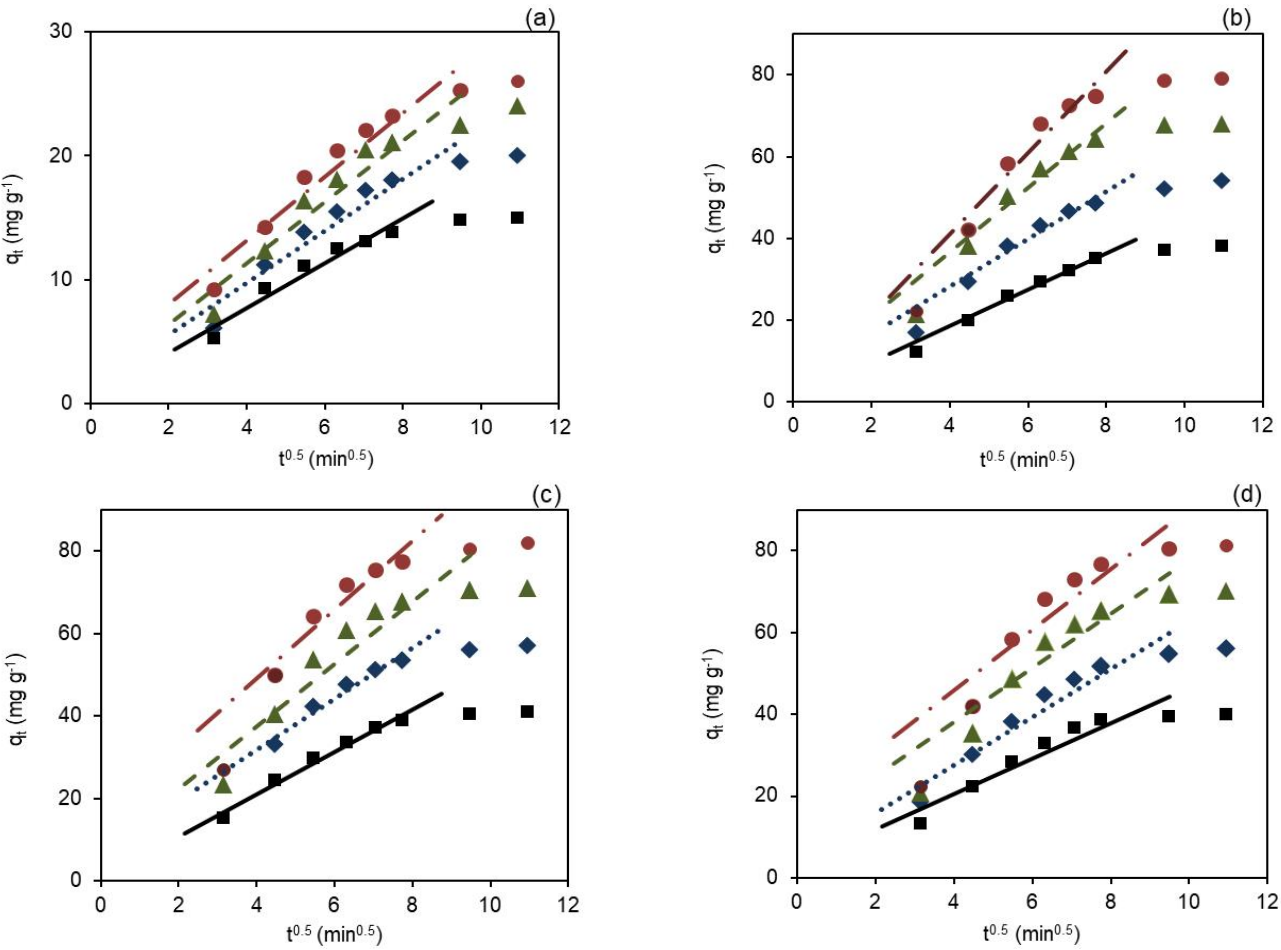Figure S3.

Figure S4.

(a)

$y = 0.0095x + 0.2278$
$R^2 = 0.9908$

C_e/q_e (g L^{-1})

C_e (mg L^{-1})

(b)

$y = 0.5228x + 2.0979$
$R^2 = 0.9358$

ln q_e

ln C_e

(c)

$y = 22.139x - 16.525$
$R^2 = 0.9588$
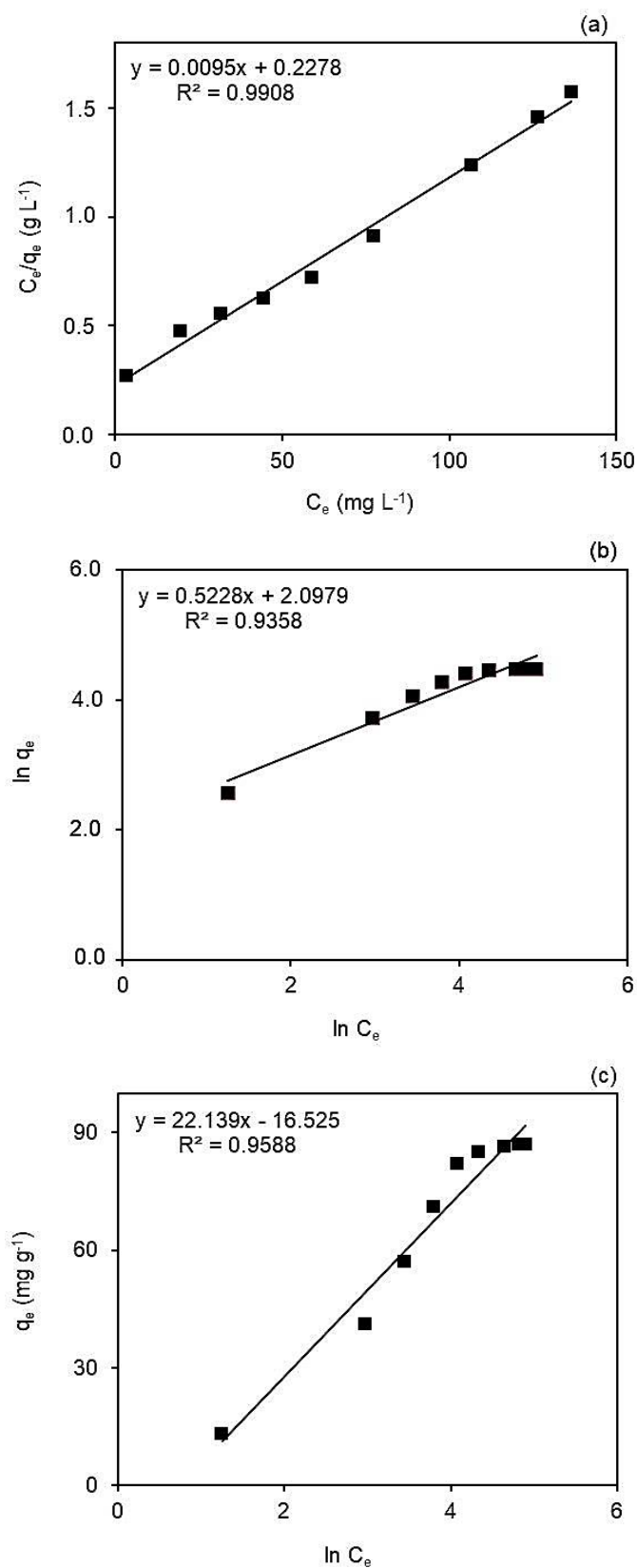
q_e (mg g^{-1})

ln C_e

Figure S5.

Residuals represent the variation left unexplained by a model. Their study, and the identification of the patterns they follow, may give an indication that the models proposed have underlying problems.

The membrane models' residuals were plotted against the adsorption capacity (Figure S5a) and used to check the linear relationship assumptions. Ideally, the residual plot will show no fitted pattern. The presence of a pattern may indicate a problem with some aspects of the linear model. For the models proposed here, no pattern was found in the residual plot. This suggests a linear relationship between the predictors and the outcome variables, as they seem to be distributed equally around the horizontal line in a random fashion manner. In addition, the histograms of the residuals were also plotted in Figure S5b.

(a)
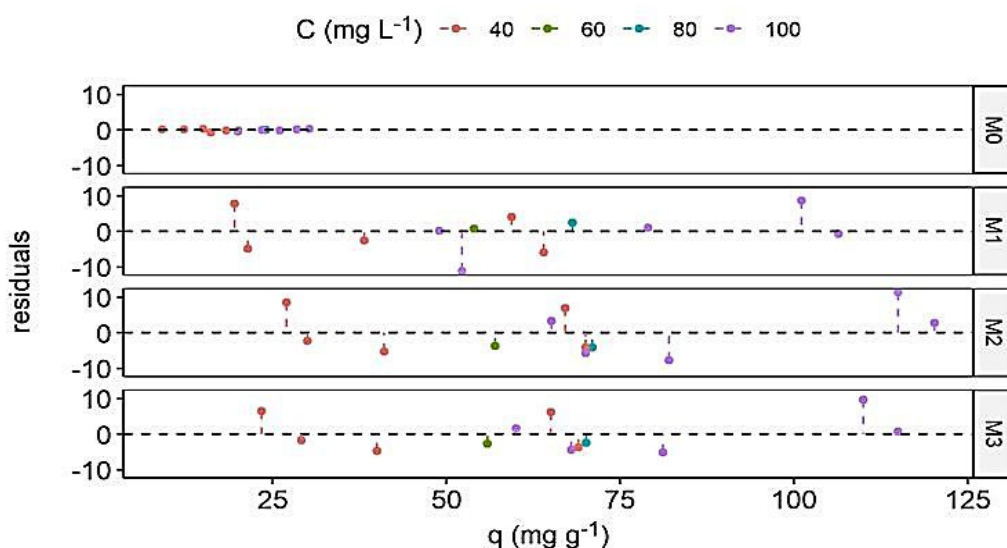
(b)
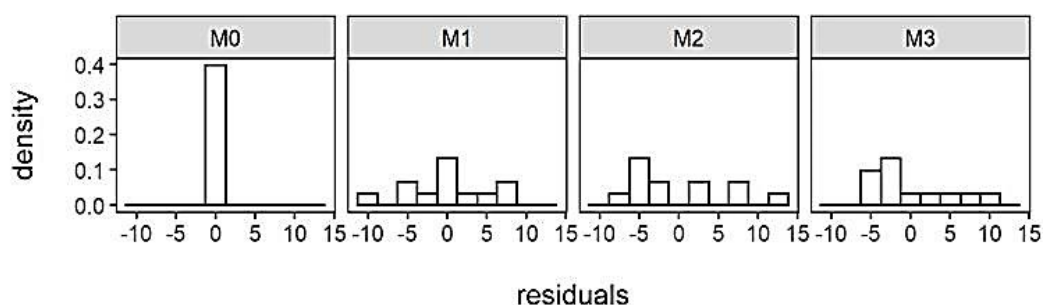
Since the residuals should be normally distributed, a Q-Q plot was used to explore the theoretical quantiles of the models against the standardized residuals (samples in Figure S5c), to visually check the normality assumption. The normal probability plot of residuals should approximately follow a straight line. As most of the residual follow the predicted line, the residuals were assumed to be normally distributed.

(c)
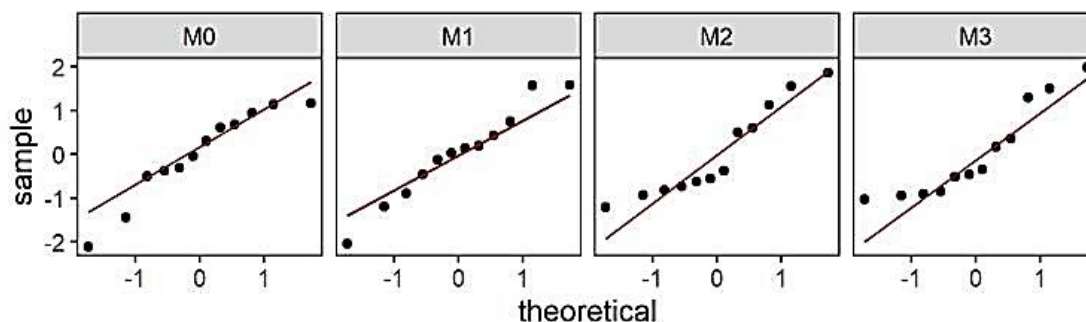
The homogeneity of variance was verified with a scaled location graph. These graphs (Figure S5d) show that the residuals are spread out along with the predictor intervals. Ideally, a horizontal line with a non-increasing or non-decreasing trend should be obtained to suggest constant variance in the residuals (homoscedasticity). From the graphs obtained, the residues are considered homoscedastic.

(d)

Figure S6.

M0.data.2.html

M1.data2.html

M2.data2.html

M3.data2.html
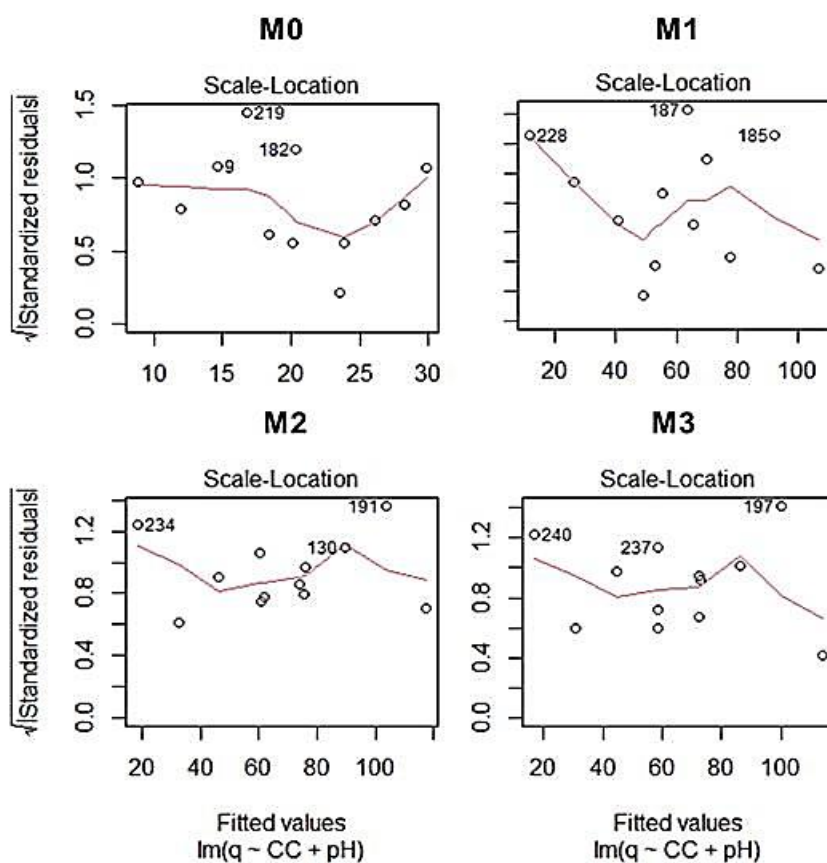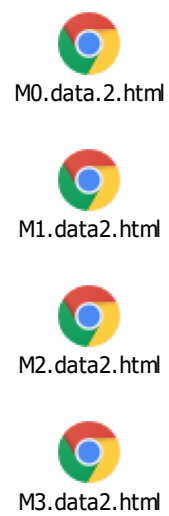
Figure S7.

The residuals of the log-logistic models with five parameters were plotted as a function of the adsorption capacity (Figure S7a), and used to verify the linear relationship assumptions. No pattern was identified in the residual plots. Residual histograms were also plotted to confirm a normal distribution (Figure S7b).

(a)

(b)

Figure S8.

M0

3   11

pH = 3

40   100

C = 100 mg L^{-1}

0   240

time = 93.6 min

0   30.2

Adsorption = 30.2 mg g^{-1}

M1

3   11

pH = 3

40   100

C = 100 mg L^{-1}

0   240

time = 62.5 min

0   106.4

Adsorption = 106.4 mg g^{-1}

M2

3   11

pH = 3

40   100

C = 100 mg L^{-1}

0   240

time = 68.0 min

0   120.2

Adsorption = 120.2 mg g^{-1}

M3

3   11

pH = 3

40   100

C = 100 mg L^{-1}

0   240

time = 67.2 min

0   115.0

Adsorption = 115.0 mg g^{-1}
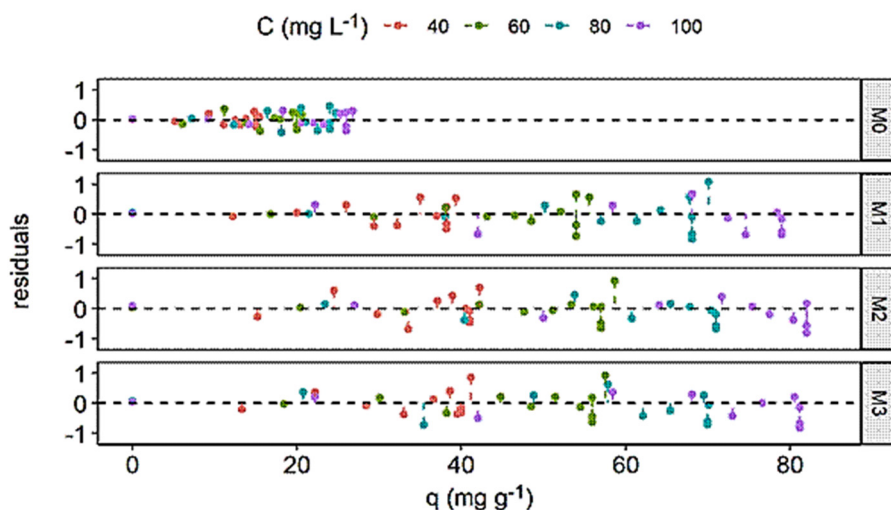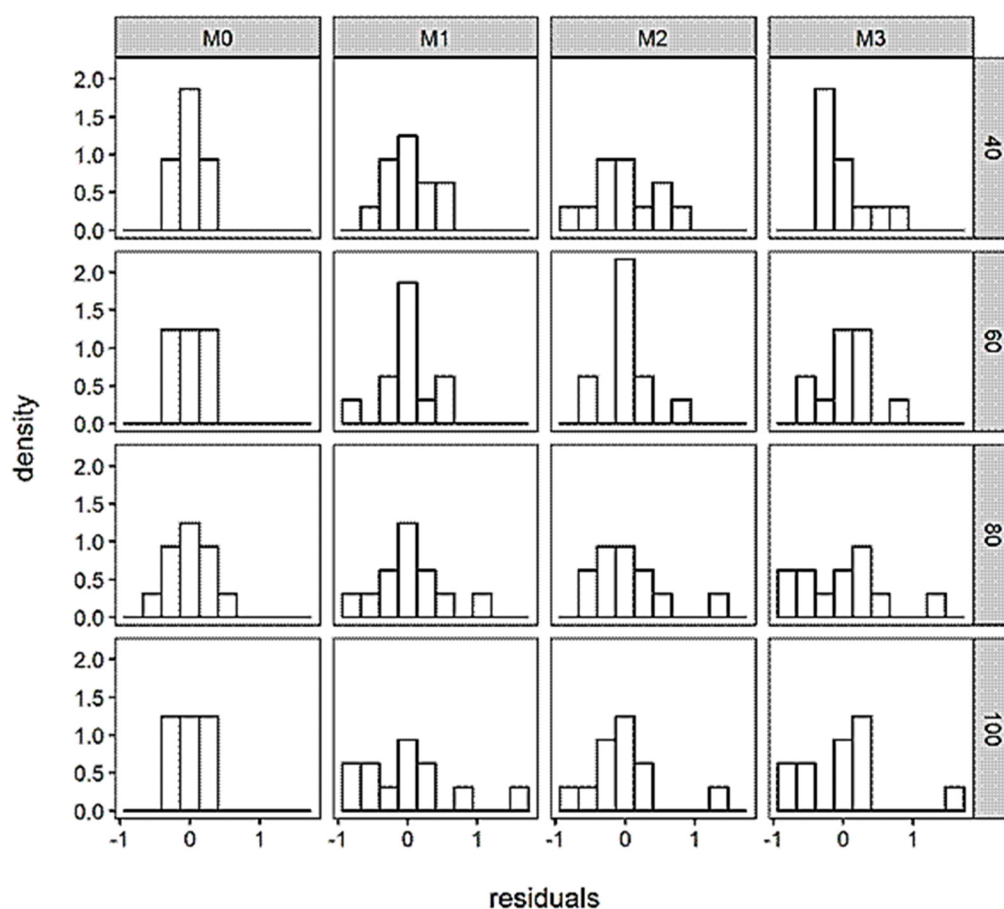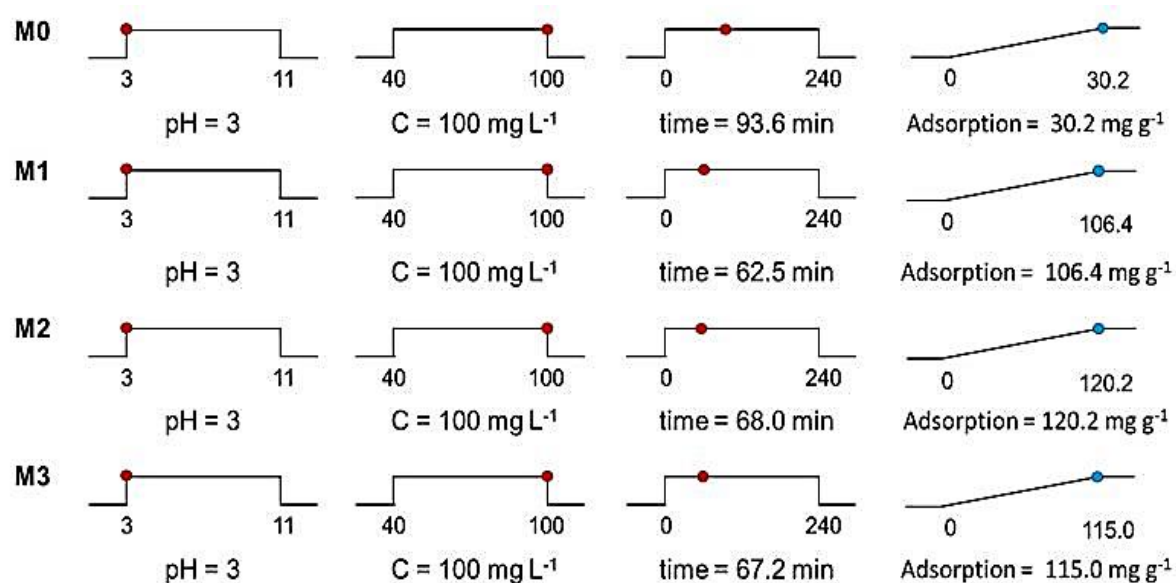
Table S1. The parameters of isothermal models for the adsorption of hydRB5 on the M2 membrane, in pH~7 and 25 °C.

Table S2. The ANOVA results for the response surface quadratic model. Significance levels are expressed as: (.) > 0.05; (*) ≤ 0.05; (**) ≤ 0.01; (***) ≤ 0.001.

Table S3. The equation models where q_t is the adsorption capacity of membranes, pH is the acidity of the medium and C is the dye concentration.

Table S4. The functions used to adjust membrane adsorption capacity as a function of time and initial dye concentration (Akaike Information Criterion, AIC, was used for each model).

Table S5. The time-dependent and parameter-dependent log-logistic formula per membrane and initial dye concentration.

Table S6. The optimal conditions found by the RSM at t = 120 min (Top position), the optimal dye concentration at 100 mg L^{-1} and pH = 2 (Medium position), and the pseudo-optimal conditions at 40 mg L^{-1} and pH = 2 (Bottom position).

Table S1.

Membrane	Adsorption isotherm	Parameters	Calculated values
M2	Langmuir	q_{max} (mg g^{-1})	105.26
		K_L (L mg^{-1})	0.042
		R^2	0.9908
	Freundlich	K_F (mg g^{-1})(L mg^{-1})$^{1/n}$	8.15
		n	1.91
		R^2	0.9358
	Temkin	K_T (L mg^{-1})	0.44
		B (J mol^{-1})	22.14
		R^2	0.9588

Table S2.

Source	M0			M1			M2			M3		
	Mean Square	F value	Prob > F; p-value	Mean Square	F value	Prob > F; p-value	Mean Square	F value	Prob > F; p-value	Mean Square	F value	Prob > F; p-value
CC	338.6	3966.32	1.08×10^{-9} (***)	3528	77.52	1.19×10^{-4} (***)	4822	94.11	6.88×10^{-5} (***)	4415	122.67	3.23×10^{-5} (***)
pH	113.8	1332.58	2.82×10^{-8} (***)	4225	92.84	7.15×10^{-5} (***)	3874	75.61	1.28×10^{-4} (***)	3895	108.22	4.62×10^{-5} (***)
CC^2	7.4	86.92	8.63×10^{-5} (***)	7.0	0.15	0.711 (.)	38	0.735	0.424 (.)	16	0.436	0.533 (.)
pH^2	1.8	21.09	0.004 (**)	3	0.068	0.803 (.)	48	0.936	0.371 (.)	15	0.409	0.546 (.)
$CC\times pH$	0.5	5.63	0.055 (.)	67	1.48	0.269 (.)	52	1.009	0.354 (.)	31	0.861	0.389 (.)
Residuals	0.1			46			51			36		
Model summary statistics												
F-statistic	813.7			99.58			87.98			134.8		
p-value	2.06×10^{-9} (***)			7.27×10^{-7} (***)			1.24×10^{-6} (***)			1.96×10^{-7} (***)		
R^2	0.998			0.957			0.951			0.968		
R_{adj}^2	0.997			0.947			0.941			0.961		
R_{pred}^2	0.994			0.917			0.910			0.941		
Std. Dev.	0.376			6.239			7.030			5.551		
PRESS	2.824			675.361			820.249			507.665		

Table S3.

Membrane	Equation model
M0	$q_t = 4.19 + 0.472 \times C - 0.305 \times pH - 0.063 \times pH^2 - 0.002 \times C^2$ (13)
M1	$q_t = 66.88 + 0.620 \times C - 7.270 \times pH$ (S14)
M2	$q_t = 66.00 + 0.720 \times C - 6.960 \times pH$ (S15)
M3	$q_t = 65.91 + 0.690 \times C - 6.980 \times pH$ (S16)

Table S4.

		AIC per model[1]												
		log-logistic (LL)			**Weibull type 1 and type 2 (W1 and W2)**				**Brain-Cousens (BC)**		**Asymptotic Regression (AR)**		**Michaelis-Menten (MM)**	
Membrane	**C (mg L^{-1})**	**LL.3**	**LL.4**	**LL.5**	**W1.3**	**W1.4**	**W2.3**	**W2.4**	**BC.4**	**BC.5**	**AR.2**	**AR.3**	**MM.2**	**MM.3**
M0	40	-2.4	-0.4	1.0	4.2	6.2	3.8	5.7	-1.0	1.0	2.7	4.7	26.1	27.8
M0	60	9.8	11.8	9.7	18.5	20.5	6.1	8.0	9.6	11.6	4.2	6.2	34.1	35.6
M0	80	16.0	18.0	15.4	25.1	27.1	15.9	17.8	16.9	18.8	14.1	16.1	37.3	38.7
M0	100	20.1	22.1	8.9	28.6	30.6	5.3	7.3	18.4	20.4	14.5	15.2	35.0	37.0
M1	40	32.7	34.7	20.4	41.2	43.2	17.6	19.6	31.6	33.5	15.8	17.7	49.6	51.6
M1	60	27.0	28.9	21.4	40.4	42.4	27.1	29.1	27.1	29.0	25.2	27.1	58.0	59.5
M1	80	42.6	44.4	28.9	53.1	55.1	26.9	28.9	42.8	44.7	40.1	40.6	70.2	71.7
M1	100	49.2	50.9	35.3	60.1	62.0	37.9	39.9	50.4	52.2	58.8	59.2	78.7	80.2
M2	40	35.8	37.9	24.9	42.2	44.2	22.0	24.0	35.6	37.6	21.1	22.8	51.6	53.5
M2	60	37.9	39.8	21.8	47.0	48.9	21.6	23.6	37.9	39.8	19.7	21.7	60.3	62.0
M2	80	46.8	48.6	30.3	55.9	57.9	30.6	32.6	47.5	49.4	44.1	44.9	72.1	73.7
M2	100	35.0	36.9	31.4	48.0	50.0	41.8	43.7	37.0	38.9	49.7	50.6	74.8	76.4
M3	40	40.7	42.7	21.8	47.4	49.4	25.6	27.6	40.9	42.8	30.0	31.5	57.5	59.5
M3	60	41.7	43.6	22.9	50.5	52.4	24.0	26.0	41.0	42.9	22.1	24.1	59.8	61.4
M3	80	51.4	53.1	32.9	60.5	62.4	34.7	36.7	51.5	53.3	47.5	48.2	72.6	74.1
M3	100	49.0	50.7	33.2	60.7	62.6	34.9	36.9	49.8	51.6	57.4	57.6	78.6	80.0
Mean		33.3	35.2	22.5	42.7	44.7	23.5	25.5	33.6	35.5	29.2	30.5	57.3	58.9

[1]log-logistic (LL), Weibull type 1 and type 2 (W1 and W2, respectively), Brain-Cousens (BC), Asymptotic Regression (AR) and Michaelis-Menten (MM) functions using 2, 3, 4 or 5 parameters (.2, .3, .4 and .5 respectively).

Table S5.

Membrane	C (mg L^{-1})	$q = \dfrac{d}{\left(1+e^{(b\times(ln(t)-e))}\right)^{f}}$				t$_{50}$ (min)	t$_{95}$ (min)
		b	d	e	f		
M0	40	-1.67	15.42	3.03	0.73	15.7 ± 0.5	99.0 ± 15.1
M0	60	-1.98	20.52	3.52	0.48	18.5 ± 0.6	101.3 ± 14.4
M0	80	-1.96	24.59	3.58	0.48	19.6 ± 0.7	109.4 ± 16.3
M0	100	-2.35	26.53	3.84	0.29	17.6 ± 0.5	93.6 ± 8.6
M1	40	-2.69	38.83	3.88	0.27	19.2 ± 0.6	86.5 ± 7.7
M1	60	-1.99	55.22	3.47	0.49	18.2 ± 0.4	97.5 ± 7.8
M1	80	-2.71	69.05	3.58	0.34	17.6 ± 0.4	69.9 ± 4.1
M1	100	-3.06	79.75	3.54	0.34	18.5 ± 0.4	62.5 ± 3.6
M2	40	-3.09	41.51	3.83	0.21	15.9 ± 0.6	69.5 ± 6.4
M2	60	-2.61	57.79	3.66	0.29	16.2 ± 0.3	73.1 ± 4.4
M2	80	-3.04	71.73	3.60	0.28	16.9 ± 0.4	62.4 ± 3.4
M2	100	-2.25	83.07	3.21	0.52	15.7 ± 0.3	68.0 ± 4.4
M3	40	-4.21	40.31	3.87	0.17	17.8 ± 0.5	60.9 ± 3.8
M3	60	-2.86	56.66	3.85	0.25	18.4 ± 0.4	79.0 ± 4.9
M3	80	-3.30	70.78	3.77	0.26	19.5 ± 0.5	68.4 ± 3.9
M3	100	-2.95	82.13	3.59	0.35	19.2 ± 0.4	67.2 ± 3.5

Table S6.

	Membrane	C^a (mg L^{-1})	pH	q_{pred}^b (mg g^1)	q_{exp}^c (mg g^{-1})	Δ^d	Δ (%)^e
Optimal conditions found	M0	100	3	29.91	30.20	0.29	0.97
	M1	100	3	107.01	106.40	0.61	0.57
	M2	100	3	117.52	120.25	2.73	2.27
	M3	100	3	114.25	115.00	0.75	0.65
Other conditions explored	M0	100	2	32.50	30.53	1.97	6.45
	M1	100	2	111.50	114.27	2.77	2.43
	M2	100	2	125.20	124.48	0.72	0.58
	M3	100	2	120.00	121.23	1.23	1.01
	M0	40	2	20.50	19.02	1.48	7.78
	M1	40	2	68.80	77.12	8.32	10.79
	M2	40	2	74.50	81.04	6.54	8.07
	M3	40	2	73.00	79.66	6.66	8.37

[a] The dye concentration in mg L^{-1}.

[b] The predicted (pred.) adsorption in mg g^{-1}.

[c] The experimental (exp.) adsorption in mg g^{-1}.

[d] The difference between the predicted and real value is calculated as $\Delta = |q_{pred.} - q_{exp.}|$

[e] The difference percentage is calculated as $\Delta\% = 100 \times \Delta / q_{exp.}$