

Supporting Information for:

How Nanoscale Surface Heterogeneity Impacts Transport of Nano- to Micro- Particles on Surfaces under Unfavorable Attachment Conditions

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Table SI-1. ζ -potential values used in simulations for CML=carboxylate modified polystyrene latex, CMS= carboxylate modified silica, UMS=unmodified silica. Values were determined from EPM measurements via the Smoluchowski equation¹. Note that measurements for colloids > 3 μm have inherent uncertainty using this method.

Material	Colloid Diameter (μm)	NaCl (mM)	pH	ζ -potential (mV)
CML	0.1	6.0	6.7	-45.3
CML	0.3	6.0	6.7	-18.3
CML	1.1	6.0	6.7	-65.4
CML	2.0	6.0	6.7	-29.9
CML	4.4	6.0	6.7	-65.0
CML	6.8	6.0	6.7	-10.2
CML	0.1	20.0	6.7	-35.9
CML	0.3	20.0	6.7	-10.5
CML	1.1	20.0	6.7	-50.1
CML	2.0	20.0	6.7	-8.2
CML	4.4	20.0	6.7	-42.8
CML	6.8	20.0	6.7	-4.5
CML	0.1	6.0	8.0	-61.4
CML	0.3	6.0	8.0	-74.9
CML	1.1	6.0	8.0	-91.0
CML	2.0	6.0	8.0	-80.5
CML	4.4	6.0	8.0	-52.0
CML	6.8	6.0	8.0	-6.9
CML	0.1	20.0	8.0	-42.0
CML	0.3	20.0	8.0	-26.5
CML	1.1	20.0	8.0	-62.2
CML	2.0	20.0	8.0	-63.9
CML	4.4	20.0	8.0	-63.1
CML	6.8	20.0	8.0	-11.8
CML	0.1	50.0	2.0	-13.8
CML	0.3	50.0	2.0	-2.3
CML	1.1	50.0	2.0	-5.1
CML	2.0	50.0	2.0	-5.4
CML	4.4	50.0	2.0	-11.7
CML	6.8	50.0	2.0	1.9
CMS	1.0	6.0	6.7	-66.7
CMS	3.0	6.0	6.7	-15.4
UMS	3.0	6.0	6.7	-27.6
CMS	1.0	50.0	2.0	5.1
CMS	3.0	50.0	2.0	37.4
UMS	3.0	50.0	2.0	-1.7

Table SI-2. ζ -potential values used in simulations for soda-lime glass collector surface (silica). ζ -potential were from representative values reported in the literature.

Material	NaCl (mM)	pH	ζ-potential (mV)
Glass ²	6.0	6.7	-70.0
Glass ²	20.0	6.7	-53.5
Glass ²	6.0	8.0	-80.0
Glass ²	20.0	8.0	-70.0
Glass ³	50.0	2.0	-10.0

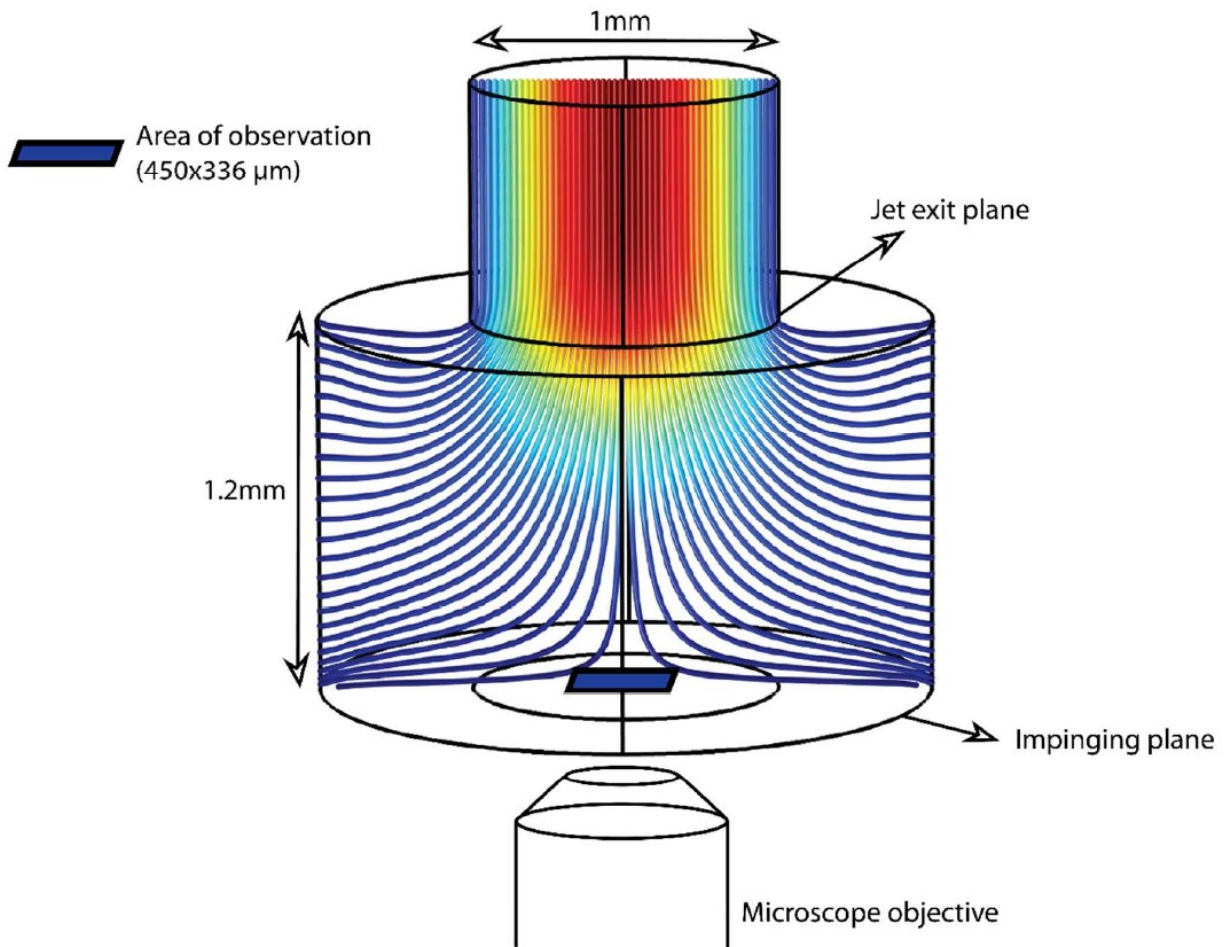


Figure SI-1. Schematic of the impinging jet flow chamber. Fluid flow field is represented by color coded low lines (red high velocity, blue low velocity). The jet is 0.5 mm in radius and the impinging plane is located 1.25 mm below the jet exit. Images of attached colloids are acquired via an inverted microscope across an area of observation of $450 \times 336 \mu\text{m}$ on the impinging plane aligned with the center of the jet.⁴⁻⁶

Table SI-3. RMS roughness values used in simulations regarding attachment of CML onto silica surfaces. Roughness values were determined as described in Rasmuson et al.⁷

CML diameter (μm)	RMS roughness (nm)
0.1	4.7
0.25	6.4
1.1	10.3
2.0	13.0
4.4	17.0
6.8	19.8

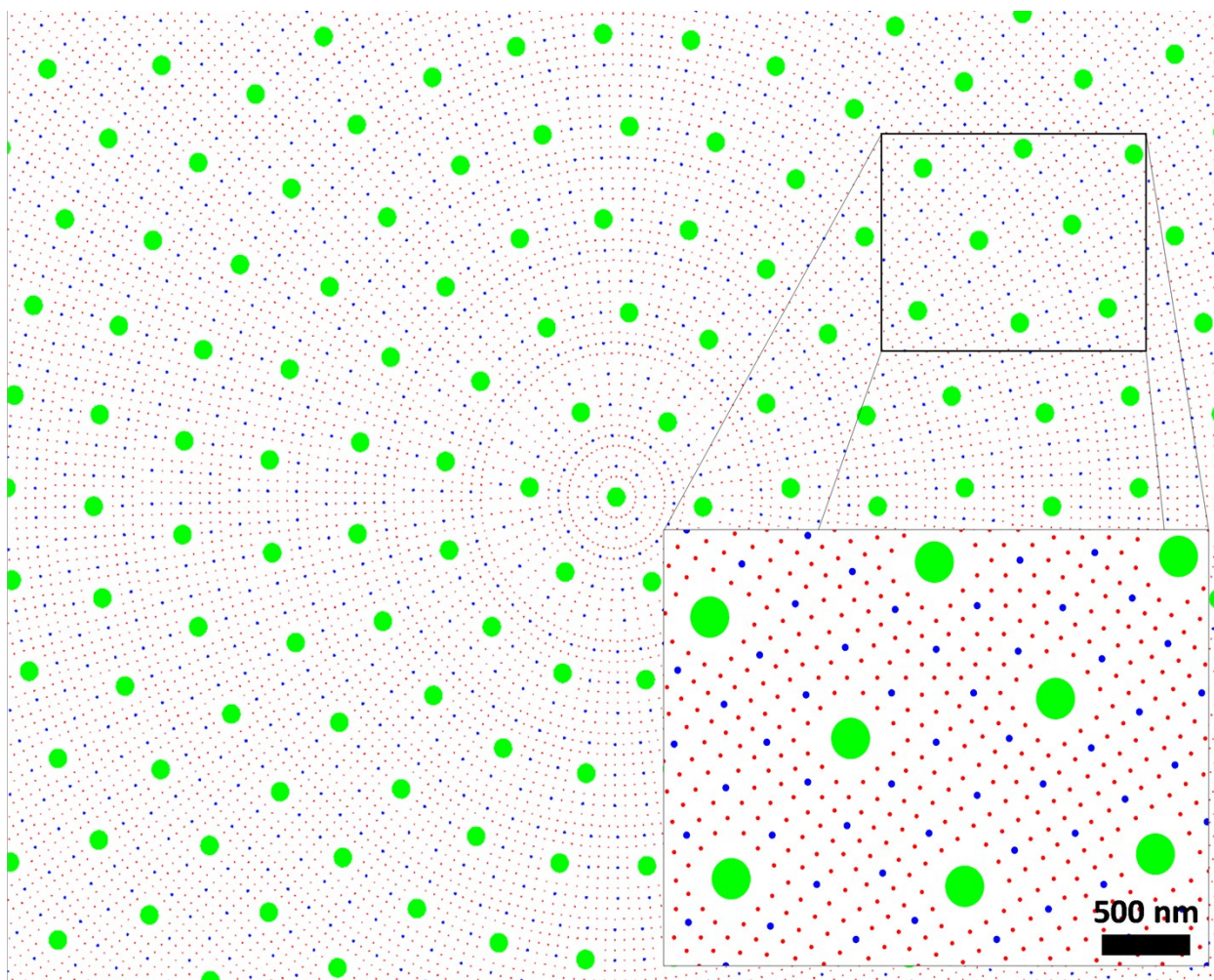


Figure SI-2. Representation of DRNH using power law-distributed heterodomains of three different sizes (green: 220 nm, blue: 40 nm, and red: 25 nm) increasing frequency ratios (1:8:64) superimposed onto the same surface. In simulations, the pattern was expanded to cover the entire surface at a defined spatial density (SD).

Table SI-4. Deposition rate constants (k_f), collector efficiencies (η), and collision efficiencies (α) from published literature meeting criteria provided in main text.

Colloid	Colloid diameter (μm)	Surface	Grain diameter (μm)	Vpore (m/day)	pH	IS (mM)	k_f (1/s)		η		α
							Favorable	Unfavorable	Favorable	Unfavorable	
<i>Cryptosporidium parvum</i> Oocysts ⁸	4.100	Silicate sand	375.0	18.506	8.0	1.0	2.46E-03	3.36E-03	5.03E-03	6.86E-03	1.363
Copolymer-modified Latex Microspheres ⁸	4.300	Silicate sand	375.0	18.506	8.0	1.0	2.48E-03	3.31E-03	5.07E-03	6.75E-03	1.331
Anatase ⁹	0.198	Silicate sand	358.5	101.647	7.0	10.0	1.67E-02	1.52E-03	5.11E-03	4.66E-04	0.091
CML ³	0.110	Glass beads	508.5	4.000	6.7	20.0	2.78E-03	2.00E-04	3.23E-02	2.36E-03	0.073
CML ³	0.200	Glass beads	508.5	4.000	6.7	20.0	2.22E-03	7.22E-05	2.59E-02	8.53E-04	0.033
CML ³	0.500	Glass beads	508.5	4.000	6.7	20.0	1.81E-03	1.75E-05	2.11E-02	2.07E-04	0.010
CML ³	1.000	Glass beads	508.5	4.000	6.7	20.0	1.11E-03	6.94E-05	1.30E-02	8.20E-04	0.063
CML ³	2.000	Glass beads	508.5	4.000	6.7	20.0	1.14E-03	2.56E-04	1.34E-02	3.01E-03	0.226
MS2 ¹⁰	0.025	Ohio river sand	245.0	38.750	7.0	6.8	1.77E-01	2.91E-04	9.05E-02	1.56E-04	0.002
PRD1 ¹⁰	0.065	Ohio river sand	245.0	38.750	7.0	6.8	8.07E-02	9.87E-05	4.23E-02	5.29E-05	0.001
OY107 ¹⁰	0.690	Ohio river sand	245.0	38.750	7.0	6.8	1.26E-02	2.70E-05	6.74E-03	1.45E-05	0.002
DA001 ¹⁰	0.850	Ohio river sand	245.0	38.750	7.0	6.8	1.10E-02	5.72E-05	5.87E-03	3.06E-05	0.005
E. Coli ¹⁰	1.000	Ohio river sand	245.0	38.750	7.0	6.8	1.00E-02	5.73E-05	5.34E-03	3.07E-05	0.006
<i>Cryptosporidium parvum</i> Oocysts ¹⁰	5.500	Ohio river sand	245.0	38.750	7.0	6.8	2.17E-02	1.83E-02	1.16E-02	9.75E-03	0.842
<i>Cryptosporidium parvum</i> Oocysts ¹¹	4.000	Glass beads	328.0	19.382	8.0	10.0	5.80E-03	1.70E-03	8.94E-03	2.63E-03	0.294
Carboxylate-modified Latex Microspheres ¹¹	3.000	Glass beads	328.0	19.382	8.0	10.0	4.17E-03	1.29E-04	6.43E-03	2.00E-04	0.031

Table SI-4 (Continued). Deposition rate constants (k_f), collector efficiencies (η), and collision efficiencies (α)

Cerium Oxide ¹²	0.063	Silicate sand	717.0	167.169	6.0	1.0	8.72E-03	3.23E-03	3.26E-03	1.21E-03	0.370
Silica ¹³	0.052	Silicate sand	220.0	3.710	10.0	1.0	5.97E-02	8.89E-06	2.66E-01	4.60E-05	0.000
Silica ¹³	0.008	Silicate sand	220.0	3.710	10.0	1.0	8.26E-01	4.17E-05	9.86E-01	2.16E-04	0.000
Zinc Oxide ¹⁴	0.300	Quartz sand	510.0	8.023	8.0	5.0	3.29E-03	4.72E-04	2.06E-02	2.98E-03	0.145
Rutile ¹⁵	0.149	Quartz sand	275.0	11.909	6.0	1.0	2.06E-02	6.37E-04	4.25E-02	1.34E-03	0.032
Zero valent Iron ¹⁶	0.065	Quartz sand	725.0	73.339	8.0	-	1.09E-02	5.37E-03	1.03E-02	5.08E-03	0.494
DA001 ¹⁷	1.100	Glass beads	508.5	4.002	6-9	4.0	1.11E-03	5.56E-05	1.30E-02	6.56E-04	0.050
<i>Escherichia Coli</i> D21g ¹⁸	1.700	Quartz sand	205.0	42.195	5.7	3.2	1.11E-03	2.40E-04	1.30E-02	1.18E-04	0.009
<i>Escherichia Coli</i> D21g ¹⁹	0.960	Quartz sand	275.0	31.930	5.7	10.0	1.70E-02	6.00E-04	1.55E-02	5.51E-04	0.036
<i>Escherichia Coli</i> XL1-Blue ¹⁹	1.100	Quartz sand	275.0	31.930	5.7	10.0	1.70E-02	4.80E-04	1.55E-02	4.41E-04	0.028
Carboxylate-modified latex microspheres ^(This study)	0.110	Soda-lime glass	Impinging jet	2.000	6.7	6.0	-	-	0.000114	4.70309E-05	0.413
Carboxylate-modified latex microspheres ^(This study)	0.250	Soda-lime glass	Impinging jet	2.000	6.7	6.0	-	-	3.16E-05	6.27953E-06	0.199
Carboxylate-modified latex microspheres ^(This study)	1.100	Soda-lime glass	Impinging jet	2.000	6.7	6.0	-	-	1.94E-05	1.85494E-06	0.096
Carboxylate-modified latex microspheres ^(This study)	2.000	Soda-lime glass	Impinging jet	2.000	6.7	6.0	-	-	1.06E-05	1.51048E-06	0.143
Carboxylate-modified latex microspheres ^(This study)	4.400	Soda-lime glass	Impinging jet	2.000	6.7	6.0	-	-	4.15E-05	3.03814E-05	0.732
Carboxylate-modified latex microspheres ^(This study)	6.800	Soda-lime glass	Impinging jet	2.000	6.7	6.0	-	-	0.000178	0.0000919	0.518
Carboxylate-modified latex microspheres ^(This study)	0.110	Soda-lime glass	Impinging jet	2.000	6.7	20.0	-	-	0.000114	5.38031E-05	0.473
Carboxylate-modified latex microspheres ^(This study)	0.250	Soda-lime glass	Impinging jet	2.000	6.7	20.0	-	-	3.16E-05	1.6654E-05	0.528
Carboxylate-modified latex microspheres ^(This study)	1.100	Soda-lime glass	Impinging jet	2.000	6.7	20.0	-	-	1.94E-05	4.36965E-06	0.225

Table SI-4 (Continued). Deposition rate constants (k_f), collector efficiencies (η), and collision efficiencies (α)

Carboxylate-modified latex microspheres ^(This study)	2.000	Soda-lime glass	Impinging jet	2.000	6.7	20.0	-	-	1.06E-05	5.2718E-06	0.498
Carboxylate-modified latex microspheres ^(This study)	4.400	Soda-lime glass	Impinging jet	2.000	6.7	20.0	-	-	4.15E-05	4.74588E-05	1.144
Carboxylate-modified latex microspheres ^(This study)	6.800	Soda-lime glass	Impinging jet	2.000	6.7	20.0	-	-	0.000178	0.000122502	0.690
Carboxylate-modified latex microspheres ^(This study)	0.110	Soda-lime glass	Impinging jet	2.000	8.0	6.0	-	-	0.000114	2.1338E-05	0.188
Carboxylate-modified latex microspheres ^(This study)	0.250	Soda-lime glass	Impinging jet	2.000	8.0	6.0	-	-	3.16E-05	4.62587E-07	0.015
Carboxylate-modified latex microspheres ^(This study)	1.100	Soda-lime glass	Impinging jet	2.000	8.0	6.0	-	-	1.94E-05	7.55524E-08	0.004
Carboxylate-modified latex microspheres ^(This study)	2.000	Soda-lime glass	Impinging jet	2.000	8.0	6.0	-	-	1.06E-05	1.60113E-07	0.015
Carboxylate-modified latex microspheres ^(This study)	4.400	Soda-lime glass	Impinging jet	2.000	8.0	6.0	-	-	4.15E-05	3.03814E-05	0.732
Carboxylate-modified latex microspheres ^(This study)	6.800	Soda-lime glass	Impinging jet	2.000	8.0	6.0	-	-	0.000178	0.000108403	0.611
Carboxylate-modified latex microspheres ^(This study)	0.110	Soda-lime glass	Impinging jet	2.000	8.0	20.0	-	-	0.000114	1.47937E-05	0.130
Carboxylate-modified latex microspheres ^(This study)	0.250	Soda-lime glass	Impinging jet	2.000	8.0	20.0	-	-	3.16E-05	5.19027E-06	0.164
Carboxylate-modified latex microspheres ^(This study)	1.100	Soda-lime glass	Impinging jet	2.000	8.0	20.0	-	-	1.94E-05	3.65429E-07	0.019
Carboxylate-modified latex microspheres ^(This study)	2.000	Soda-lime glass	Impinging jet	2.000	8.0	20.0	-	-	1.06E-05	4.96712E-07	0.047
Carboxylate-modified latex microspheres ^(This study)	4.400	Soda-lime glass	Impinging jet	2.000	8.0	20.0	-	-	4.15E-05	1.85407E-05	0.447
Carboxylate-modified latex microspheres ^(This study)	6.800	Soda-lime glass	Impinging jet	2.000	8.0	20.0	-	-	0.000178	0.000201867	1.137
Carboxylate-modified silica microspheres ^(This study)	1.000	Soda-lime glass	Impinging jet	2.000	6.7	6.0	-	-	9.33E-05	2.21681E-06	0.024
Carboxylate-modified silica microspheres ^(This study)	3.000	Soda-lime glass	Impinging jet	2.000	6.7	6.0	-	-	0.000366	0.000387712	1.060

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