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Supporting information

Structure evolution of aluminosilicate sol and its structure-directing effect on the synthesis of NaY zeolite

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Cascade tangent rule

The cascade tangent rule assumes that the particle size distribution is discrete of the polydisperse system. The scattering intensity can be described as:

$$\mathrm{I}(q) = I_e N_1 n_1^2 e^{-q^2 R_1^2/3} + I_e N_2 n_2^2 e^{-q^2 R_2^2/3} + \cdots + I_e N_i n_i^2 e^{-q^2 R_i^2/3} \quad \text{(Eq. S1)}$$

Where I_e is the scattering intensity of one electron, R_i, N_i and n_i are the gyration radius, the particle number and the electron number of particle with the ith size level, respectively, V_i is the volume of particle with R_i, so V_i be represented as:

$$V_i = P_1 R_i^3 \quad \text{(Eq. S2)}$$

$$n_i = \rho P_1 R_i^3 \quad \text{(Eq. S3)}$$

P_1 is a constant, ρ is the electron density in particle with R_i.

If we define

$$K_i = I_e N_i n_i^2 \quad \text{(Eq. S4)}$$

When $q = 0$,

$$I(0) = K_1 + K_2 + \cdots + K_i \quad \text{(Eq. S5)}$$

$$K_i = I_e N_i n_i^2 = I_e N_i (\rho V_i)^2 = I_e N_i V_i \cdot V_i \rho^2 = I_e W_i V_i \rho^2 = I_e W_i P_1 R_i^3 \rho^2 \quad \text{(Eq. S6)}$$

$$W_i = N_i V_i \quad \text{(Eq. S7)}$$

W_i is the total volume of particles with R_i

If we define

$$\sum W_i = 1, \quad \text{(Eq. S8)}$$

the Wi is the volume percentage of particles with R_i.

$$W_1{:}W_2{:}\ldots{:}W_i = \frac{K_1}{R_1^3}{:}\frac{K_2}{R_2^3}{:}\ldots{:}\frac{K_\mathrm{i}}{R_\mathrm{i}^3} \quad \text{(Eq. S9)}$$

A tangent to the experimental curve is drawn at the greatest angle of scattering studied. This tangent intersects the axis of ordinates at the value K_1 or $\ln(K_1)$, R_1 can be obtained from the slope ($-R_1^2/3$). The values corresponding to this tangent are then subtracted in linear intensity scale from the original curve ($\ln[I_1(q)]$) and a new corrected curve ($\ln[I_2(q)]$) is obtained (shown in Figure S3). Repeating the above procedure, the second tangent is performed on curve $\ln[I_2(q)]$ with its intercept K_2 or $\ln(K_2)$. The procedure is repeated until the final points yield a straight line of intercept. Finally, all the parameters (K_i, R_i) can be obtained. The cascade tangent rule for different samples are illustrated in Figure S3.

Monte-Carlo method

Monte Carlo method (Pauw *et al.*, 2013; Bressler *et al.*, 2015) is a novel way to obtain accurate, form-free size distributions from SAXS data of non-interacting low-concentration scatterers. Briefly, the method starts from a set of non-interacting scatterers of predefined shape (e.g. spheres, rods, ellipsoids). The SAXS pattern is then calculated and compared to the experimental one. A change in the size distribution is then performed and if this change results in a better fit, the change is accepted. This step is reproduced until a convergence criterium is met.

The MC method calculates a scattering pattern $I_{\mathrm{MC}}(q)$ using the general equation (spheres):

$$I_{\mathrm{MC}}(q) = b + A\sum_{k=1}^{n_s}\left|F_{sph,k}(qR_k)\right|^2\left(\frac{3}{4}\pi\right)^2 R_k^{(6-p_c)} \quad \text{(Eq. S10)}$$

Is the Rayleigh from factor for sphere k, normalized to 1 for q = 0. R_k is the radius for sphere k. p_c is a parameter adjustable in the range $0 \leq p_c \leq 6$, the recommended valve for $p_c = 3$.

b is a constant background term. A is a scaling factor.

$$\mathrm{A} = \varphi\Delta\rho^2\sum_{k=1}^{n_s} 1\Big/\left[\frac{4}{3}\pi R_k^{(3-p_c)}\right] \quad \text{(Eq. S11)}$$

$\Delta\rho$ is the scattering contrast. In our paper the electron density contrast of the scattering phases is ill-defined and the absolute volume fractions will be affected, but the size distribution is still correct.

The volume fraction of the scatterers is defined as

$\varphi = V_{scatt}/V_{irr}$ (Eq. S12)

Where V_{irr} is the irradiated sample volume and V_{scatt} is the total scatterer volume in Virr.

Supporting Figures

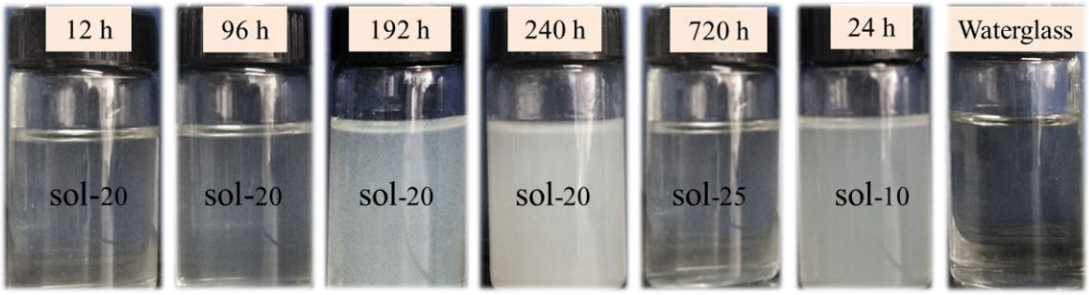

Figure S1 Appearance condition of aluminosilicate sols and water glass during the aging process for each period.

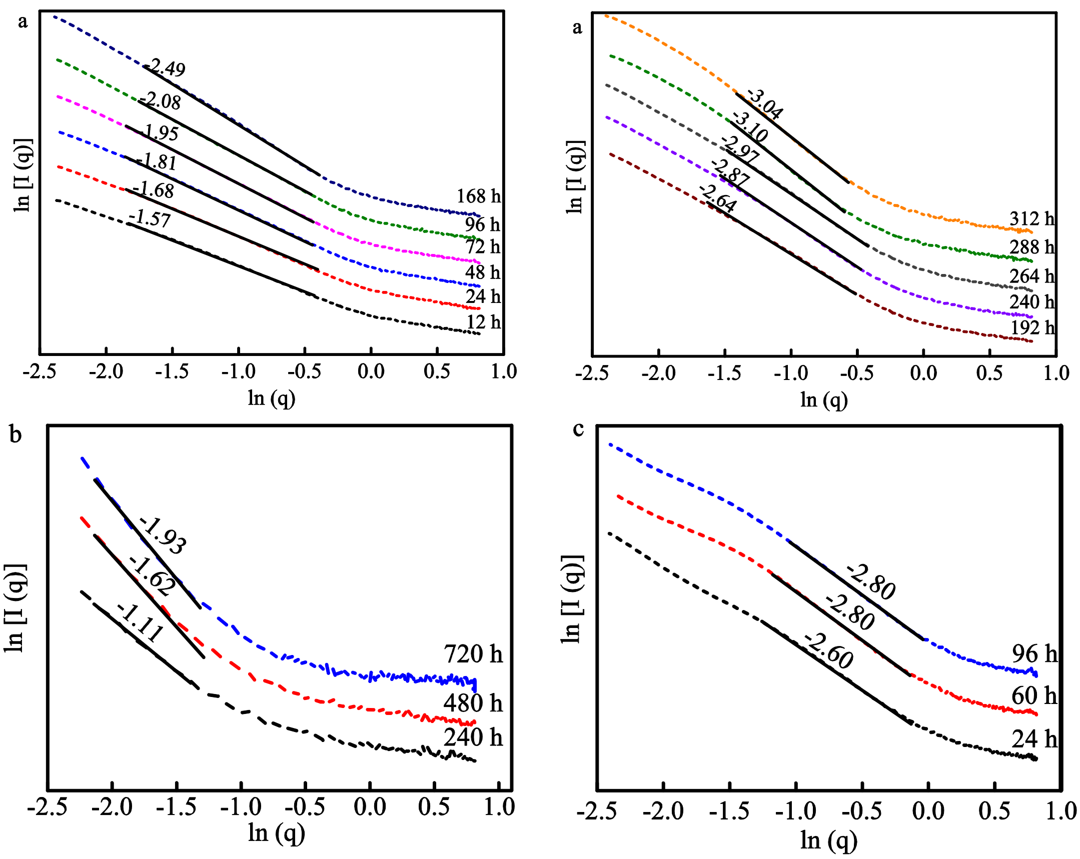

Figure S2 Shifted scattering curves of samples. (a), the sol-20, range of linearity: 0.1353 < q <1.65 nm^{-1}, the figure on the left: aging time from 12 h to 168, the figure on the right: aging time from 192 h to 312 h; (b), sol-25, range of linearity: 0.1054 < q < 0.223 nm^{-1}; (c), and sol-10, range of linearity: 0.223 < q <1.00 nm^{-}

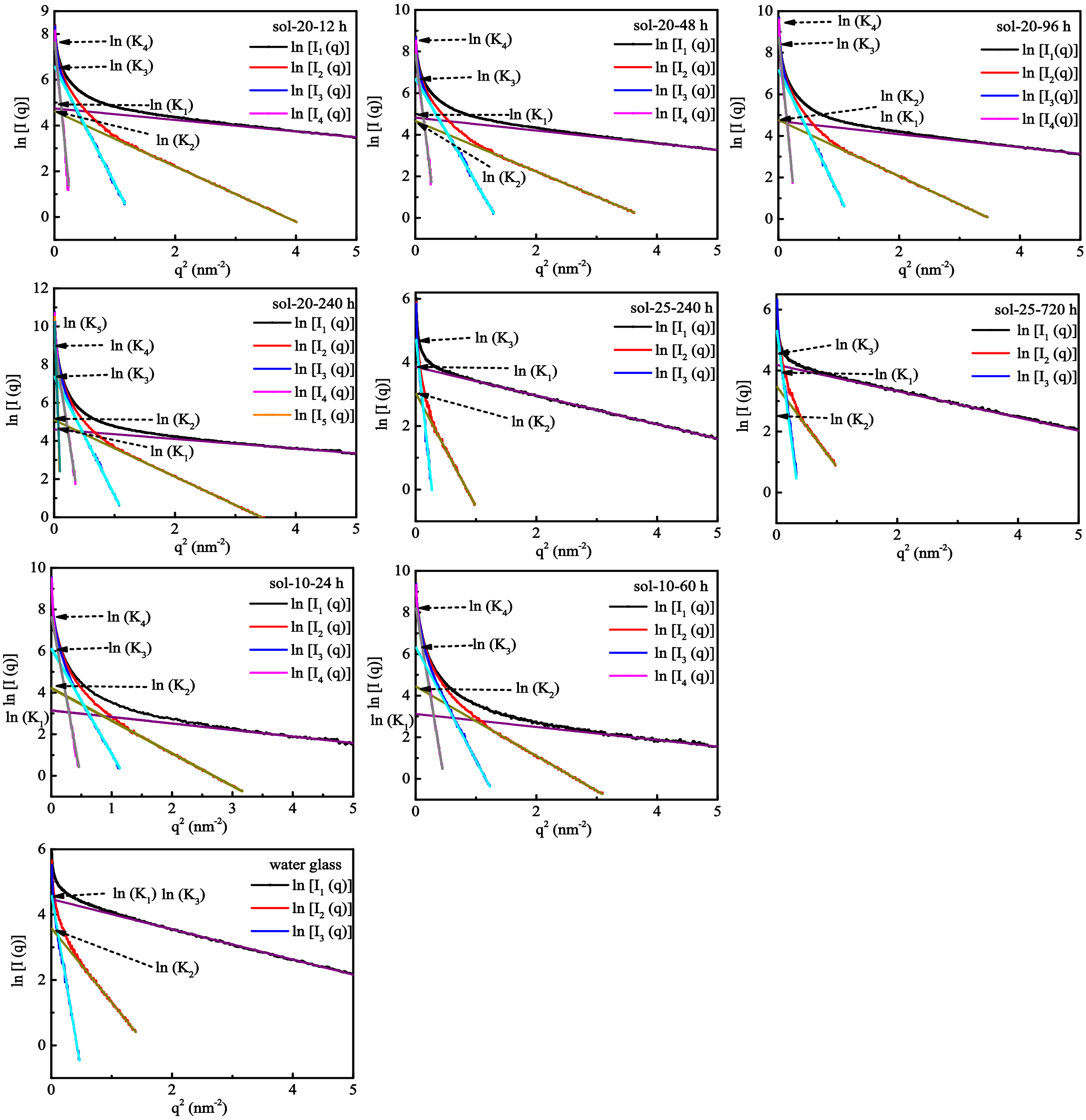

Figure S3 $\ln[I_i(q)]{\sim}q^2$ curves for samples (sol-20-12 h, sol-20-48 h, sol-20-96 h, sol-20-240 h, sol-25-240 h, sol-25-720 h, sol-10-24 h, sol-10-60 h, sol-10-96 h and water glass) and detailed illustrations of the cascade tangent rule.

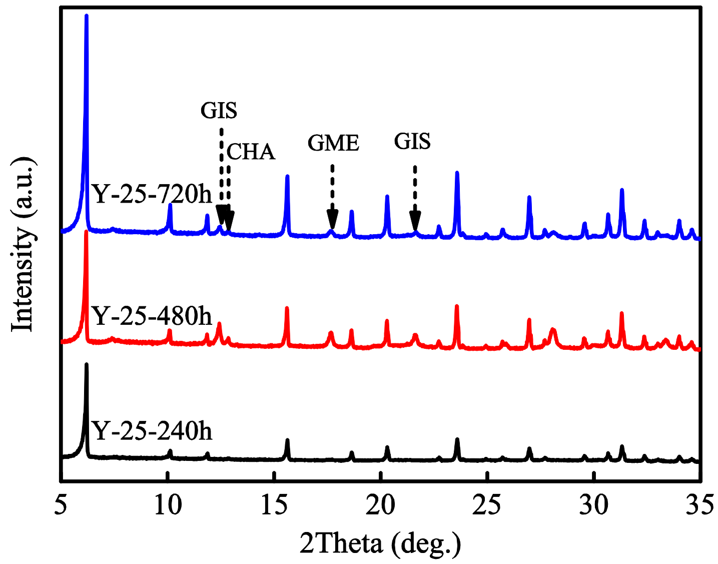

Figure S4 XRD patterns of the so-synthesized products using sol-25-240 h, sol-25-480 h and sol-25-720 h as SDA.

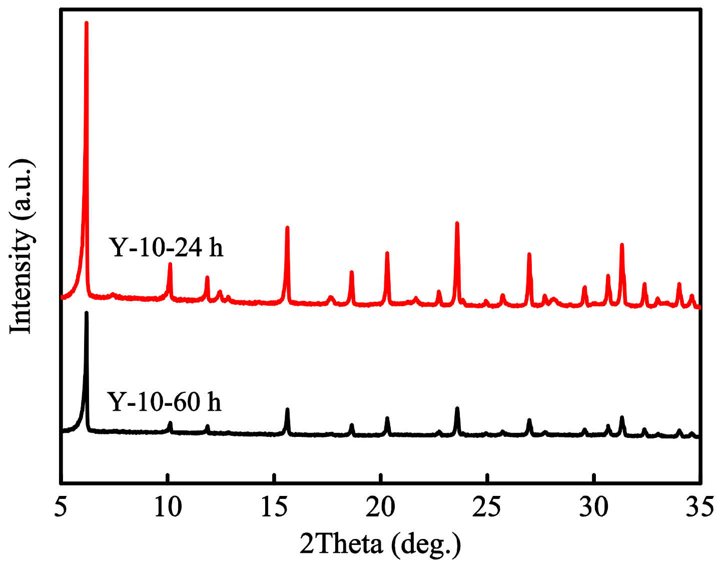

Figure S5 XRD patterns of the so-synthesized products using sol-10- 24 h and sol-10-60 h as SDA.

Table S1

Calculation of particle sizes by cascade tangent rule from scattering data: sol-20-12h.

Intercept		linear fitting range of q^2		Calculation of the slope		R_i	$\frac{K_i}{R_i^3}$	Volume fraction/ W_i
$\ln K_i$	K_i	Maximum (nm^{-2})	Minimum (nm^{-2})	$\Delta\ln\ [I\ (q)]$	$\frac{\Delta\ln\ [I\ (q)]}{\Delta q^2}$			
4.7465	115.181	5.106	3.416	0.425	0.252	0.87	174.913	0.852
4.6252	102.021	4.000	1.342	3.198	1.208	1.90	14.787	0.072
6.6002	735.212	1.167	0.395	3.974	5.148	3.93	12.110	0.059
7.8549	2578.321	0.395	0.009	10.445	27.060	9.04	3.490	0.017

Table S2

Calculation of particle sizes by cascade tangent rule from scattering data: sol-20-48h.

Intercept		linear fitting range of q^2		Calculation of the slope		R_i	$\frac{K_i}{R_i^3}$	Volume fraction /W_i
$\ln K_i$	K_i	Maximum (nm^{-2})	Minimum (nm^{-2})	$\Delta\ln\ [I\ (q)]$	$\frac{\Delta\ln\ [I\ (q)]}{\Delta q^2}$			
4.821	124.104	5.100	3.414	0.518	0.307	0.96	138.6629	0.801
4.662	105.864	3.626	1.214	2.933	1.216	1.91	15.19311	0.088
5.772	321.078	1.297	0.438	4.288	4.992	3.87	5.539591	0.082
8.039	3100.168	0.438	0.008	10.162	23.632	8.42	5.193367	0.030

Table S3

Calculation of particle sizes by cascade tangent rule from scattering data: sol-20-96h.

Intercept		linear fitting range of q^2		Calculation of the slope		R_i	$\frac{K_i}{R_i^3}$	Volume fraction $/W_i$
$\ln K_i$	K_i	Maximum (nm^{-2})	Minimum (nm^{-2})	$\Delta\ln\,[I\ (q)]$	$\frac{\Delta\ln\ [I\ (q)]}{\Delta q^2}$			
4.708	110.819	5.103	3.414	0.530	0.314	0.97	121.423	0.747
4.766	117.437	3.466	1.616	2.491	1.351	2.01	14.462	0.089
7.206	1347.628	1.093	0.370	4.353	6.021	4.25	17.555	0.108
8.980	7943.745	0.370	0.008	10.890	30.083	9.50	9.265	0.057

Table S4

Calculation of particle sizes by cascade tangent rule from scattering data: sol-20-240h.

Intercept		linear fitting range of q^2		Calculation of the slope		R_i	$\frac{K_i}{R_i^3}$	Volume fraction $/W_i$
$\ln K_i$	K_i	Maxi-mum (nm^{-2})	Mini-mum (nm^{-2})	$\Delta\ln\,[I\ (q)]$	$\frac{\Delta\ln\ [I\ (q)]}{\Delta q^2}$			
4.602	99.728	5.101	3.413	0.426	0.251	0.87	151.447	0.679
5.096	163.326	3.479	1.165	3.434	1.487	2.11	17.386	0.077
7.414	1659.262	1.079	0.365	4.483	6.267	4.34	20.298	0.091
9.218	10078.8	0.350	0.122	4.731	20.751	7.89	20.520	0.092
10.966	57850.13	0.122	0.008	10.084	88.455	16.29	13.383	0.060

Table S5

Calculation of particle sizes by cascade tangent rule from scattering data: sol-10-24h.

Intercept		linear fitting range of q^2		Calculation of the slope		R_i	$\frac{K_i}{R_i^3}$	Volume fraction $/W_i$
$\ln K_i$	K_i	Maximum (nm^{-2})	Minimum (nm^{-2})	$\Delta\ln[I(q)]$	$\frac{\Delta\ln[I(q)]}{\Delta q^2}$			
3.144	23.206	5.103	3.414	0.530	0.314	0.97	25.427	0.541
4.223	68.206	3.100	1.060	3.202	1.570	2.17	6.675	0.142
6.141	464.557	1.100	0.384	3.630	5.078	3.90	7.813	0.166
7.761	2346.289	0.400	0.158	3.863	15.962	6.92	7.081	0.151

Table S6

Calculation of particle sizes by cascade tangent rule from scattering data: sol-10-60h.

Intercept		linear fitting range of q^2		Calculation of the slope		R_i	$\frac{K_i}{R_i^3}$	Volume fraction $/W_i$
$\ln K_i$	K_i	Maximum (nm^{-2})	Minimum (nm^{-2})	$\Delta\ln[I(q)]$	$\frac{\Delta\ln[I(q)]}{\Delta q^2}$			
3.122	22.686	5.103	3.414	0.530	0.3136	0.97	23.388	0.464
4.447	85.346	3.100	1.060	3.412	1.673	2.24	7.593	0.146
6.362	579.271	1.100	0.384	3.915	5.468	4.05	8.720	0.173
8.352	4239.567	0.400	0.158	4.275	17.666	7.28	10.988	0.218

Table S7

Calculation of particle sizes by cascade tangent rule from scattering data: sol-10-96h.

Intercept		linear fitting range of q^2		Calculation of the slope		R_i	$\frac{K_i}{R_i^3}$	Volume fraction $/W_i$
$\ln K_i$	K_i	Maximum (nm^{-2})	Minimum (nm^{-2})	$\Delta\ln\ [I\ (q)]$	$\frac{\Delta\ln\ [I\ (q)]}{\Delta q^2}$			
3.071	21.573	5.070	3.411	0.520	0.314	0.97	23.637	0.404
4.687	108.570	3.200	1.080	3.674	1.733	2.28	9.160	0.156
6.760	862.775	1.170	0.396	4.443	5.741	4.15	12.071	0.205
8.237	3777.988	0.300	0.138	2.949	18.204	7.39	9.361	0.160
9.069	8684.621	0.138	0.008	6.771	52.083	12.5	4.447	0.076

Table S8

Calculation of particle sizes by cascade tangent rule from scattering data: water glass.

Intercept		linear fitting range of q^2		Calculation of the slope		R_i	$\frac{K_i}{R_i^3}$	Volume fraction $/W_i$
$\ln K_i$	K_i	Maximum (nm^{-2})	Minimum (nm^{-2})	$\Delta\ln\ [I\ (q)]$	$\frac{\Delta\ln\ [I\ (q)]}{\Delta q^2}$			
4.474	87.728	5.094	1.400	1.708	0.462	1.18	53.394	0.954
3.594	36.390	1.400	0.466	2.125	2.275	2.61	2.0467	0.036
4.679	107.681	0.460	0.010	4.939	10.975	5.74	0.569	0.010

Table S9

Calculation of particle sizes by cascade tangent rule from scattering data: sol-25-240h.

Intercept		linear fitting range of q^2		Calculation of the slope		R_i	$\frac{K_i}{R_i^3}$	Volume fraction $/W_i$
$\ln K_i$	K_i	Maximum (nm^{-2})	Mini-mum (nm^{-2})	$\Delta\ln\ [I\ (q)]$	$\frac{\Delta\ln\ [I\ (q)]}{\Delta q^2}$			
3.859	47.409	5.086	1.000	1.847	0.452	1.16	30.373	0.970
3.046	21.031	0.980	0.330	2.326	3.578	3.28	0.596	0.019
4.888	132.748	0.270	0.020	4.530	18.118	7.37	0.332	0.011

Table S10

Calculation of particle sizes by cascade tangent rule from scattering data: sol-25-720h.

Intercept		linear fitting range of q^2		Calculation of the slope		R_i	$\frac{K_i}{R_i^3}$	Volume fraction $/W_i$
$\ln K_i$	K_i	Maximum (nm^{-2})	Mini-mum (nm^{-2})	$\Delta\ln\ [I\ (q)]$	$\frac{\Delta\ln\ [I\ (q)]}{\Delta q^2}$			
4.048	57.283	5.097	1.000	2.491	0.608	1.35	23.282	0.956
3.686	39.885	0.581	0.200	1.809	4.750	3.79	0.733	0.030
5.520	249.635	0.200	0.030	6.021	35.42	8.95	0.348	0.014

References

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