Supporting Information

for

Critical Role of Coexistence Order and Interfacial Forces in the Aggregation of Polystyrene Nanoplastics Induced by Nano-SiO_2 and Metal Cations

Pengju Ren, Yihan Chi, Lijuan Wang, Yuanyuan Tang*

Guangdong Provincial Key Laboratory of Soil and Groundwater Pollution Control, School of Environmental Science and Engineering, Southern University of Science and Technology, Shenzhen 518055, P. R. China

*Corresponding author: Yuanyuan Tang; E-mail address: tangyy@sustech.edu.cn

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Text S1. Method of data processing

Before starting the aggregation experiments, we conducted preliminary experiments on pure SiO_2 in a metal cation system. As shown in Fig. S2, we observed rapid aggregation of SiO_2, which usually occurs before the initial DLS sampling. This aggregation exceeded the particle size of PS NPs within the same timeframe under the same experiment conditions (Fig. S2). To differentiate between SiO_2 and PS NPs aggregates in the DLS-measured data, we identified anomalously large data points as potential SiO_2 aggregates. These points were replaced with interpolated values based on the average of neighboring points.

Table S1. Experimental program setting

Experiment system	System name	Ion concentration (mmol/L)										PS (mg/L)	SiO_2 (mg/L)
Single system	PS-Na	50	80	100	150	200	300	400	500	600	800	10	N.A.
	PS-Mg	1	5	10	20	30	40	50	60	80	100	10	N.A.
	PS-Cr	0.1	0.5	1	1.5	2	3	4	5	6	8	10	N.A.
	PS-SiO_2 (10mg/L)	N.A.										10	10
	PS-SiO_2 (100mg/L)	N.A.										10	100
Simultaneous addition system	PS-Na-SiO_2 (10mg/L)	50	80	100	150	200	300	400	500	600	800	10	10
	PS-Na-SiO_2 (100mg/L)	50	80	100	150	200	300	400	500	600	800	10	100
	PS-Mg-SiO_2 (10mg/L)	1	5	10	20	30	40	50	60	80	100	10	10
	PS-Mg-SiO_2 (100mg/L)	1	5	10	20	30	40	50	60	80	100	10	100
	PS-Cr-SiO_2 (10mg/L)	0.1	0.5	1	1.5	2	3	4	5	6	8	10	10
	PS-Cr-SiO_2 (100mg/L)	0.1	0.5	1	1.5	2	3	4	5	6	8	10	100
Sequential addition system	PS-Na(2h)+SiO_2 (10 mg/L)	50			200				600			10	10
	PS-Na(24h)+SiO_2 (10 mg/L)	50			200				600			10	10
	PS-Na(2h)+SiO_2 (100 mg/L)	50			200				600			10	100
	PS-Na(24h)+SiO_2 (100 mg/L)	50			200				600			10	100
	PS-Mg(2h)+SiO_2 (10 mg/L)	1			30				100			10	10
	PS-Mg(24h)+SiO_2 (10 mg/L)	1			30				100			10	10
	PS-Mg(2h)+SiO_2 (100 mg/L)	1			30				100			10	100
	PS-Mg(24h)+SiO_2 (100 mg/L)	1			30				100			10	100
	PS-Cr(2h)+SiO_2 (10 mg/L)	0.1			2				8			10	10
	PS-Cr(24h)+SiO_2 (10 mg/L)	0.1			2				8			10	10
	PS-Cr(2h)+SiO_2 (100 mg/L)	0.1			2				8			10	100
	PS-Cr(24h)+SiO_2 (100 mg/L)	0.1			2				8			10	100

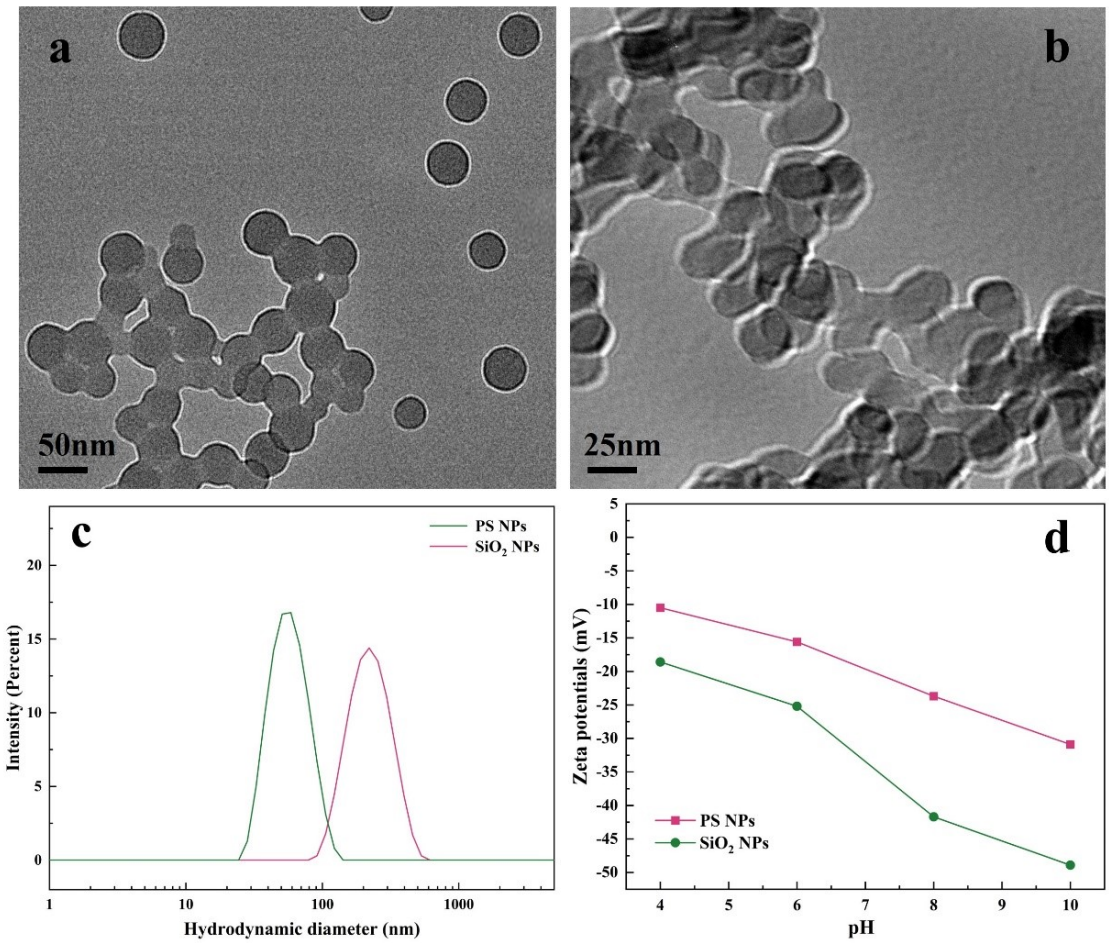

Figure S1. TEM images of standard PS NPs (a) and SiO2 (b) samples, and hydrodynamic particle size (c) and zeta potential (d) measured by DLS.

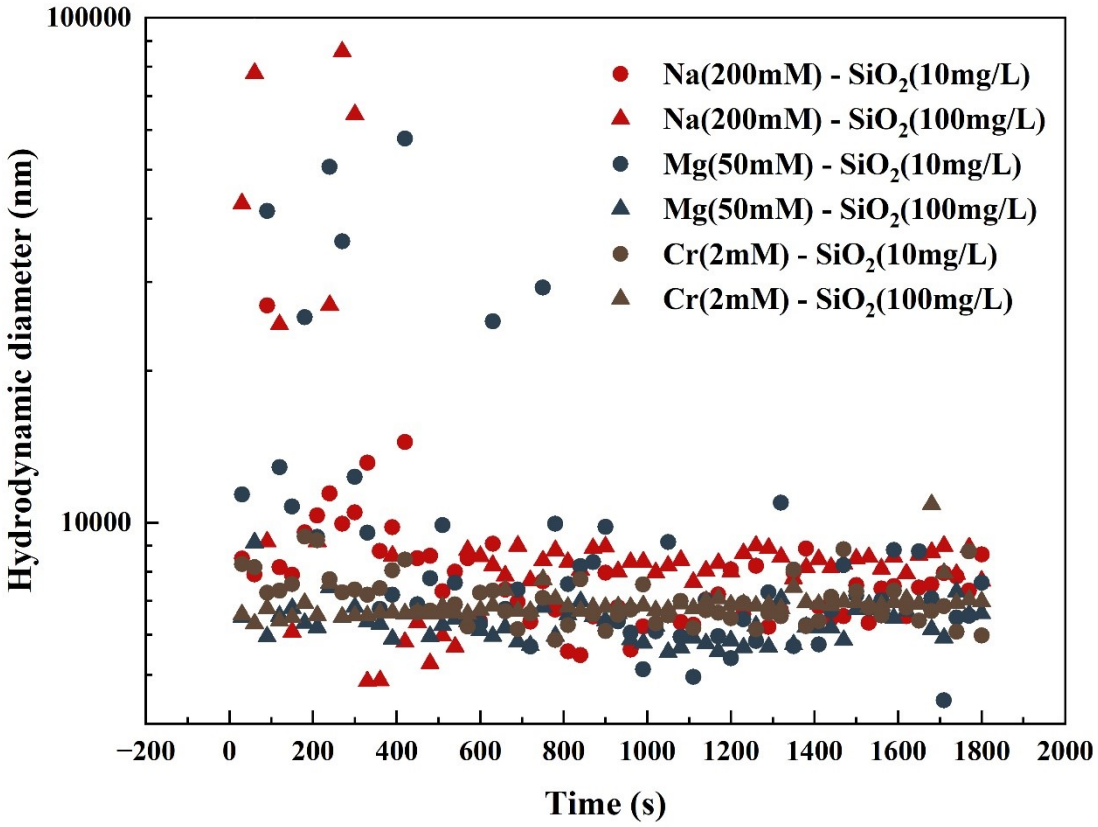

Figure S2. Aggregation dynamics particle size of SiO_2 (10,100mg/L) affected by ions (Na^+ (200mmol/L), Mg^{2+} (50mmol/L), Cr^{3+} (2mmol/L))

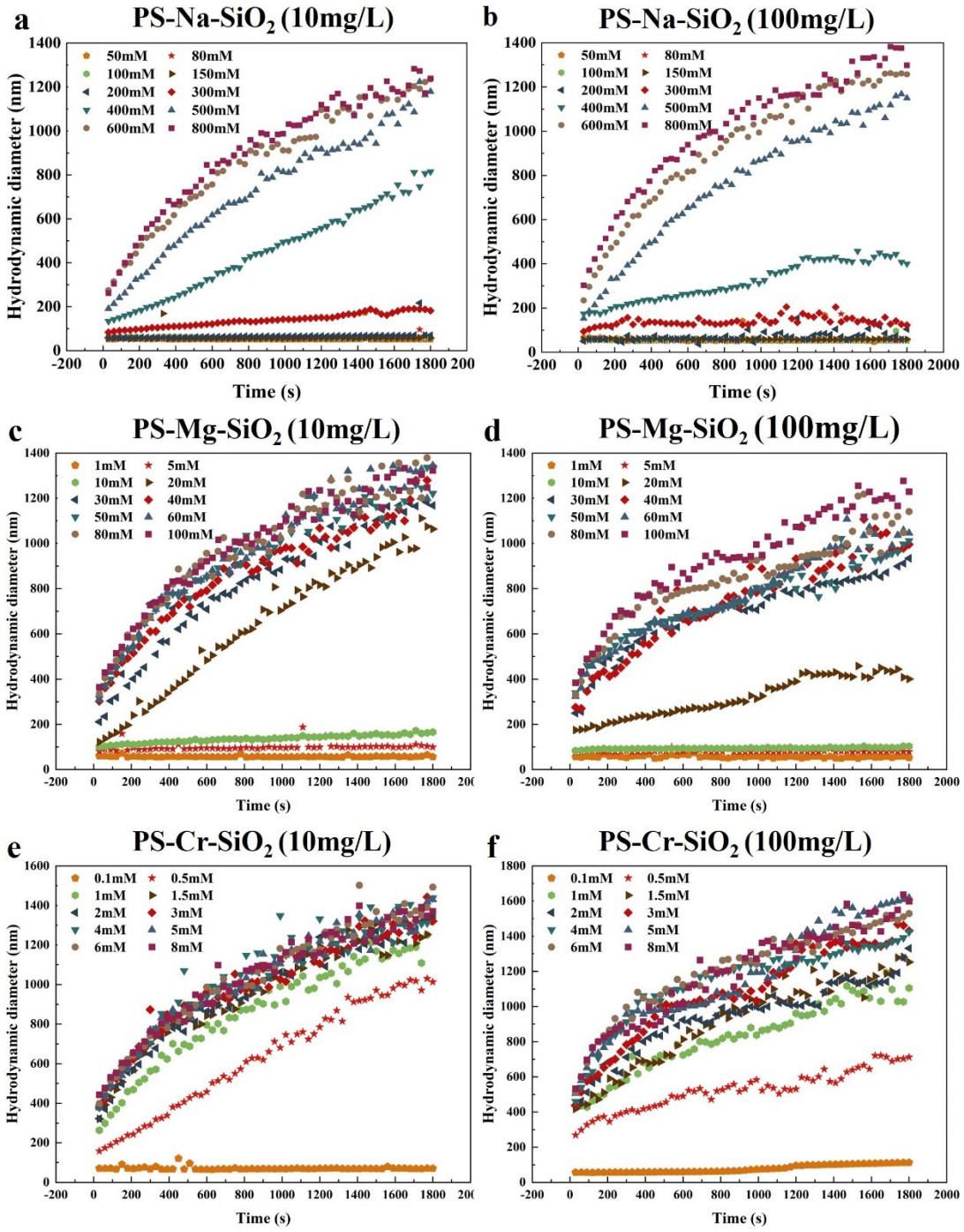

Figure S3. Aggregation dynamics particle size of PSNPs in simultaneously added solutions of different concentrations of ions (Na^+ (a, b), Mg^{2+} (c, d), Cr^{3+} (e, f) and silica (10mg/L (a, c, e), 100mg/L (b, d, f)).

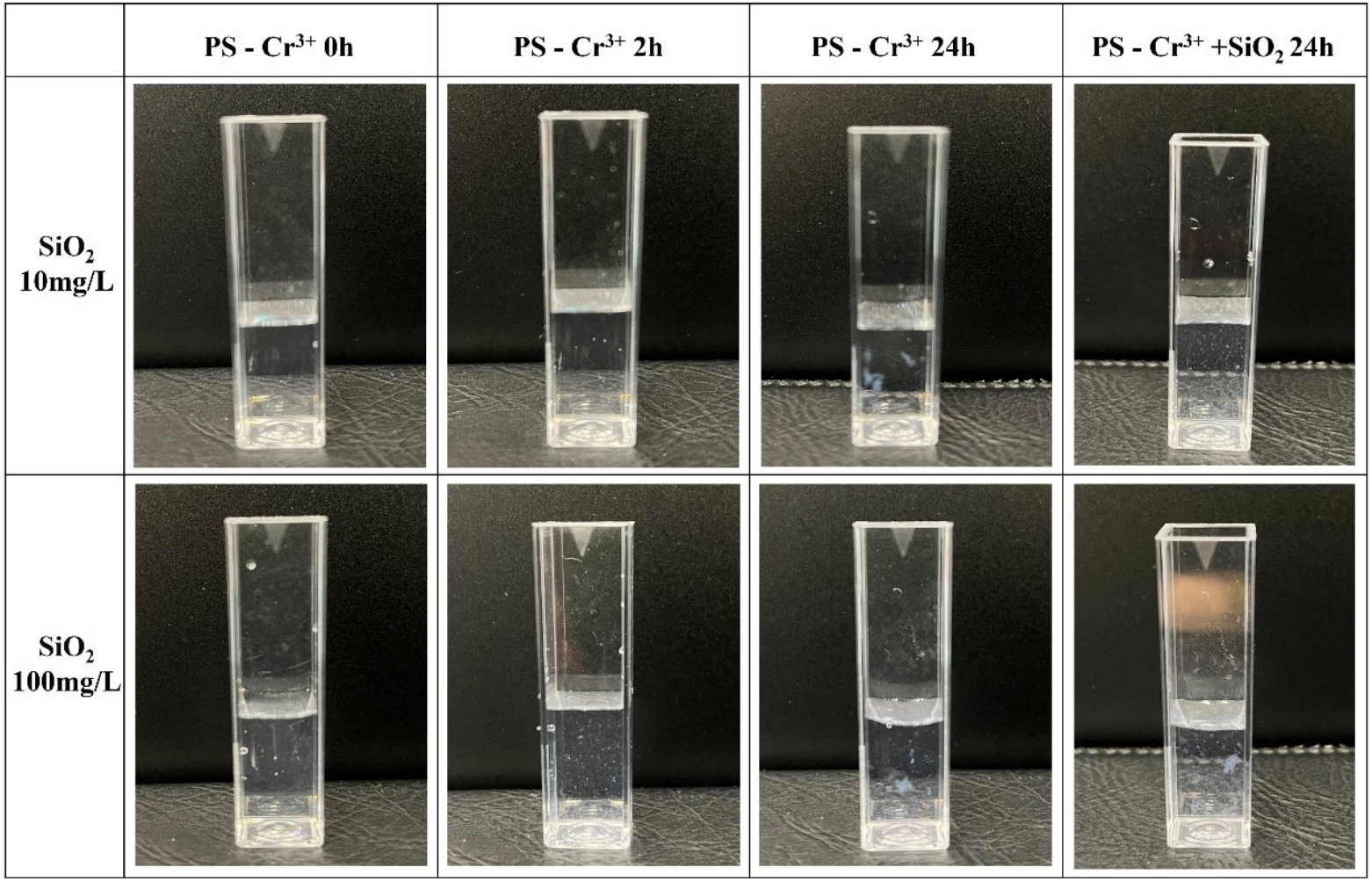

Figure S4. Aggregation of PS NPs at different times of reaction in 8 mmol/L Cr^{3+} solution.

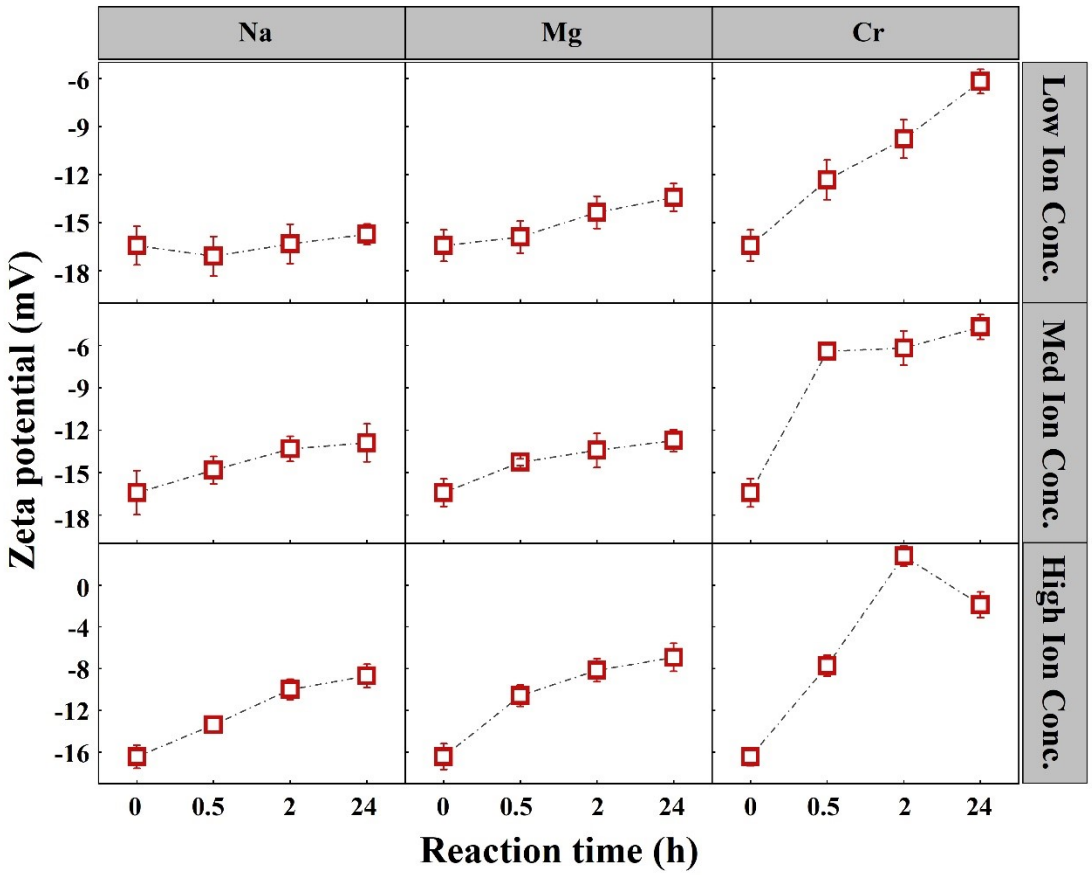

Figure S5. Zeta potential of PS NPs in PS-Cations system at 0, 0.5, 2, and 24 hours under low, medium, and high concentrations of Na^{+} (50, 200, 600 mmol/L), Mg^{2+} (1, 30, 100 mmol/L), and Cr^{3+} (0.1, 2, 8 mmol/L) solutions, respectively.