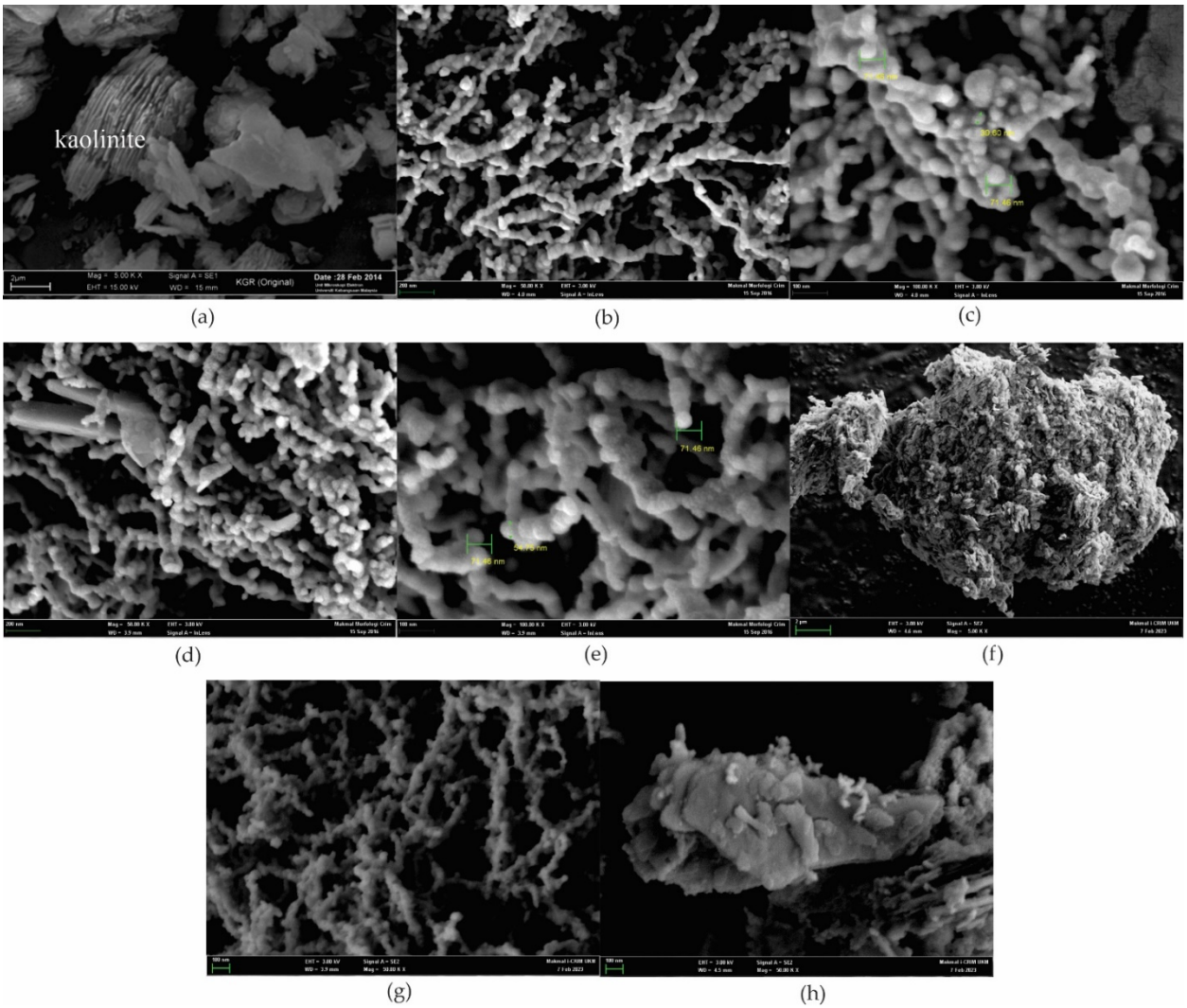


Figure S1. The morphologies before treatment; SEM for (a) Gr; 5000× and FESEM for (b) nZVI; Mag: 50,000×, (c) nZVI; Mag: 100,000×, (d) Gr-nZVI; Mag: 50,000×, (e) Gr-nZVI; Mag: 100,000× while the morphologies after treatment; FESEM for (f) Gr; Mag: 5000× (g) nZVI; Mag: 50,000×, (h) Gr-nZVI; Mag: 50,000×.

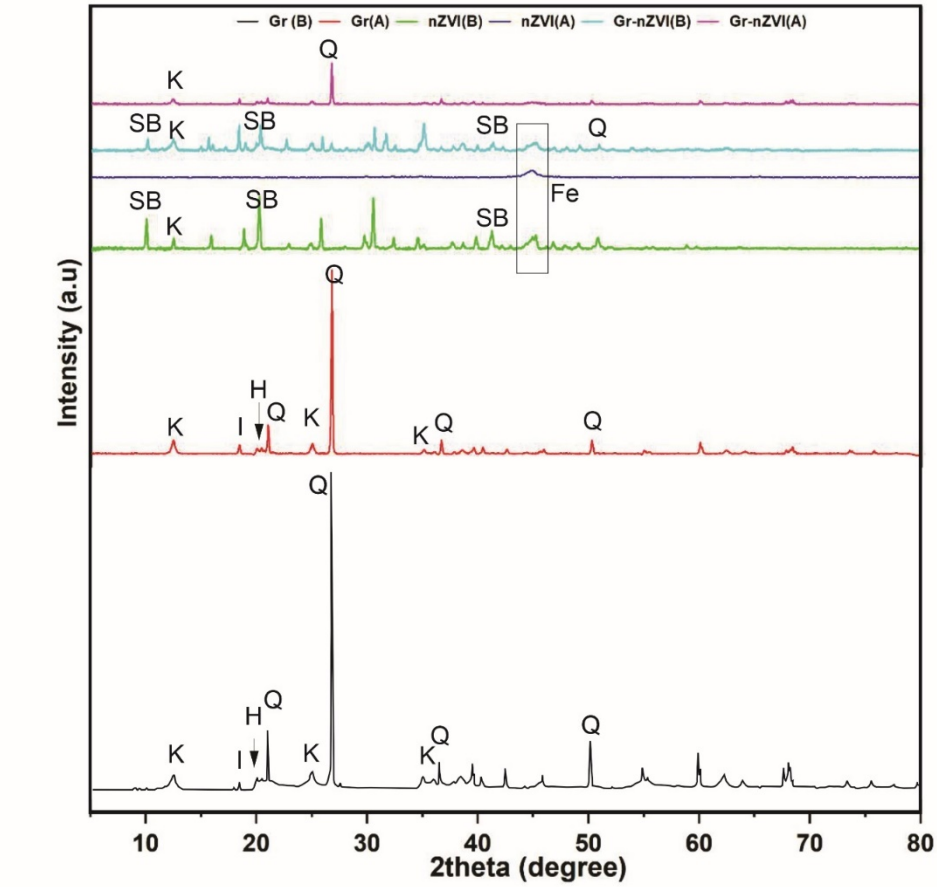


Figure S2. The XRD results for all samples. [SB=sodium borate, $Na_2B_4O_7$, Q=Quartz, K=kaolinite, H=halloysite, Fe= iron Fe^0]

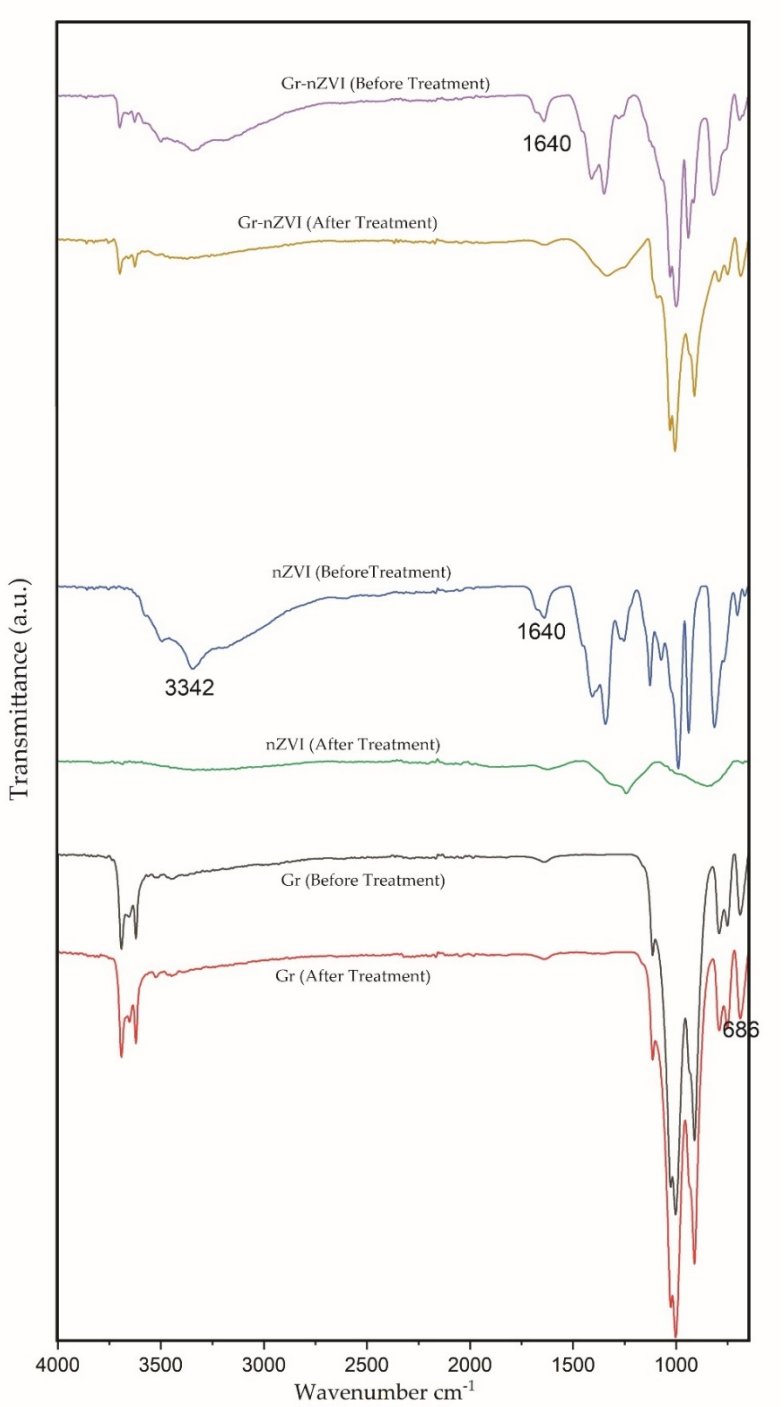


Figure S3. The FTIR spectrum of nZVI, Gr and Gr-nZVI before and after the treatment

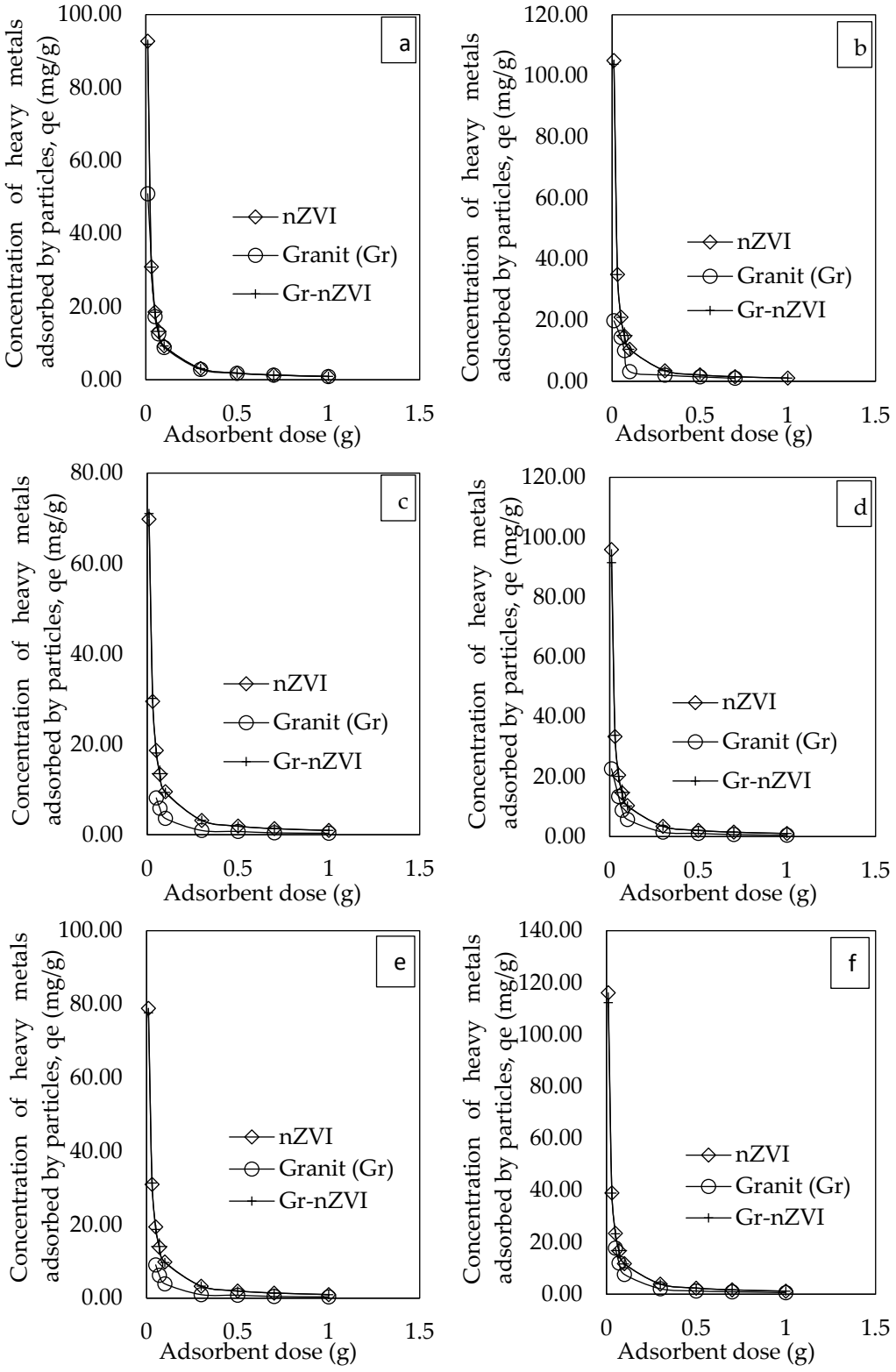
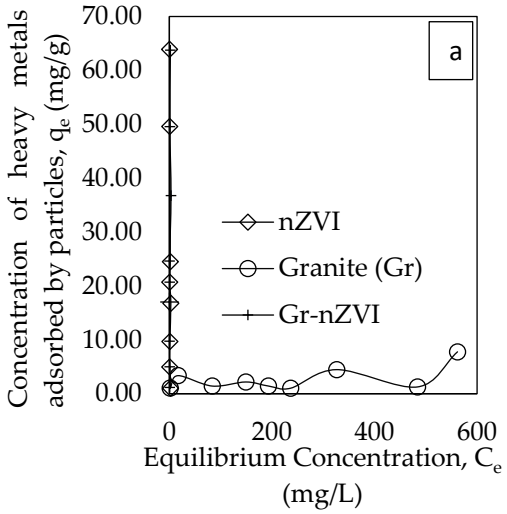
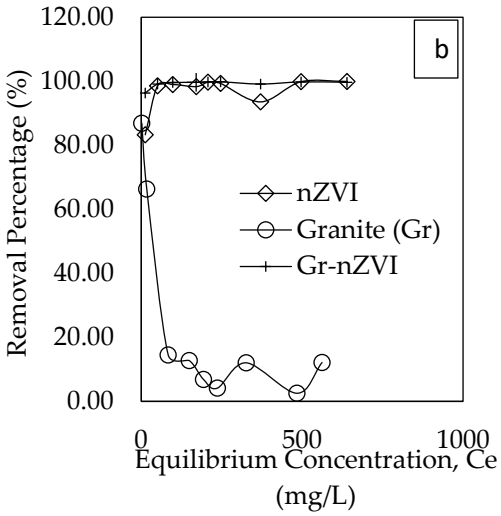
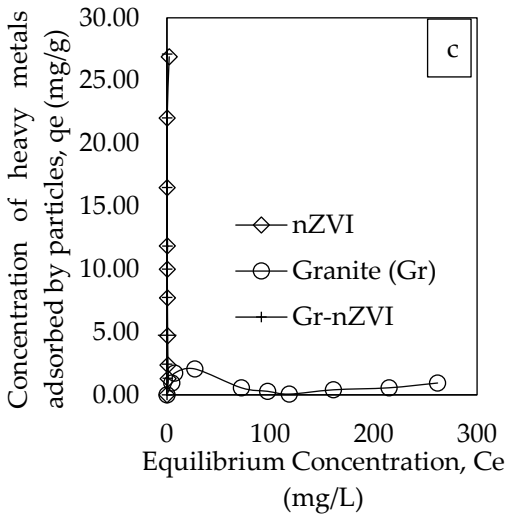
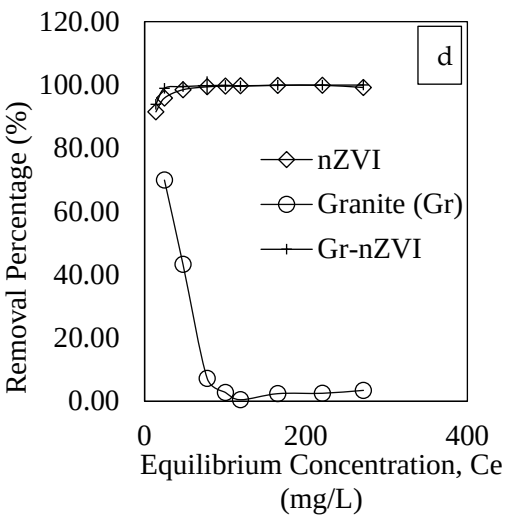


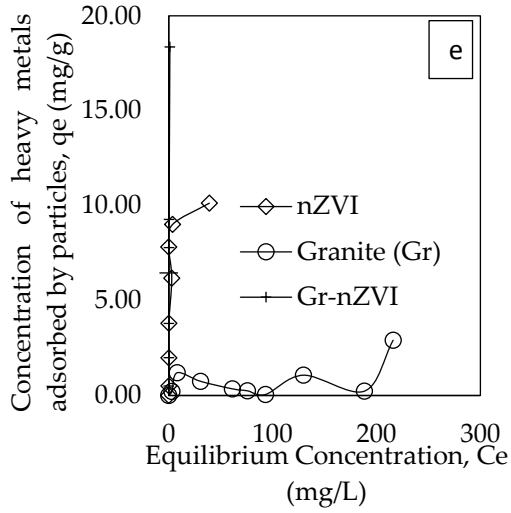
Figure S4. Effect of adsorbent dosage on heavy metals (a) Pb, (b) Cu, (c) Co, (d) Cd, (e) Ni, (f) Zn adsorption capacity (Co= 50 mg/L, V= 30 ml, pH=6, shaking time= 3h, Temperature= 25^{o}C).

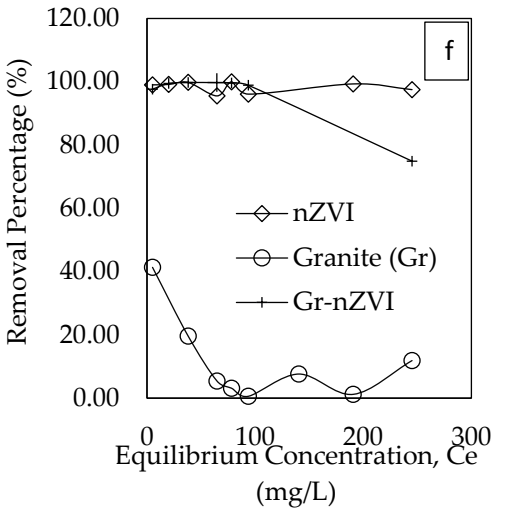

a
Concentration of heavy metals adsorbed by particles, q_e (mg/g)
70.00
60.00
50.00
40.00
30.00
20.00
10.00
0.00
0
200
400
600
Equilibrium Concentration, C_e (mg/L)
nZVI
Granite (Gr)
Gr-nZVI


b
Removal Percentage (%)
120.00
100.00
80.00
60.00
40.00
20.00
0.00
0
500
1000
Equilibrium Concentration, Ce (mg/L)
nZVI
Granite (Gr)
Gr-nZVI


c
Concentration of heavy metals adsorbed by particles, qe (mg/g)
30.00
25.00
20.00
15.00
10.00
5.00
0.00
0
100
200
300
Equilibrium Concentration, Ce (mg/L)
nZVI
Granite (Gr)
Gr-nZVI


d
Removal Percentage (%)
120.00
100.00
80.00
60.00
40.00
20.00
0.00
0
200
400
Equilibrium Concentration, Ce (mg/L)
nZVI
Granite (Gr)
Gr-nZVI


e
Concentration of heavy metals adsorbed by particles, qe (mg/g)
20.00
15.00
10.00
5.00
0.00
0
100
200
300
Equilibrium Concentration, Ce (mg/L)
nZVI
Granite (Gr)
Gr-nZVI


f
Removal Percentage (%)
120.00
100.00
80.00
60.00
40.00
20.00
0.00
0
100
200
300
Equilibrium Concentration, Ce (mg/L)
nZVI
Granite (Gr)
Gr-nZVI

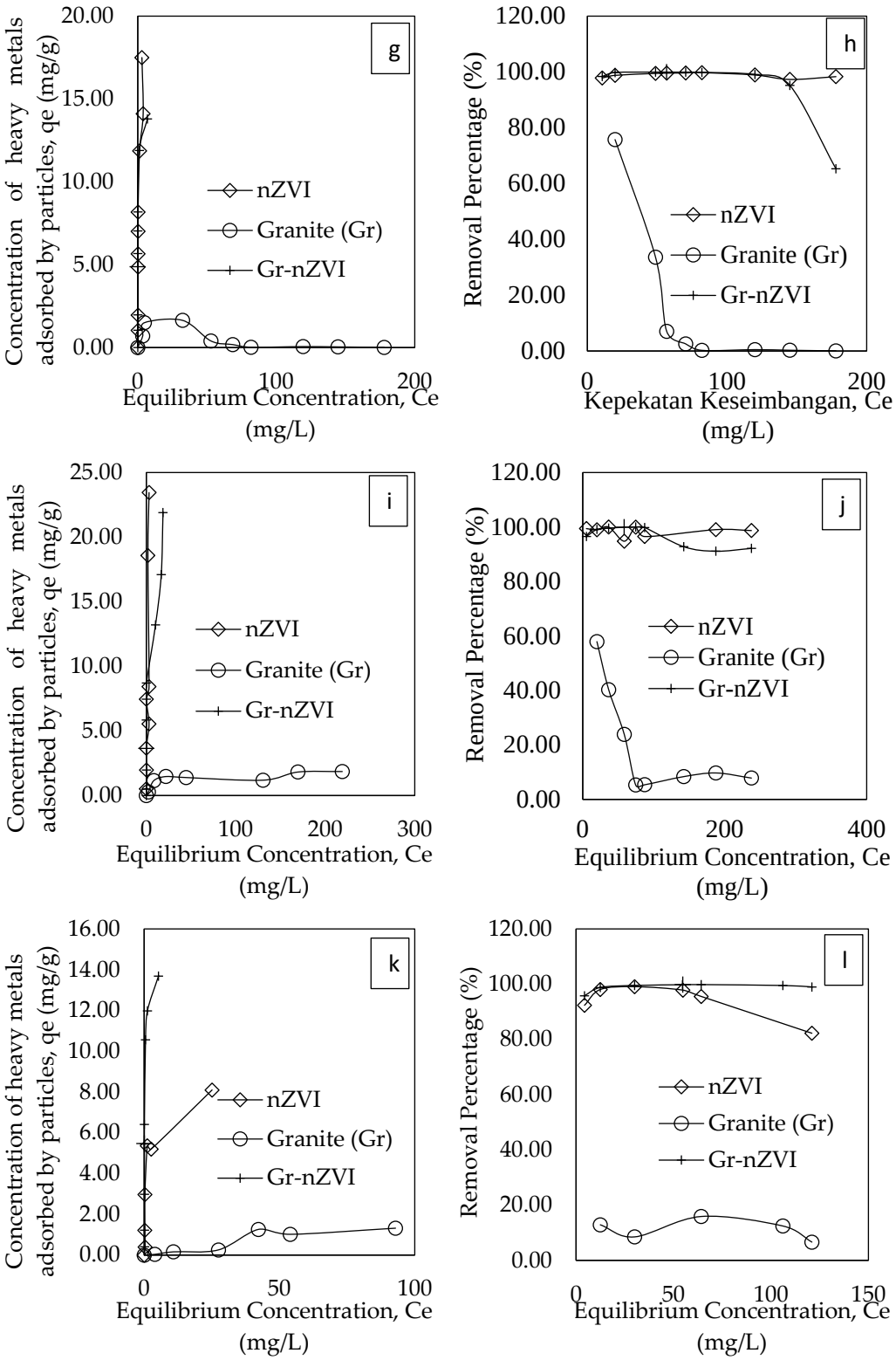


Figure S5. Effect of initial concentration and removal percentage on heavy metals Pb (a and b), Cu (c and d), Co (e and f), Cd (g and h), Ni (i and j) and Zn (k and l) adsorption capacity (M= 0.5 g, V= 50 ml, pH=6, shaking time= 3h, Temperature= 25^{o}C).

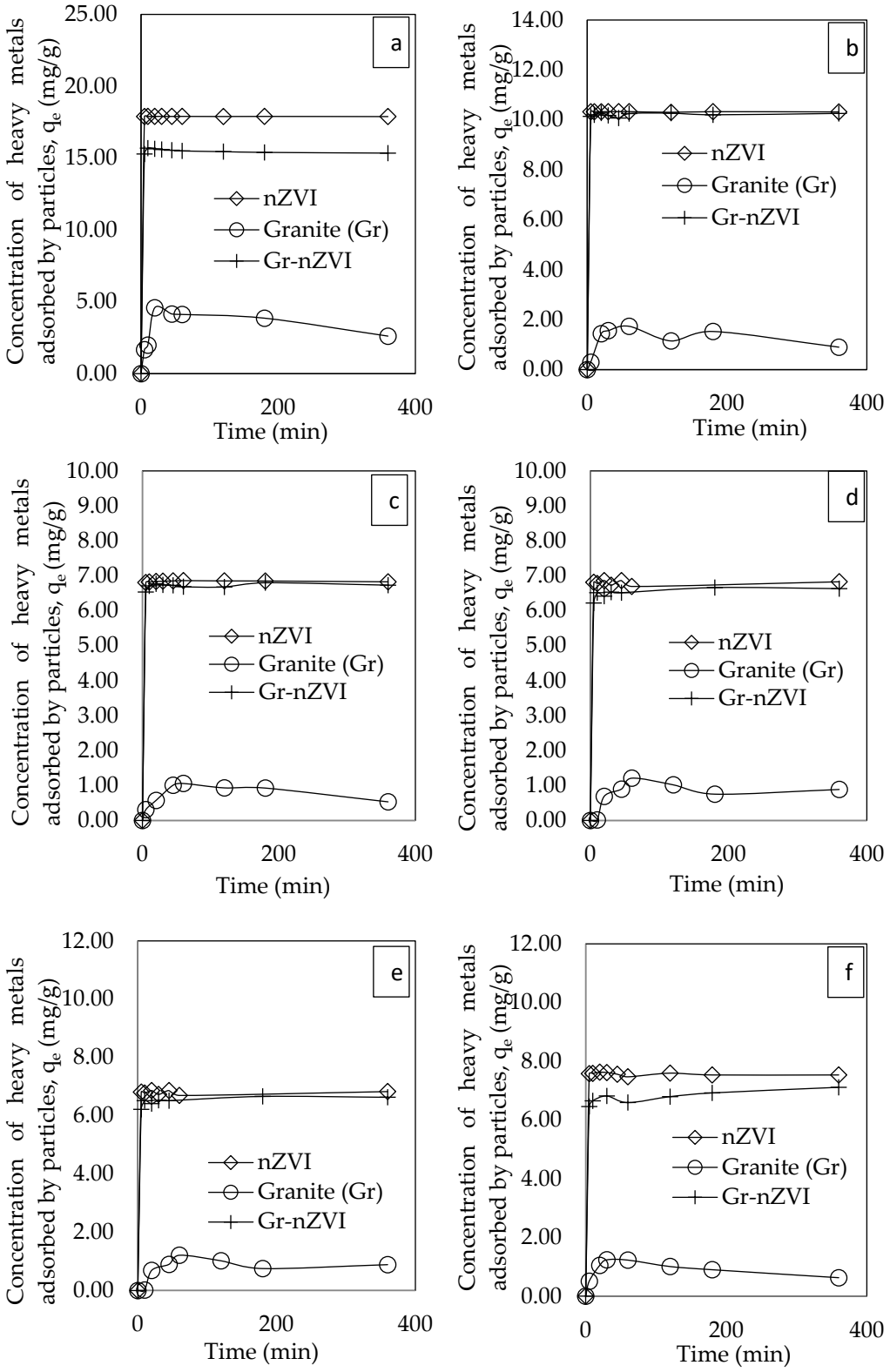


Figure S6. Effect of contact time on heavy metals (a) Pb, (b) Cu, (c) Co, (d) Cd, (e) Ni, (f) Zn adsorption capacity (Co= 50 mg/L, M= 0.5 g, V= 50 ml, pH=6, Temperature= 25°C)

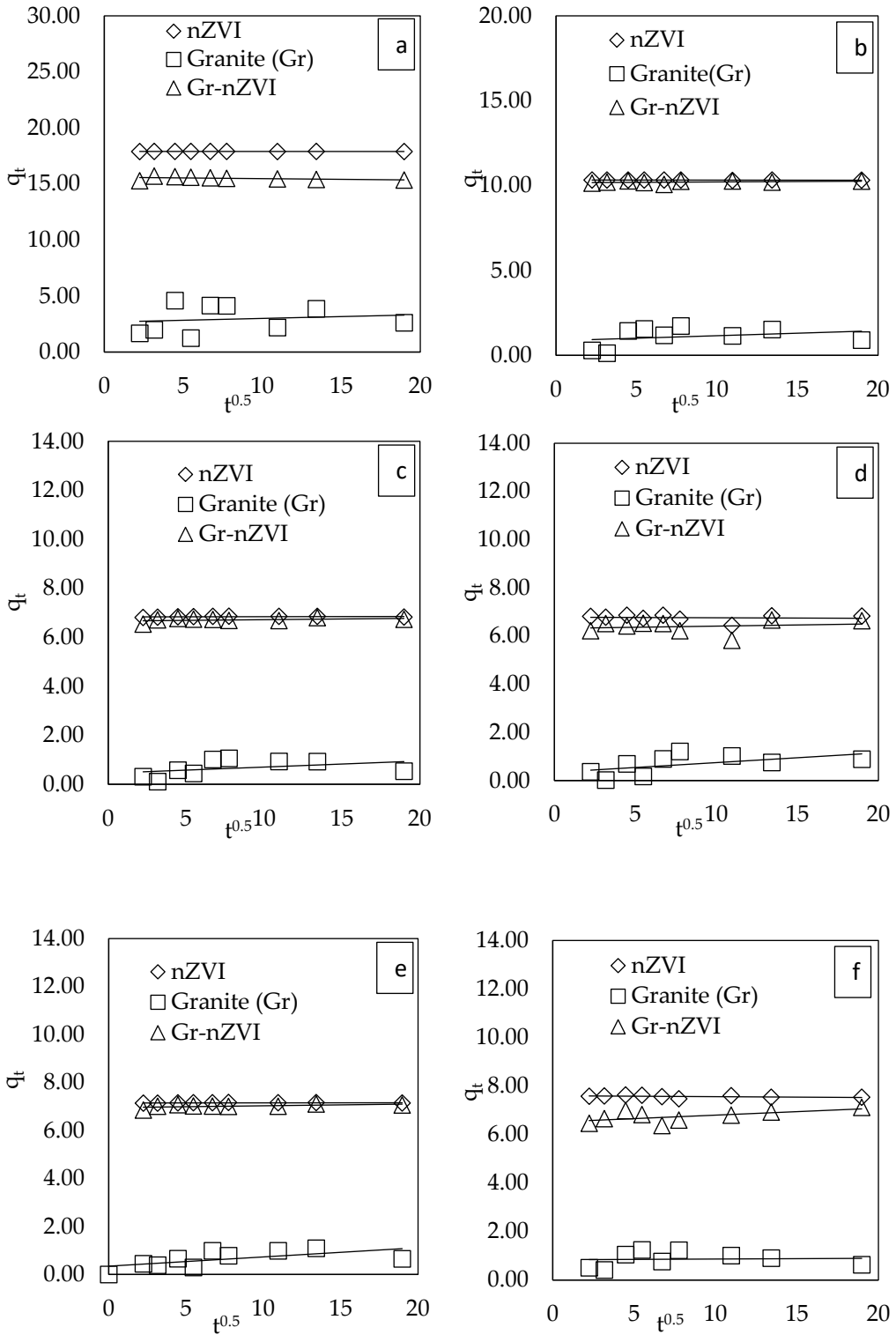


Figure S7. Plot of intraparticle diffusion modelling of heavy metals (a) Pb, (b) Cu, (c) Co, (d) Cd, (e) Ni, (f) Zn onto adsorbents (nZVI, Gr and Gr-nZVI)

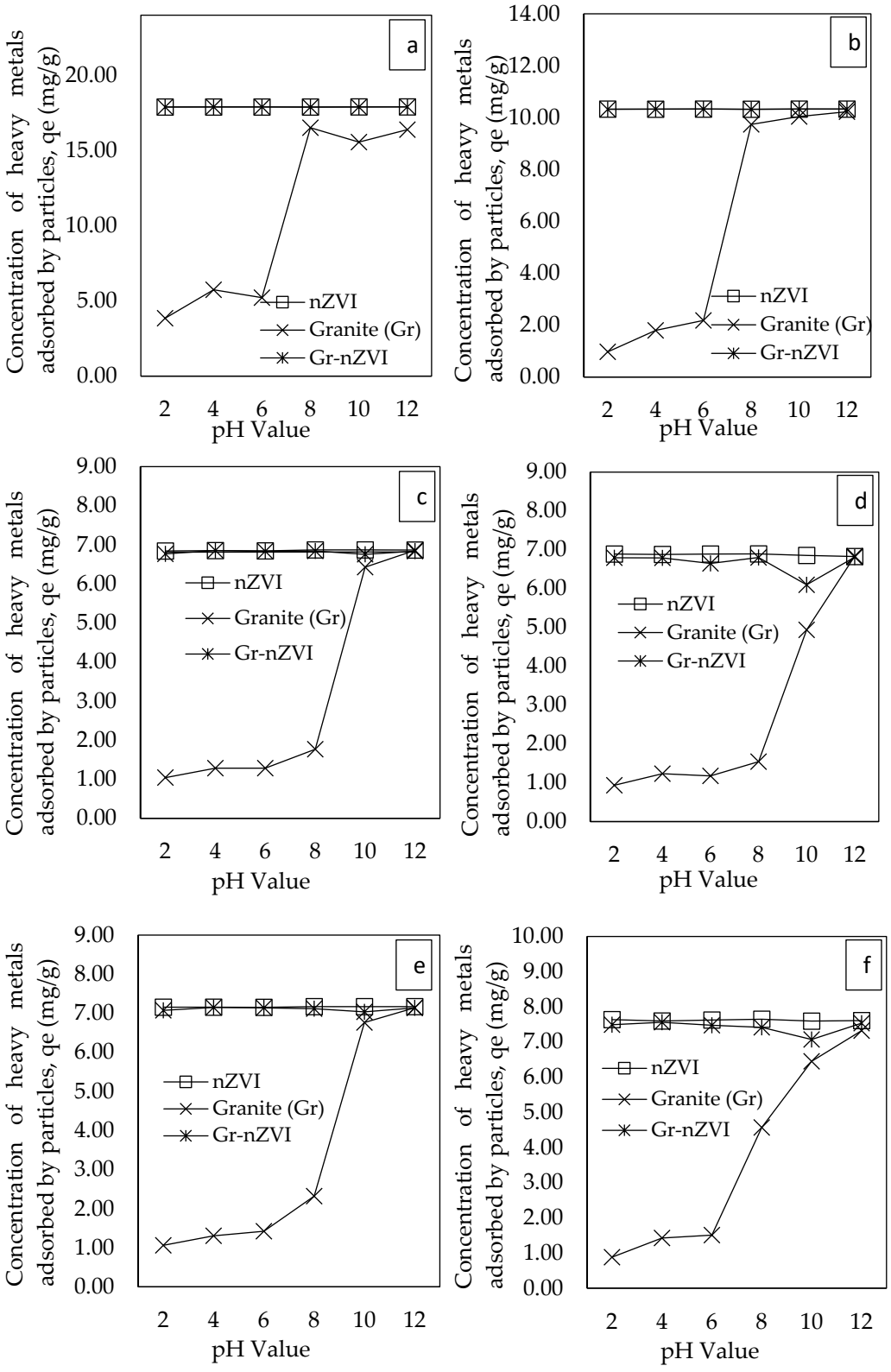


Figure S8. Effect of initial pH on heavy metals (a) Pb, (b) Cu, (c) Co, (d) Cd, (e) Ni, (f) Zn adsorption capacity (Co= 50 mg/L, M= 0.5 g, V= 50 ml, shaking time= 3h, Temperature= 25°C)

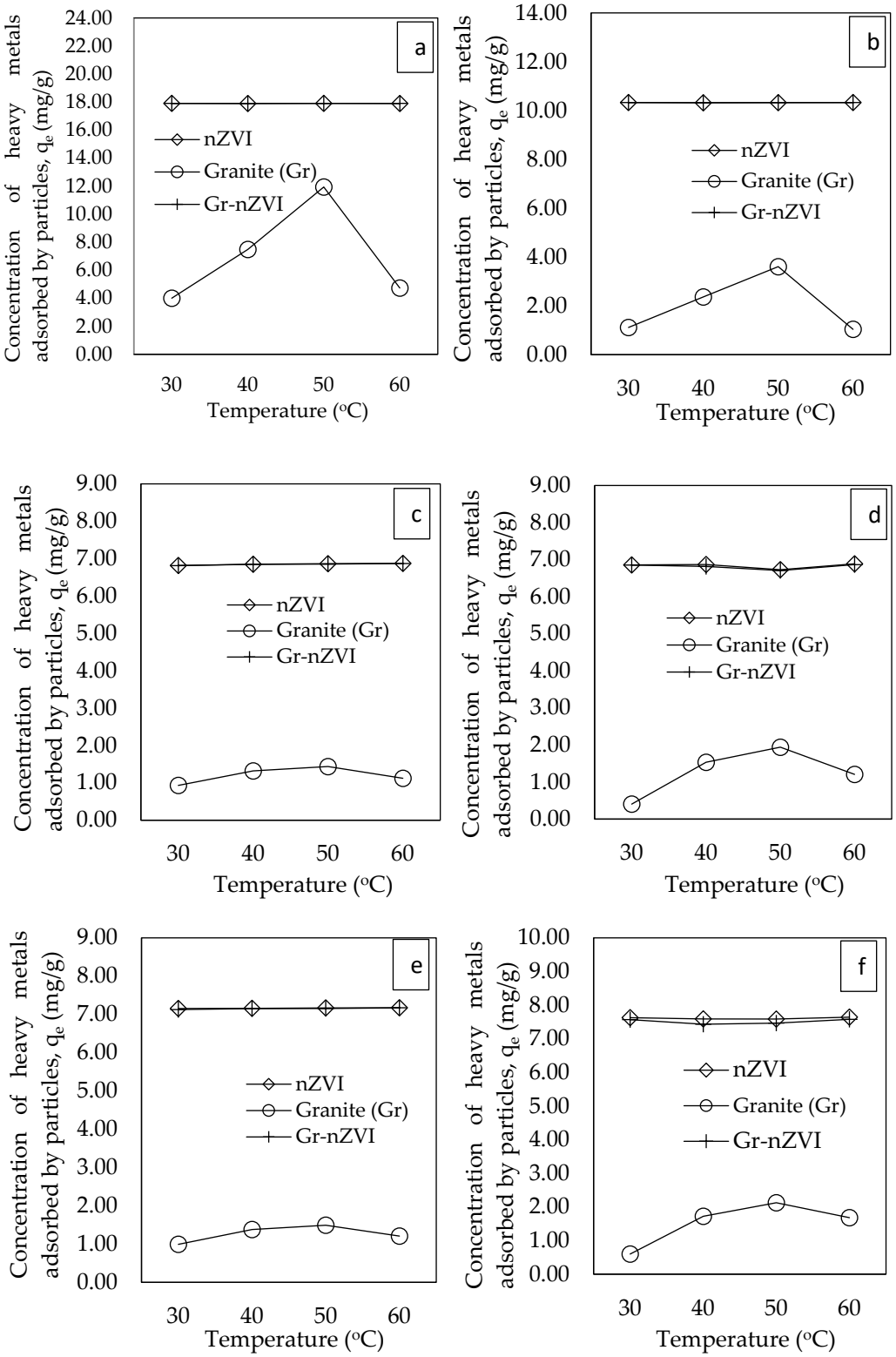


Figure S9. Effect of temperature on heavy metals (a) Pb, (b) Cu, (c) Co, (d) Cd, (e) Ni, (f) Zn adsorption capacity (Co= 50 mg/L, M= 0.5 g, V= 50 ml, pH=6, shaking time= 3h)

Table S1. Linear equation of adsorption isotherm and kinetic model

Model	Equation	Significance	Plots
Langmuir	$C_e/q_e = 1/K_L . A_m + C_e/A_m$ Where, K_L is the Langmuir binding constant (L/mg), A_m is the saturated adsorption amount of metal ions (mg/g) and C_e is the equilibrium concentration of metal ions (mg/L)	Signifies monolayer adsorption of adsorbate molecules onto adsorbent	C_e/q_e vs C_e
Freundlich	$\log q_e = \log K_F + (1/n) \log C_e$ Where, K_F is the Freundlich sorption coefficient, 1/n is Freundlich sorption exponent and C_e is the metal concentration in final contact concentration (mg/L).	Surface heterogeneity and exponential distribution of active sites and their energies	Loq q_e vs Log C_e
Pseudo first order	$\ln (q_e - q_t) = \ln(q_e) - \frac{k_1}{2.303} t$ where q_e and q_t represents the adsorption capacity of adsorbent at equilibrium and time t in mg/g, respectively, k_1 is the pseudo first-order rate constant, and t is the contact time (min)	Adsorption at the solid-liquid interface	$\ln (q_e - q_t)$ vs t
Pseudo second order	$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}$ where q_e and q_t represents the adsorption capacity of adsorbent at equilibrium and time t in mg/g, respectively, k_2 is the pseudo second-order rate constant, and t is the contact time (min)	Rate-limiting step in adsorption is controlled by chemical interactions	t/q_t versus t
Interparticle diffusion	$ln \frac{C_t}{C_o} = -k_f \frac{A}{V} t$ where C_o is the initial metal concentration, C_t is the concentration at time t, A/V is the external sorption area to the total solution volume, t is the sorption time and k_f is the external diffusion coefficient ($m^2g^{-1}minL^{-1}$)	The ionic adsorption of the contaminant is adsorbed only on the outer surface of the adsorbent.	$\ln (C_t - C_o)$ vs t
Intraparticle diffusion	$q_t = k_d t^{1/2}$ where q_t is the adsorption capacity at time t, $t^{0.5}$ is the half-life time in second, and k_d (mg/g $min^{0.5}$) is the rate constant of intraparticle diffusion	Rate-limiting step includes the transference of solute from solution into adsorbent pores	q_t versus $t^{0.5}$

Table S2. The BET results for all samples

Sample	BET Surface Area (m^2/g)	Pore Volume (cm^3/g)	Pore Size (Å)
nZVI	1.5724	0.0042	107.0115
Gr	14.1724	0.0484	136.6348
Gr-nZVI	6.7619	0.0298	176.0732

Table S3. The CEC for all samples

Sample	Cation Exchange Capacity (meq/100g)
nZVI	4.03±5.57
Gr	2.70±9.03
Gr-nZVI	1.68±7.24

Table S4. The XRF results for all adsorbent materials

Elements	nZVI	Gr	Gr-nZVI
SiO_2	0.26%	42.86%	20.62%
Al_2O_3	-	34.12%	16.65%
Fe_2O_3	54.99%	4.08%	30.81%
Na_2O	34.19%	-	19.09%
Cl	0.31%	-	0.12%
Cr_2O_3	0.02%	-	0.02%
K_2O	-	1.53%	0.95%
TiO_2	-	0.27%	0.18%
MgO	-	0.13%	0.08%
CaO	-	-	0.05%
P_2O_5	-	0.05%	0.11%
SO_3	-	0.04%	-
ZrO_2	-	0.03%	0.06%
Rb_2O	-	0.02%	0.02%
MnO	0.01%	0.01%	-
Ga_2O_3	-	0.01%	-
V_2O_5	-	0.01%	-
ZnO	-	0.01%	-
LOI	10.12%	16.56%	11.58%
Total	99.89%	99.69%	100.34%

Table S5. Model parameters for concentration

Sample	Heavy Metals Element	Linear Equation		Langmuir Equation			Freundlich Equation		
		K_d (L/g)	R^2	K_L (L/g)	A_m (mg/g)	R^2	K_F(L/g)	1/n	R^2
nZVI	Pb	7.55	0.56	820.00	36.90	0.95	1.18	0.14	0.02
	Cu	5.47	0.85	266.87	14.14	0.11	1.62	0.63	0.97
	Co	2.21	0.72	1600.92	10.25	0.99	31.27	1.62	0.24
	Cd	16.95	0.79	679.54	16.92	0.96	8.37	0.92	0.50
	Ni	8.29	0.92	305.07	11.07	0.99	22.87	1.45	0.40
	Zn	2.34	0.87	71.67	9.22	0.98	16.38	2.34	0.78
Granite (Gr)	Pb	0.01	0.68	0.19	1.10	0.68	195.16	0.15	0.02
	Cu	0.22	1.00	0.03	0.56	0.77	19.61	0.75	0.52
	Co	0.01	0.56	0.00	0.22	0.25	36.37	0.50	0.26
	Cd	0.27	0.96	0.97	1.82	0.98	14.25	0.60	0.62
	Ni	0.08	0.91	0.23	1.81	0.92	47.29	1.05	0.25
	Zn	0.02	0.89	0.04	1.89	0.24	63.33	0.83	0.84
Gr-nZVI	Pb	33.99	0.91	3667.44	54.64	0.66	3.96	0.37	0.36
	Cu	14.53	0.40	5372.88	9.67	0.49	1.48	0.54	0.63
	Co	19.79	0.89	62.47	8.06	0.96	1149.74	3.94	0.37
	Cd	121.84	0.90	956.44	11.67	1.00	215575.80	5.12	0.91
	Ni	1.14	0.84	464.61	16.45	0.98	242.21	2.43	0.51
	Zn	21.71	0.75	872.33	14.31	0.99	7271.10	3.75	0.90

Table S6. Kinetic parameters on heavy metals adsorption onto adsorbents

Sample	Heavy Metals	Type of Solution	qe experimental (mg/g)	Pseudo first Order			Pseudo Second Order			External Diffusion Models (Interparticle diffusion)		Internal Diffusion Models (Intraparticle diffusion)		
				K_1 (1/min)	q_e calculation (mg/g)	R^2	K_2 (g/ mg min)	q_e calculation (mg/g)	R^2	K_I ($mg^{-1}minL^{-1}$)	R^2	K_d ($mg\ g^{-1}\ min^{-0.5}$)	C	R^2
nZVI	Pb	Mix	20.69	8.00E-04	2.37	0.07	2.60	17.89	1.00	0.5	0.82	4.15	3.18	0.76
	Cu	Mix	11.85	3.00E-04	1.42	0.07	6.71	10.32	1.00	1.21	0.89	3.52	0.56	0.92
	Co	Mix	7.82	2.00E-05	0.98	0.00	1.73	6.85	1.00	0.80	0.91	2.32	0.37	0.92
	Cd	Mix	7.02	1.20E-03	0.30	0.05	0.21	6.82	1.00	0.44	0.84	2.31	0.37	0.96
	Ni	Mix	7.45	9.00E-04	0.37	0.07	10.84	7.16	1.00	0.87	0.90	2.43	0.39	0.92
	Zn	Mix	8.10	2.00E-04	0.58	0.01	2.72	7.58	1.00	0.74	0.91	2.58	0.41	0.92
Gr	Pb	Mix	2.18	1.60E-02	0.50	0.07	0.73	2.74	0.95	2.40E-03	0.99	0.65	0.05	0.98
	Cu	Mix	2.06	2.00E-04	0.85	0.00	0.72	0.40	0.89	9.00E-04	0.93	0.14	0.20	0.51
	Co	Mix	1.07	2.90E-03	0.27	0.01	0.03	1.11	0.81	5.00E-04	0.95	0.10	0.02	0.76
	Cd	Mix	1.64	1.30E-03	0.3	0.13	0.33	0.49	0.86	5.00E-04	0.88	0.05	0.24	0.41
	Ni	Mix	1.19	2.50E-03	0.54	0.14	0.03	1.19	0.83	4.00E-04	0.83	0.14	0.03	0.92
	Zn	Mix	1.32	2.30E-03	0.31	0.02	1.25	0.95	0.98	1.00E-03	0.97	0.15	0.11	0.74
Gr-nZVI	Pb	Mix	17.02	7.00E-04	1.37	0.25	0.18	15.34	1.00	0.08	0.92	5.31	0.77	0.93
	Cu	Mix	11.84	2.00E-04	1.52	0.03	0.55	10.26	1.00	0.17	0.93	3.47	0.54	0.92
	Co	Mix	7.79	3.00E-05	1.06	0.00	1.91	6.74	1.00	0.14	0.95	2.27	0.33	0.93

Cd	Mix	7.04	1.60E-03	0.30	0.21	0.08	6.61	1.00	0.11	0.96	2.19	0.30	0.94
Ni	Mix	7.45	1.20E-03	0.52	0.21	0.72	7.07	1.00	0.14	0.95	2.37	0.35	0.93
Zn	Mix	10.56	6.00E-04	3.16	0.03	0.06	7.10	1.00	0.08	0.93	2.25	0.32	0.93