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Supplementary Materials

Geopolymers Based on Fly Ash for Organic Dye Removal from Water

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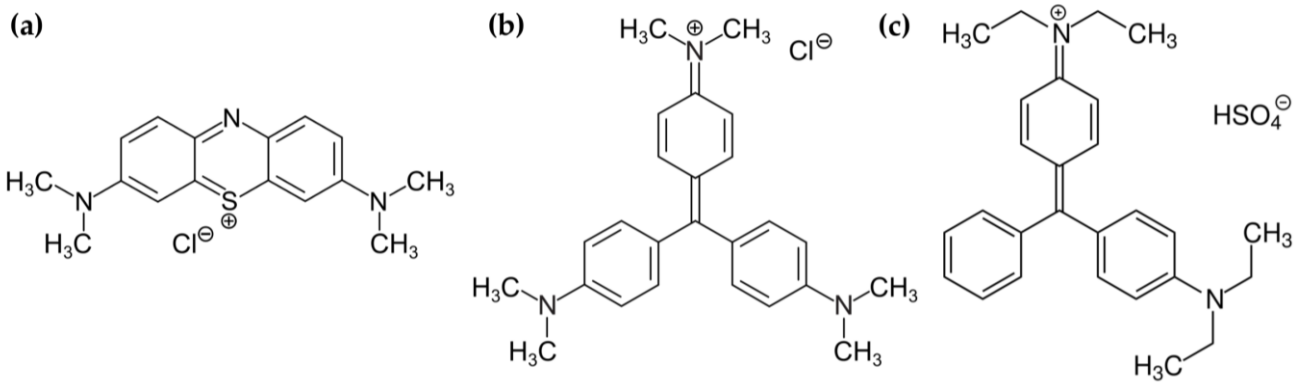


Figure S1. Chemical structures of organic dyes: (a) MB, (b) CV and (c) BG.

Table S1. Physicochemical properties of selected dyes.

	Methylene Blue	**Crystal Violet**	**Brilliant Green**
Molecular Formula	$C_{16}H_{18}ClN_3S$	$C_{25}H_{30}ClN_3$	$C_{27}H_{34}N_2O_4S$
Molecular Weight, g/mol	319.9	408.0	482.6
Melting Point, °C	100-110	215	210
Solubility in water, g/L	43.6	4.0	40.0
pK_a	3.8 7.5	8.64	5.36

Table S2. The sampling sites of raw wastewater and pH of wastewater samples.

Sample	**Latitude**	**Longitude**	**pH**
W1	44.8741	20.4548	9.16
W2	44.8084	20.4566	8.20
W3	44.8271	20.4672	8.28

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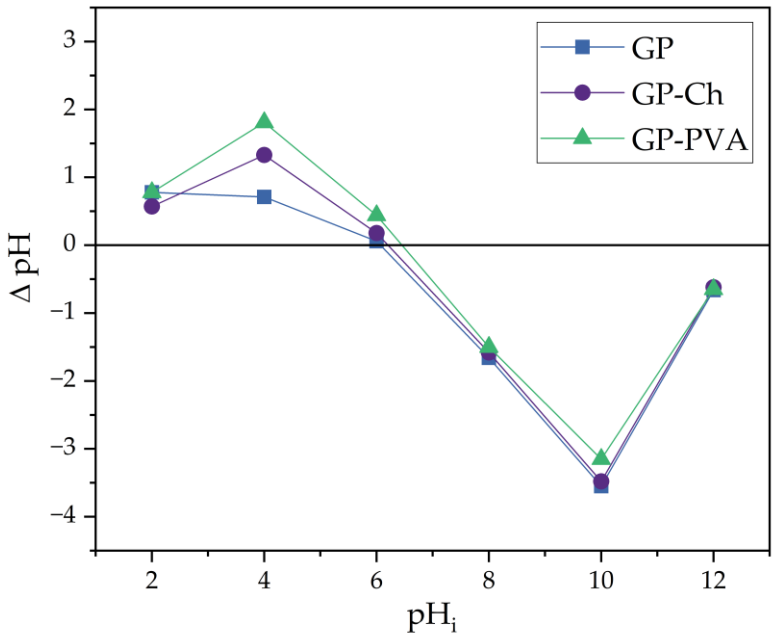


Figure S2. pH$_{PZC}$ values of GP, GP-Ch, and GP-PVA materials

Table S3. Analysis of Variance (ANOVA) of the Box–Behnken Design Model

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Model	31	6.34922	0.204814	37.35	0.000
Linear	8	2.50308	0.312885	57.06	0.000
pH	1	0.00024	0.000241	0.04	0.834
Initial concentration	1	0.00105	0.001046	0.19	0.663
Temperature	1	0.00035	0.000347	0.06	0.802
Contact time	1	0.99732	0.997322	181.89	0.000
Geopolymer	2	0.00580	0.002902	0.53	0.590
Dye	2	0.43540	0.217700	39.70	0.000
Square	4	0.86735	0.216837	39.55	0.000
pH*pH	1	0.17520	0.175201	31.95	0.000
Initial concentration*Initial concentration	1	0.06592	0.065916	12.02	0.001
Temperature*Temperature	1	0.00932	0.009316	1.70	0.195
Contact time*Contact time	1	0.49813	0.498132	90.85	0.000
2-Way Interaction	19	0.70889	0.037310	6.80	0.000
pH*Initial concentration	1	0.00013	0.000132	0.02	0.877
pH*Temperature	1	0.00727	0.007266	1.33	0.252
pH*Contact time	1	0.06990	0.069898	12.75	0.001
pH*Geopolymer	2	0.00670	0.003352	0.61	0.544
pH*Dye	2	0.17354	0.086772	15.83	0.000
Initial concentration*Geopolymer	2	0.00376	0.001879	0.34	0.710
Initial concentration*Dye	2	0.21834	0.109171	19.91	0.000
Temperature*Geopolymer	2	0.00146	0.000729	0.13	0.876
Contact time*Geopolymer	2	0.01662	0.008308	1.52	0.224
Geopolymer*Dye	4	0.06996	0.017491	3.19	0.015
Error	130	0.71280	0.005483		
Lack-of-Fit	121	0.71047	0.005872	22.66	0.000
Pure Error	9	0.00233	0.000259		

Table S4. Model equations obtained using the Box–Behnken design.

Geopolymer	Dye	Equations
GP	MB	$E\ (\%) = -1.643 + 0.503\ pH + 0.0032\ C_0 + 0.0093\ T + 0.01211\ t - 0.03690\ pH^2 - 0.000019\ C_0^2 - 0.000322\ T^2 - 0.000030\ t^2 - 0.00076\ pH \cdot C_0 + 0.00242\ pH \cdot T - 0.000594\ pH \cdot t$
	CV	$E\ (\%) = -1.653 + 0.497\ pH + 0.0065\ C_0 + 0.0093\ T + 0.01211\ t - 0.03690\ pH^2 - 0.000019\ C_0^2 - 0.000322\ T^2 - 0.000030\ t^2 - 0.00076\ pH \cdot C_0 + 0.00242\ pH \cdot T - 0.000594\ pH \cdot t$
	BG	$E\ (\%) = -2.796 + 0.620\ pH + 0.0062\ C_0 + 0.0093\ T + 0.01211\ t - 0.03690\ pH^2 - 0.000019\ C_0^2 - 0.000322\ T^2 - 0.000030\ t^2 - 0.00076\ pH \cdot C_0 + 0.00242\ pH \cdot T - 0.000594\ pH \cdot t$
GP-Ch	MB	$E\ (\%) = -1.700 + 0.514\ pH + 0.0031\ C_0 + 0.0090\ T + 0.01204\ t - 0.03690\ pH^2 - 0.000019\ C_0^2 - 0.000322\ T^2 - 0.000030\ t^2 - 0.00076\ pH \cdot C_0 + 0.00242\ pH \cdot T - 0.000594\ pH \cdot t$
	CV	$E\ (\%) = -1.716 + 0.508\ pH + 0.0065\ C_0 + 0.0090\ T + 0.01204\ t - 0.03690\ pH^2 - 0.000019\ C_0^2 - 0.000322\ T^2 - 0.000030\ t^2 - 0.00076\ pH \cdot C_0 + 0.00242\ pH \cdot T - 0.000594\ pH \cdot t$
	BG	$E\ (\%) = -2.760 + 0.631\ pH + 0.0062\ C_0 + 0.0090\ T + 0.01204\ t - 0.03690\ pH^2 - 0.000019\ C_0^2 - 0.000322\ T^2 - 0.000030\ t^2 - 0.00076\ pH \cdot C_0 + 0.00242\ pH \cdot T - 0.000594\ pH \cdot t$
GP-PVA	MB	$E\ (\%) = -1.471 + 0.494\ pH + 0.0035\ C_0 + 0.0078\ T + 0.01172\ t - 0.03690\ pH^2 - 0.000019\ C_0^2 - 0.000322\ T^2 - 0.000030\ t^2 - 0.00076\ pH \cdot C_0 + 0.00242\ pH \cdot T - 0.000594\ pH \cdot t$
	CV	$E\ (\%) = -1.481 + 0.488\ pH + 0.0068\ C_0 + 0.0078\ T + 0.01172\ t - 0.03690\ pH^2 - 0.000019\ C_0^2 - 0.000322\ T^2 - 0.000030\ t^2 - 0.00076\ pH \cdot C_0 + 0.00242\ pH \cdot T - 0.000594\ pH \cdot t$
	BG	$E\ (\%) = -2.624 + 0.611\ pH + 0.0065\ C_0 + 0.0078\ T + 0.01172\ t - 0.03690\ pH^2 - 0.000019\ C_0^2 - 0.000322\ T^2 - 0.000030\ t^2 - 0.00076\ pH \cdot C_0 + 0.00242\ pH \cdot T - 0.000594\ pH \cdot t$

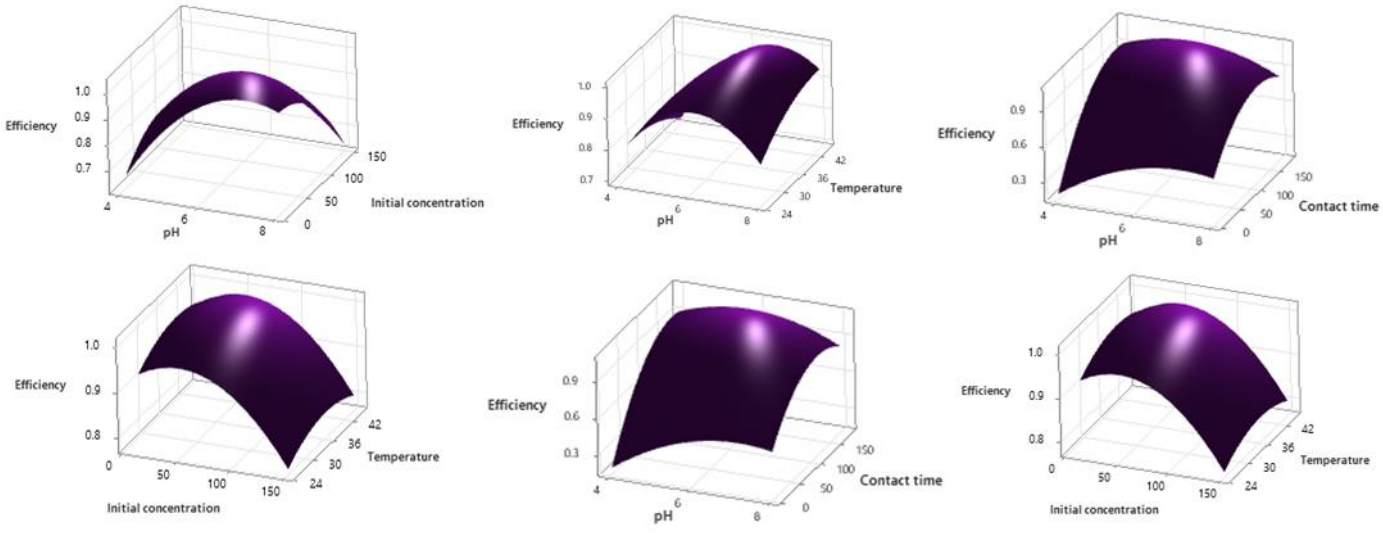


Figure S3. Effect of process parameters on the adsorption efficiency of GP-Ch for CV dye.

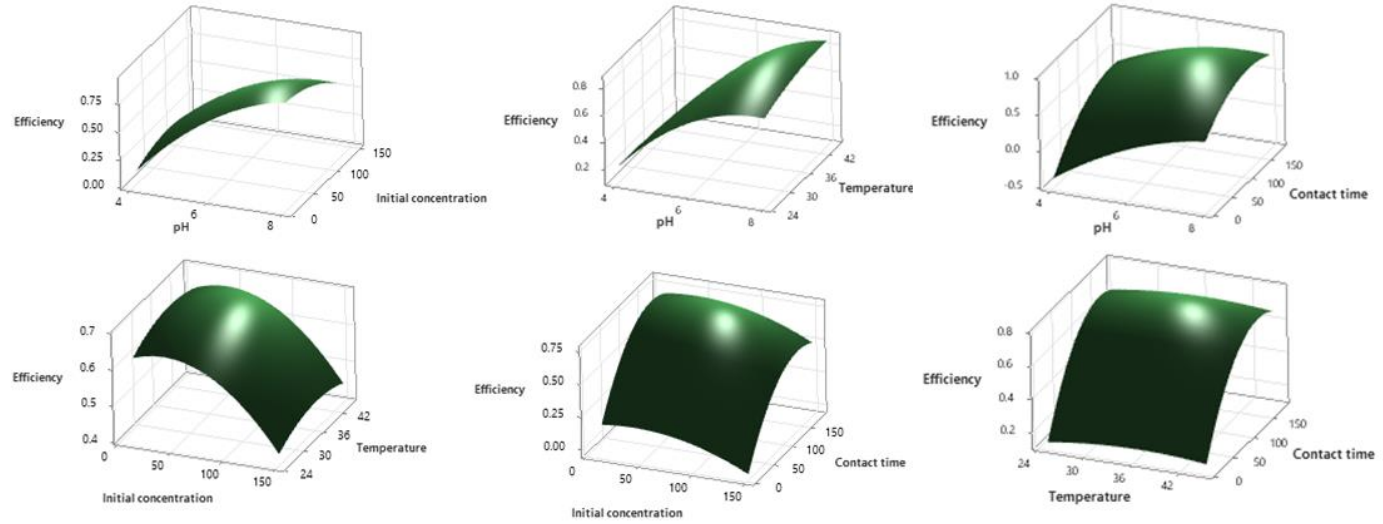


Figure S4. Effect of process parameters on the adsorption efficiency of GP-Ch for BG dye.

Table S5. Intraparticle diffusion parameters and correlation coefficients for dye adsorption onto geopolymers.

Geopolymer	Dye	R_1^2	k_1, mg/(g·min$^{1/2}$)	C_1 mg/g	R_2^2	k_2, mg/(g·min$^{1/2}$)	C_2 mg/g	R_3^2	k_3, mg/(g·min$^{1/2}$)	C_3 mg/g
GP	MB	0.920	1.250	1.805	0.925	0.444	6.636	1.00	0.0148	11.21
	CV	0.996	0.737	4.713	0.815	0.225	7.680	1.00	0.1167	8.77
	BG	0.986	1.417	-2.573	1.00	0.994	-0.368	0.958	0.1712	6.04
GP-Ch	MB	0.981	0.990	2.202	1.00	0.390	6.997	1.00	0.0738	10.46
	CV	0.993	1.019	3.421	0.775	0.247	7.755	1.00	0.0214	10.11
	BG	0.985	1.163	0.547	1.00	0.486	4.163	0.997	0.2078	6.30
GP-PVA	MB	0.996	1.145	3.797	1.00	0.496	7.310	0.936	0.0516	10.76
	CV	1.000	0.731	5.196	0.818	0.187	8.068	1.00	0.0997	9.13
	BG	0.962	1.547	-2.286	1.00	0.644	2.457	0.886	0.1667	6.21

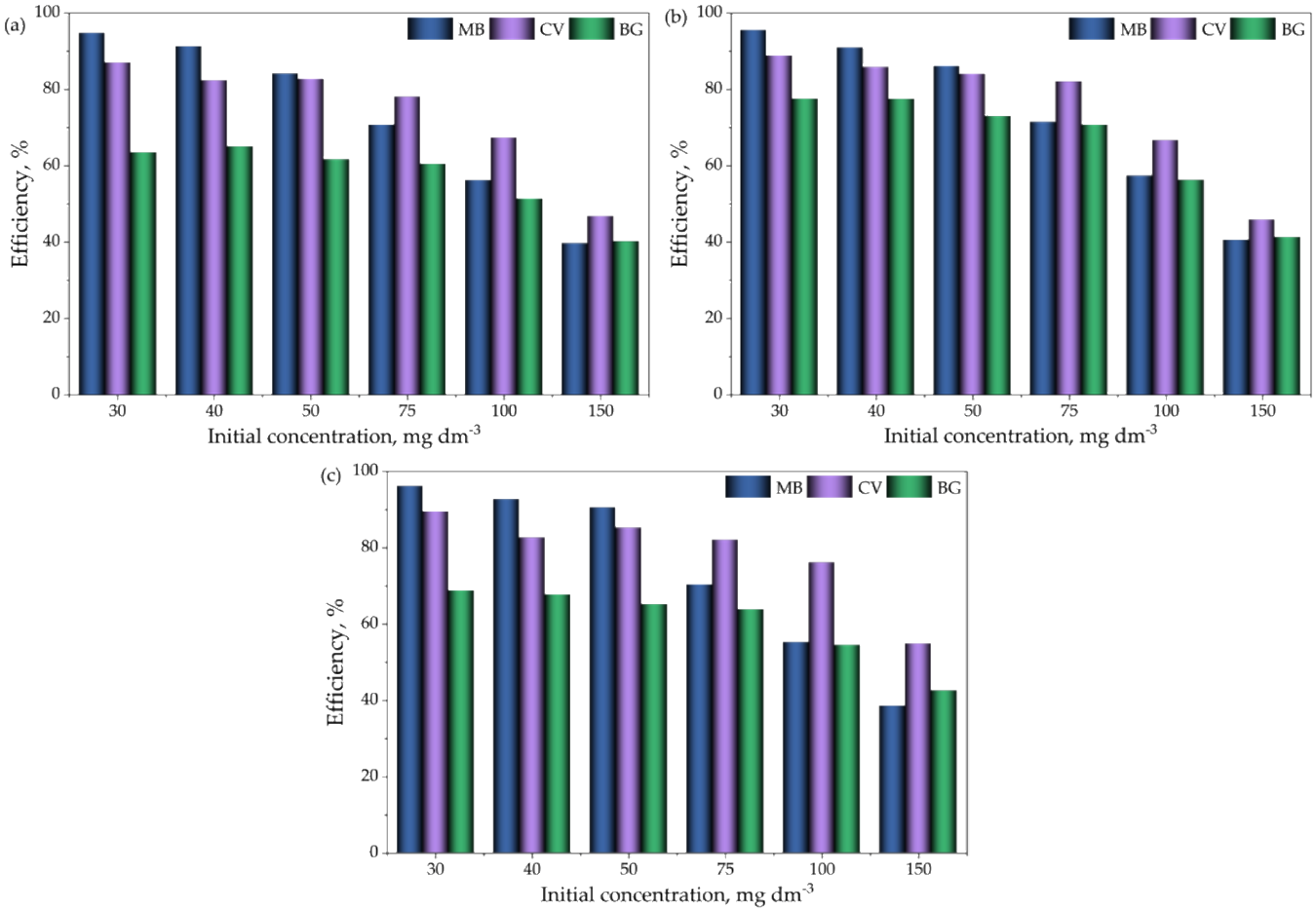


Figure S5. Influence of initial dye concentration on the adsorption efficiency.

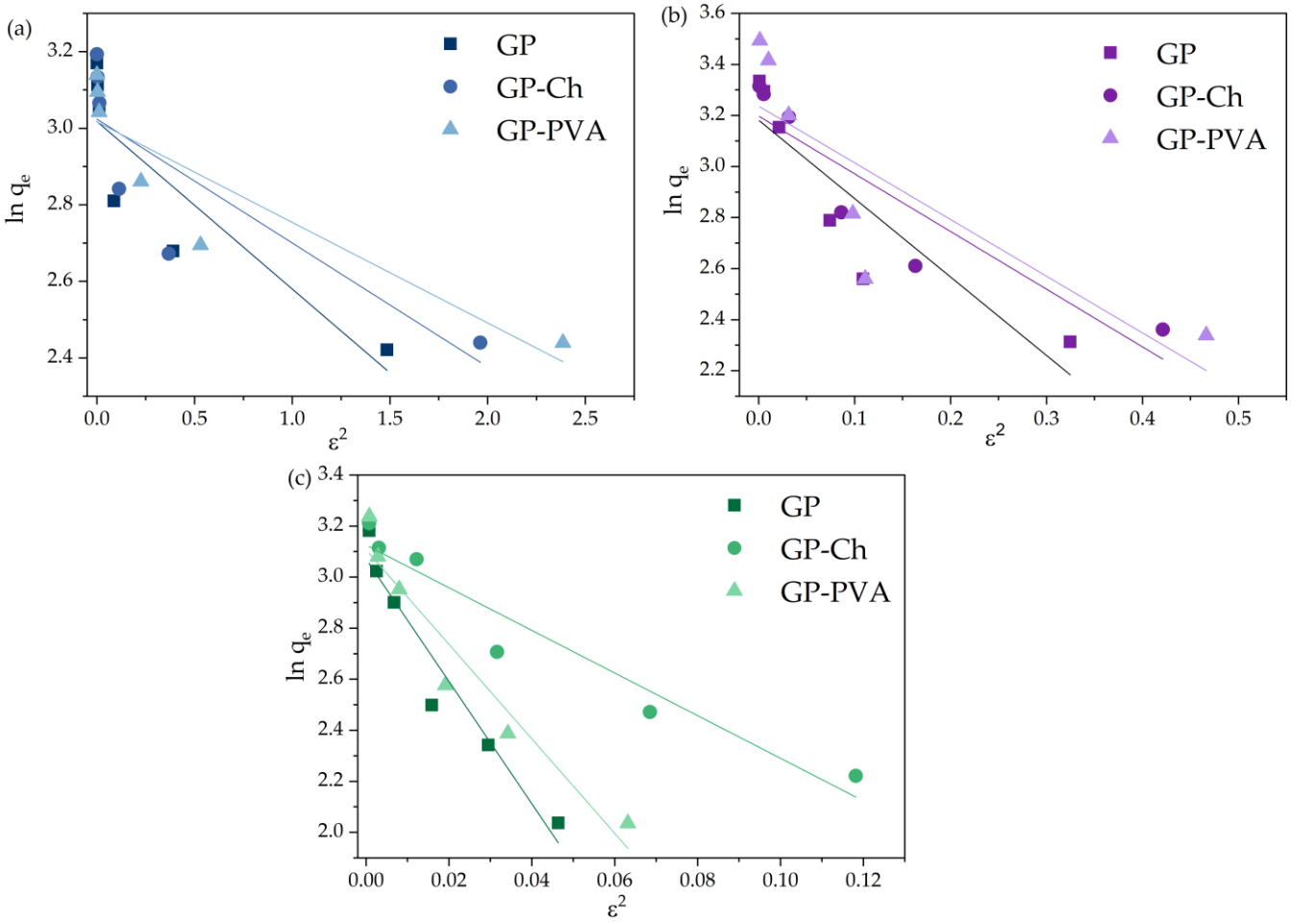


Figure S6. The influence of initial concentration of a) MB, b) CV, and c) BG solution on geopolymers' adsorption capacities and fitting of experimental data with Dubinin–Radushkevich isotherm model.

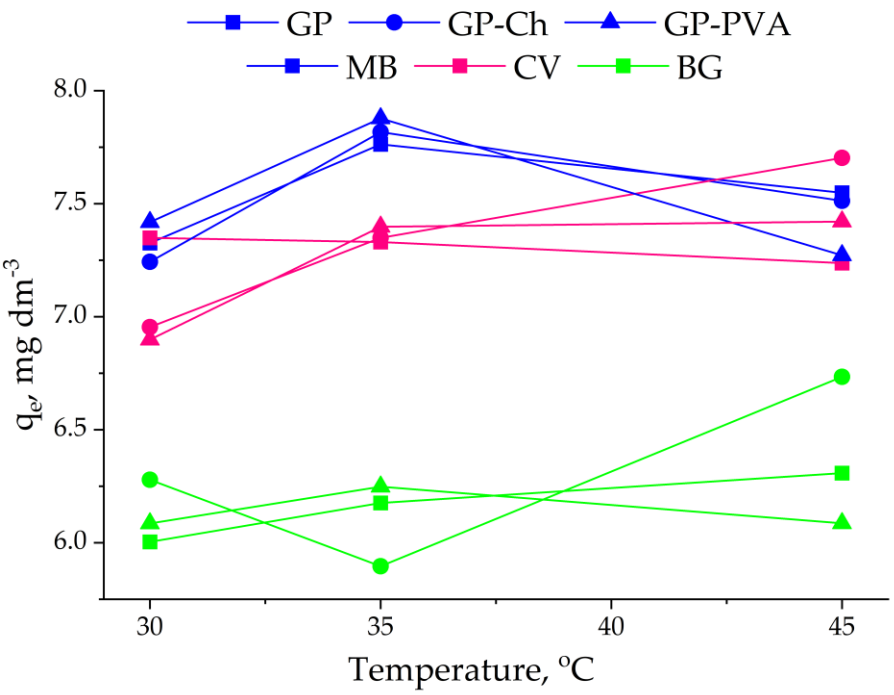


Figure S7. Impact of Temperature on Dye Adsorption onto Geopolymer.