Supporting Information for

Time-lapse Structural Insights Into the Self-assembly Event in a Slow Evolving and Mechanically Robust Supramolecular Gel

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Materials and method

1-(*N*-Boc-aminomethyl)-4-(aminomethyl)benzene, Benzyl isocyanate and silica gel were purchased from Sigma-Aldrich. Solvents were purchased from Fisher Scientific. DMSO-d_6 and CDCl$_3$ were purchased from Cambridge Isotope Laboratories.

NMR spectra were recorded in a Bruker AV 400 MHz spectrometer. The chemical shifts are expressed in parts per million (δ, ppm).

ESI-MS was carried out in Orbitrap Fusion Lumos, from Thermo Scientific, San Jose California.
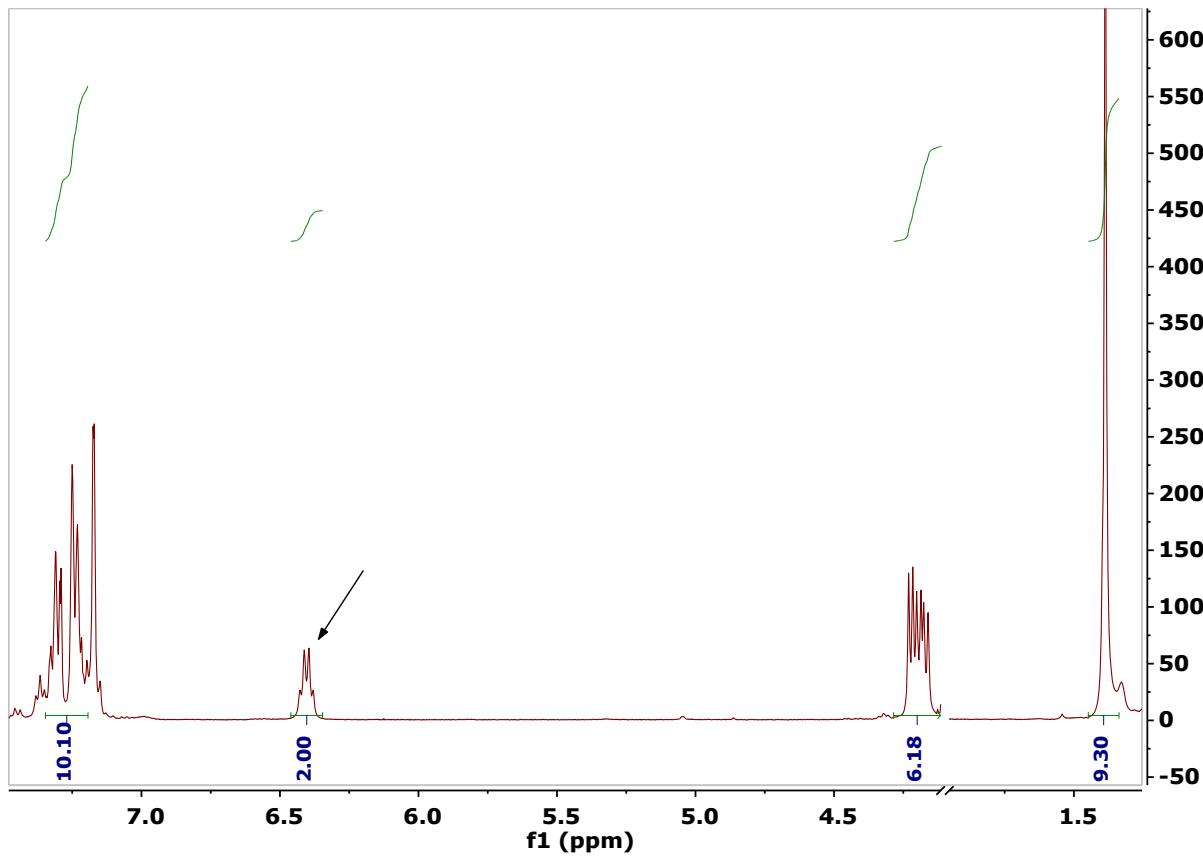
Figure S1. ^{1}H NMR of **1** in DMSO-d$_6$ (signal associated with the urea hydrogens are marked with arrow).
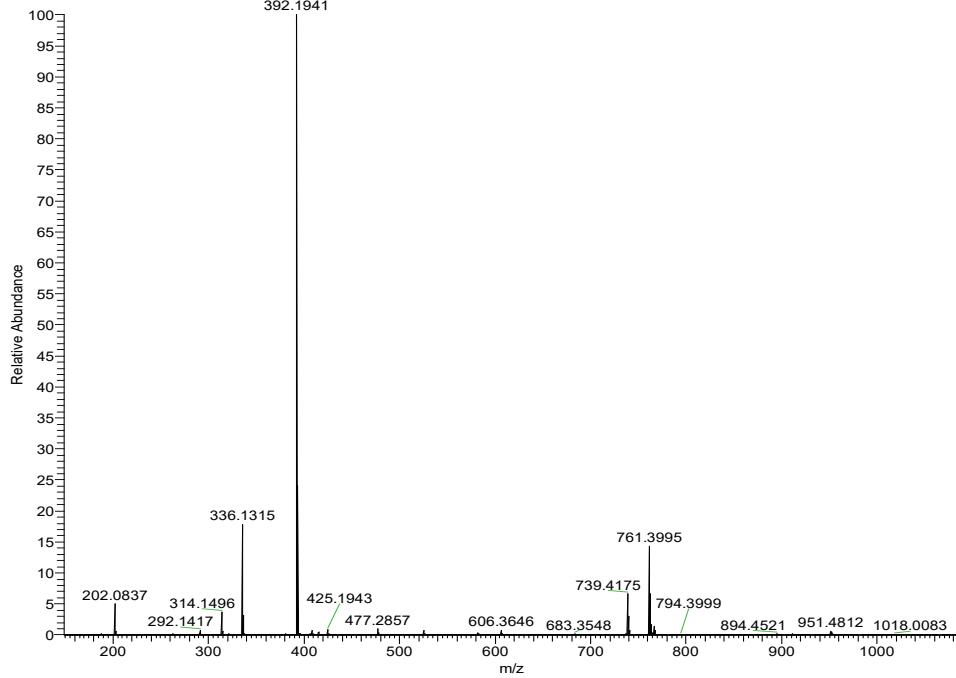

**Figure S2.** ESI mass spectra of **1**.
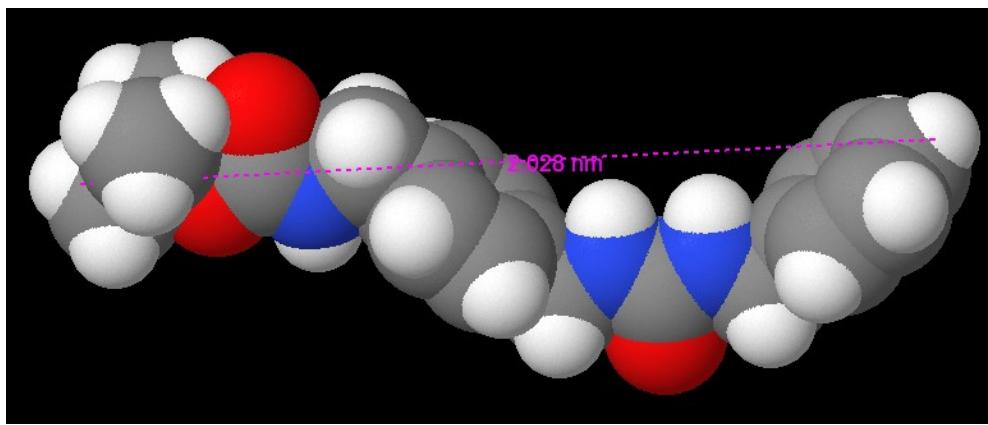
Figure S3. Molecular length based on hypothetically extended **1**, from molview.org.

SANS Section

The scattering length density (SLD) of chloroform-D was calculated from its density and molecular formula using NIST's Neutron Activation and Scattering Calculator (https://www.ncnr.nist.gov/resources/activation/). The SLD of **1** was determined using the Contrast module of MULCh, available from the University of Sydney (https://smb-research.smb.usyd.edu.au/NCVWeb/index.jsp), which also calculated the density. Three hydrogens of **1** were assumed to be exchangeable for the contrast calculation. The molecular formulas, densities, and SLDs of **1** and chloroform-D are shown in the table below. A graph depicting the fractal dimension as a function of time, indicating the degree of self-similarity over long length scales, for the nonsonicated and sonicated gels is shown in Figure S4.

Table S1: Molecular formulas, densities, and SLDs of **1** and the solvent.

| Compound	Molecular Formula	Density (g/mL)	SLD (Å^{-2})
1	$C_{21}O_3N_3H_{27}$	0.846	1.62e-6
Chloroform-D	$CDCl_3$	1.5	3.16e-6
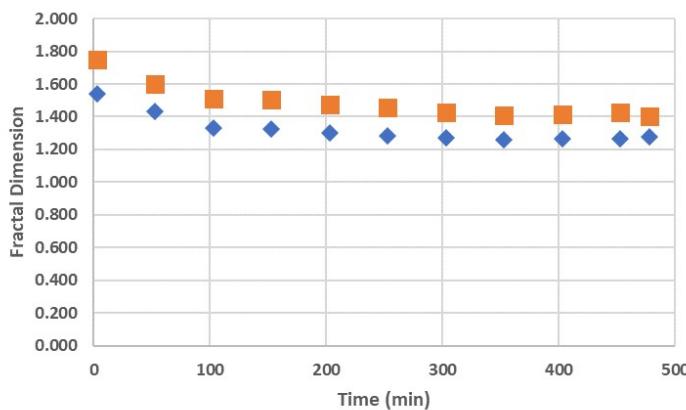
Figure S4. Fractal dimension (degree of self-similarity) of the nonsonicated (blue diamonds) and sonicated (orange squares) gels as a function of time, determined from the SANS data analysis using a smeared fractal core-shell cylinder model fit to the data.
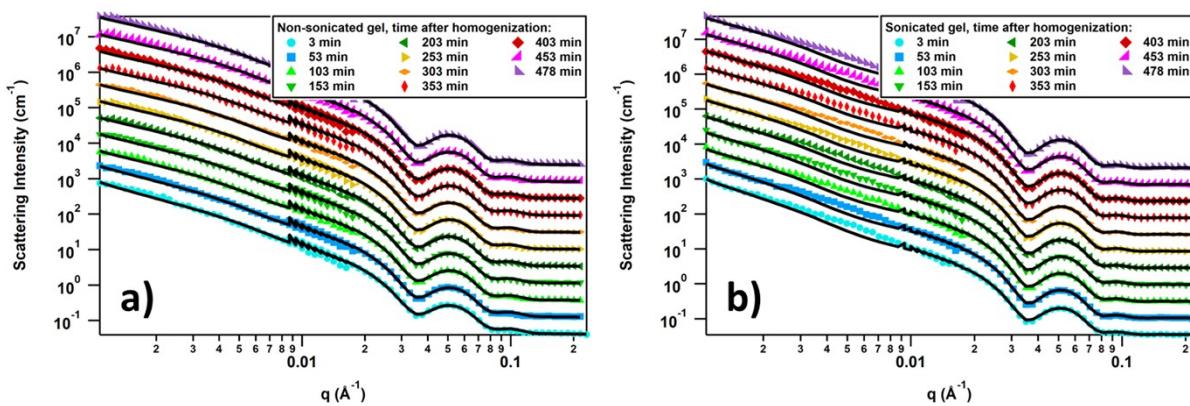

Figure S5. SANS curves of a) the nonsonicated gel and b) the sonicated gel of 1 in deuterated chloroform at a concentration of 2 w/v% showing the fits to the data over the full q-range using a fractal core-shell cylinder model (solid black lines). All curves are offset by powers of 3 for visual clarity. The oscillations in the fit curve in the overlap region (0.009 Å-1 < q < 0.017 Å-1) are caused by the model being smeared, resulting in the model alternating between fitting the low q points and the high q points.

Non-sonicated Gel (3 min after homogenization)—Smeared Fractal Core-Shell Cylinder
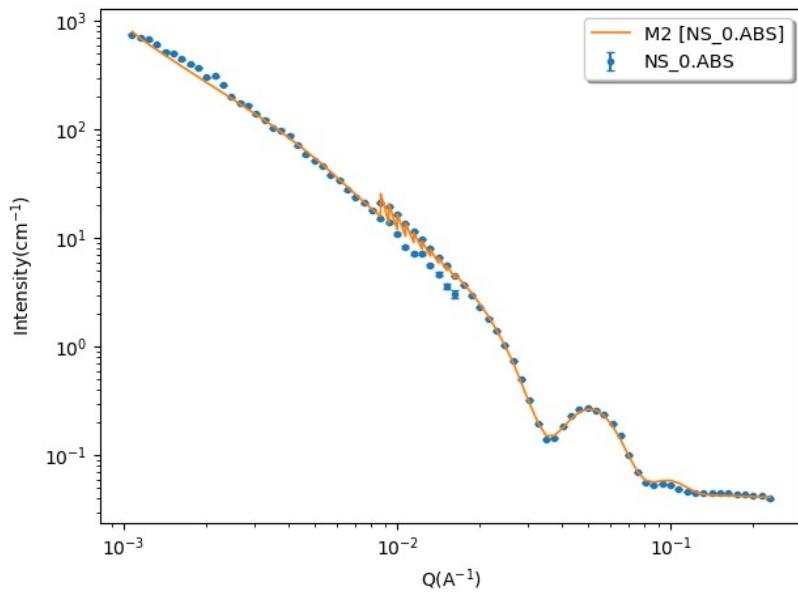
Volume Fraction (scale)	0.0163	±	2.92E-05
Background (cm^{-1})	0.040877	±	2.93E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	49.449	±	0.018798
Thickness (Å)	33.446	±	0.071014
Thickness Polydispersity	0.35202	±	0.0018475
Length (Å)	880.25	±	1.2096
Fractal Dimension	1.5475	±	0.0021275
Fitting Range	0.001072	$< q <$	0.231

Non-sonicated Gel (53 min after homogenization)—Smeared Fractal Core-Shell Cylinder
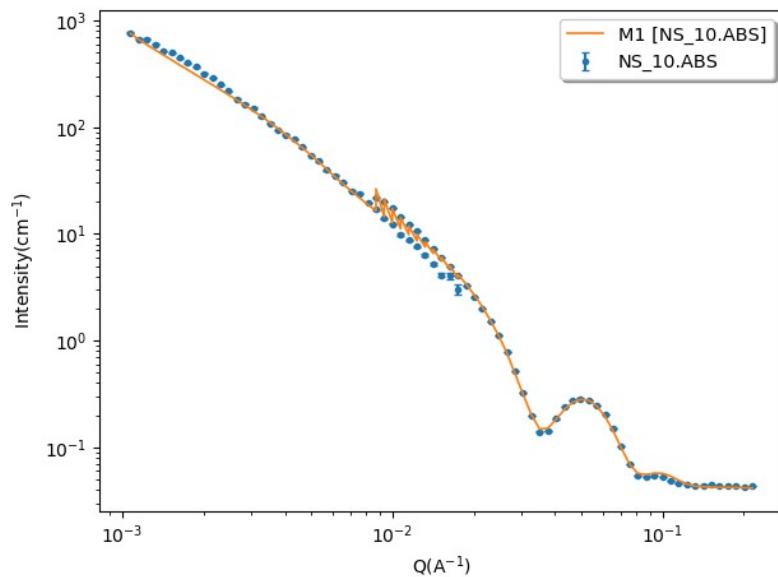
Volume Fraction (scale)	0.015612	±	1.95E-05
Background (cm^{-1})	0.041303	±	2.76E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	47.706	±	0.017626
Thickness (Å)	38.534	±	0.052674
Thickness Polydispersity	0.26189	±	0.0011786
Length (Å)	1029.7	±	1.5199
Fractal Dimension	1.4338	±	0.0022488
Fitting Range	0.001072	$< q <$	0.2154

Non-sonicated Gel (103 min after homogenization)—Smeared Fractal Core-Shell Cylinder
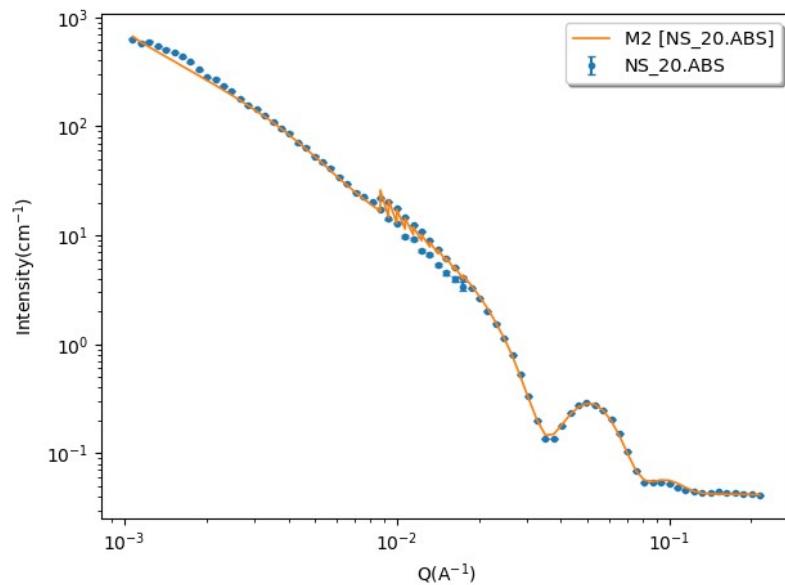
Volume Fraction (scale)	0.01555	±	1.76E-05
Background (cm^{-1})	0.041165	±	2.71E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	47.149	±	0.016934
Thickness (Å)	39.826	±	0.048796
Thickness Polydispersity	0.24069	±	0.0010637
Length (Å)	1116.2	±	1.7413
Fractal Dimension	1.3308	±	0.0024375
Fitting Range	0.001072	$< q <$	0.2154

Non-sonicated Gel (153 min after homogenization)—Smeared Fractal Core-Shell Cylinder
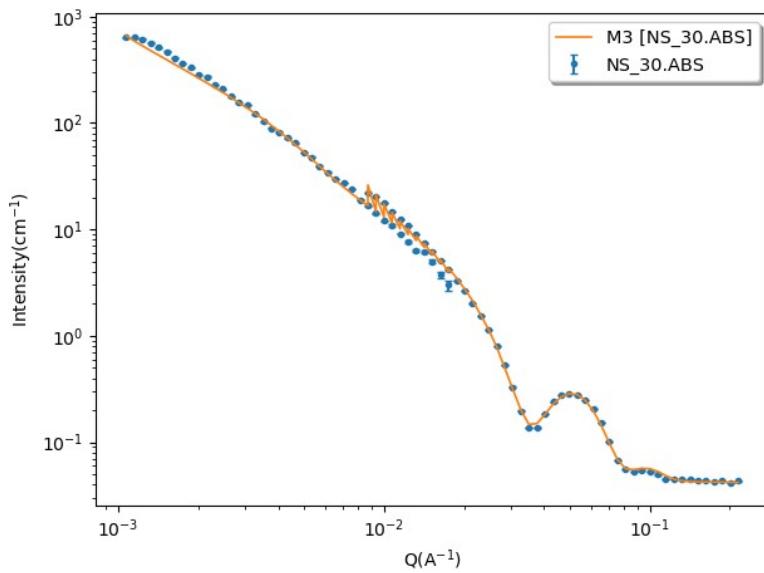
Volume Fraction (scale)	0.015465	±	1.72E-05
Background (cm^{-1})	0.041428	±	2.70E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	47.047	±	0.017005
Thickness (Å)	40.158	±	0.048389
Thickness Polydispersity	0.23731	±	0.0010445
Length (Å)	1113.1	±	1.7362
Fractal Dimension	1.3241	±	0.0024344
Fitting Range	0.001072	$< q <$	0.2154

**Non-sonicated Gel (203 min after homogenization)—Smeared Fractal Core-Shell Cylinder**
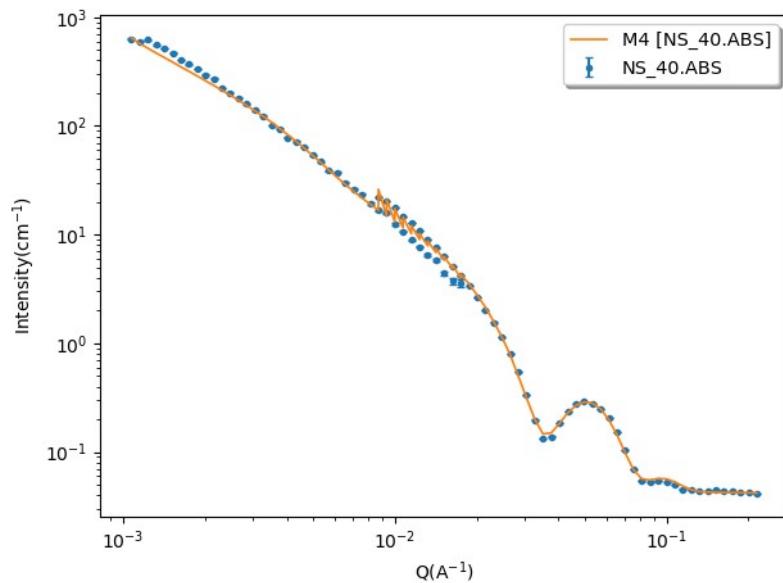
Volume Fraction (scale)	0.015422	±	1.65E-05
Background (cm^{-1})	0.041502	±	2.70E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	47.009	±	0.016561
Thickness (Å)	40.495	±	0.046174
Thickness Polydispersity	0.22945	±	0.0010156
Length (Å)	1121.2	±	1.7749
Fractal Dimension	1.3014	±	0.002477
Fitting Range	0.001072	$< q <$	0.2154

**Non-sonicated Gel (253 min after homogenization)—Smeared Fractal Core-Shell Cylinder**
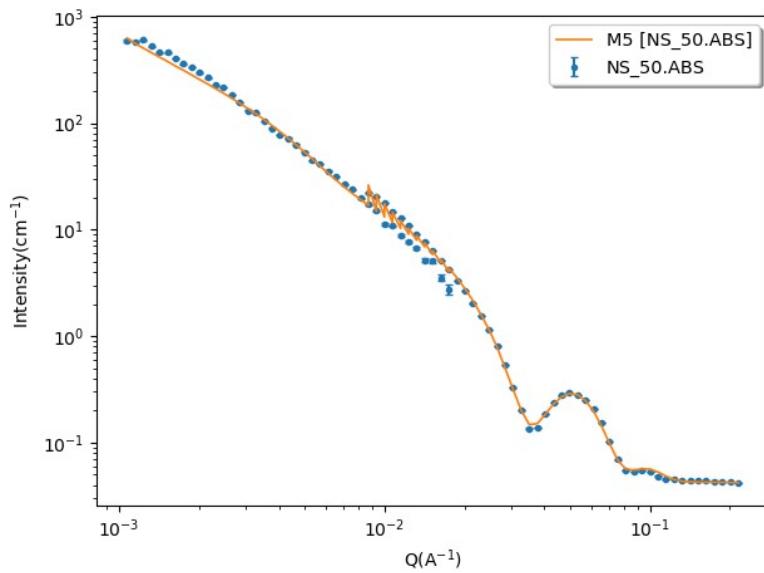
Volume Fraction (scale)	0.015391	±	1.65E-05
Background (cm^{-1})	0.041562	±	2.69E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	46.909	±	0.01677
Thickness (Å)	40.603	±	0.047062
Thickness Polydispersity	0.23298	±	0.0010179
Length (Å)	1116.7	±	1.7653
Fractal Dimension	1.2841	±	0.0024847
Fitting Range	0.001072	$< q <$	0.2154

Non-sonicated Gel (303 min after homogenization)—Smeared Fractal Core-Shell Cylinder
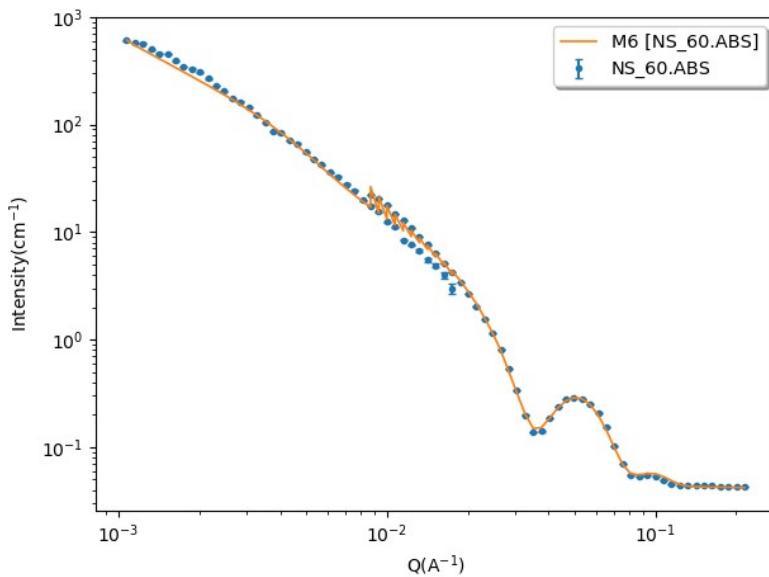
Volume Fraction (scale)	0.015512	±	1.70E-05
Background (cm⁻¹)	0.041485	±	2.71E-05
Core SLD (Å⁻²)	3.16E-06	±	0
Shell SLD (Å⁻²)	1.62E-06	±	0
Solvent SLD (Å⁻²)	3.16E-06	±	0
Radius (Å)	47.006	±	0.017314
Thickness (Å)	40.272	±	0.047951
Thickness Polydispersity	0.24005	±	0.0010264
Length (Å)	1100.5	±	1.8232
Fractal Dimension	1.2696	±	0.0024814
Fitting Range	0.001072	$< q <$	0.2154

**Non-sonicated Gel (353 min after homogenization)—Smeared Fractal Core-Shell Cylinder**
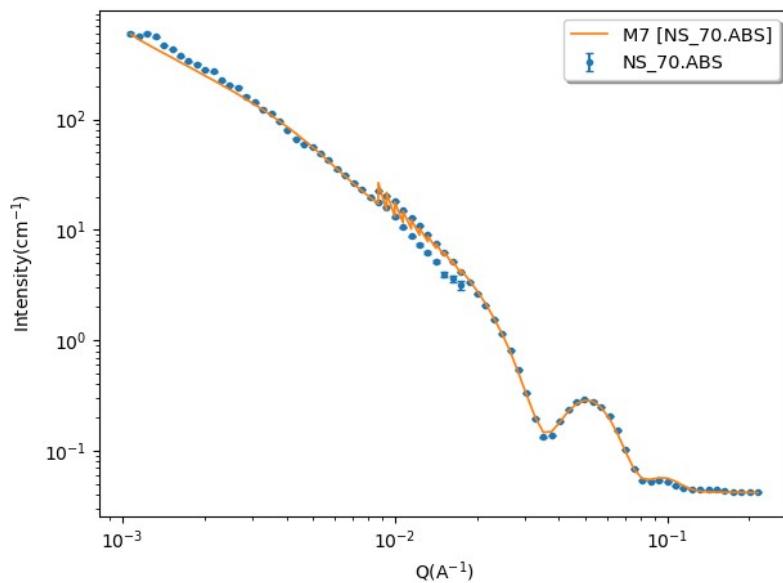
Volume Fraction (scale)	0.015382	±	1.66E-05
Background (cm^{-1})	0.041489	±	2.70E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	47.178	±	0.01708
Thickness (Å)	40.372	±	0.046118
Thickness Polydispersity	0.22454	±	0.0010186
Length (Å)	1031	±	1.5316
Fractal Dimension	1.2612	±	0.0024279
Fitting Range	0.001072	$< q <$	0.2154

Non-sonicated Gel (403 min after homogenization)—Smeared Fractal Core-Shell Cylinder
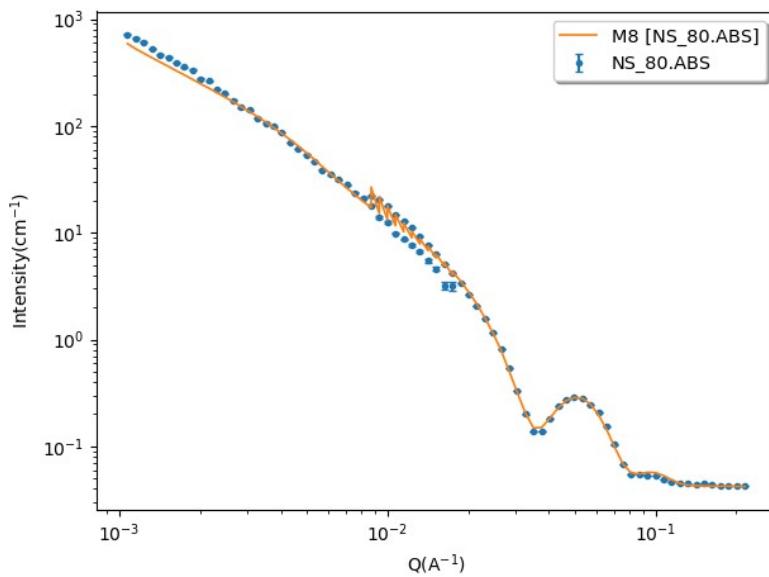
Volume Fraction (scale)	0.015391	±	1.67E-05
Background (cm^{-1})	0.041542	±	2.70E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	47.144	±	0.017151
Thickness (Å)	40.314	±	0.046713
Thickness Polydispersity	0.22879	±	0.001026
Length (Å)	1029.5	±	1.5412
Fractal Dimension	1.2643	±	0.0024316
Fitting Range	0.001072	$< q <$	0.2154

**Non-sonicated Gel (453 min after homogenization)—Smeared Fractal Core-Shell Cylinder**
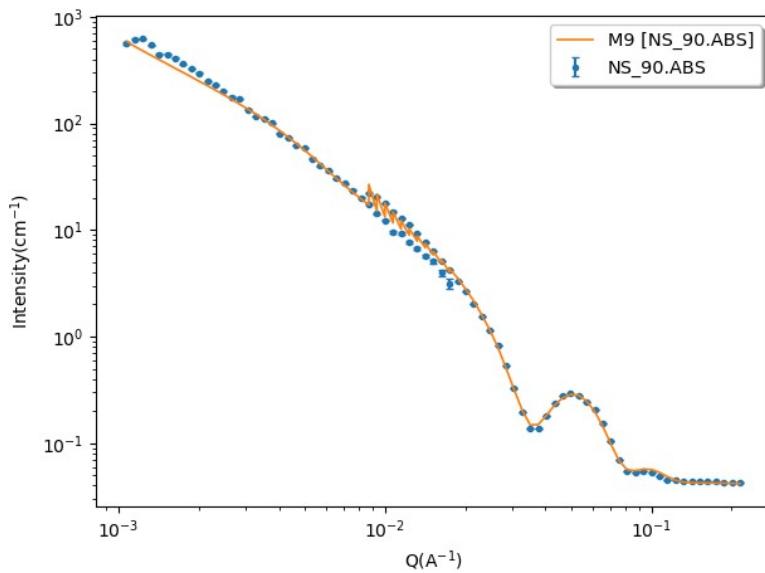
Volume Fraction (scale)	0.015438	±	1.70E-05
Background (cm^{-1})	0.041443	±	2.70E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	47.163	±	0.0172
Thickness (Å)	40.131	±	0.047053
Thickness Polydispersity	0.23019	±	0.0010343
Length (Å)	1028.9	±	1.5451
Fractal Dimension	1.2645	±	0.0024321
Fitting Range	0.001072	$< q <$	0.2154

Non-sonicated Gel (478 min after homogenization)—Smeared Fractal Core-Shell Cylinder
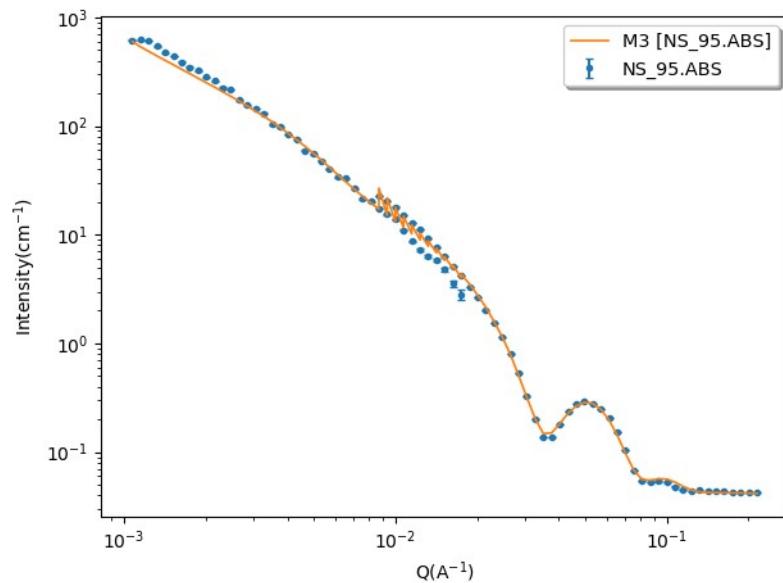
Volume Fraction (scale)	0.015384	±	1.68E-05
Background (cm^{-1})	0.041311	±	2.70E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	47.141	±	0.017215
Thickness (Å)	40.247	±	0.047032
Thickness Polydispersity	0.23014	±	0.0010294
Length (Å)	1026.3	±	1.5496
Fractal Dimension	1.2749	±	0.0024069
Fitting Range	0.001072	$< q <$	0.2154

Sonicated Gel (3 min after homogenization)— Smeared Fractal Core-Shell Cylinder
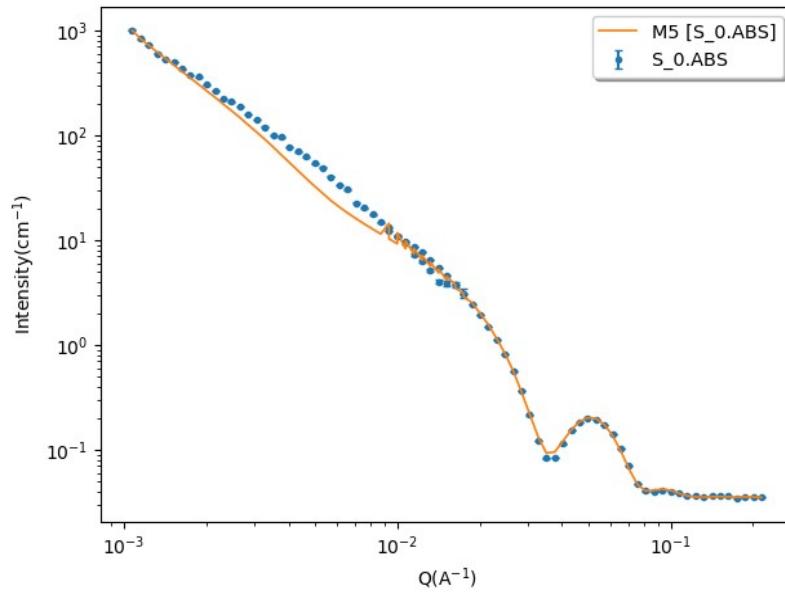
Volume Fraction (scale)	0.0091526	±	7.57E-06
Background (cm^{-1})	0.034888	±	2.25E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	43.253	±	0.019331
Thickness (Å)	49.357	±	0.039112
Thickness Polydispersity	0.091155	±	0.0012627
Length (Å)	1391.3	±	3.4947
Fractal Dimension	1.7541	±	0.0028381
Fitting Range	0.001072	$< q <$	0.2154

Sonicated Gel (53 min after homogenization)— Smeared Fractal Core-Shell Cylinder
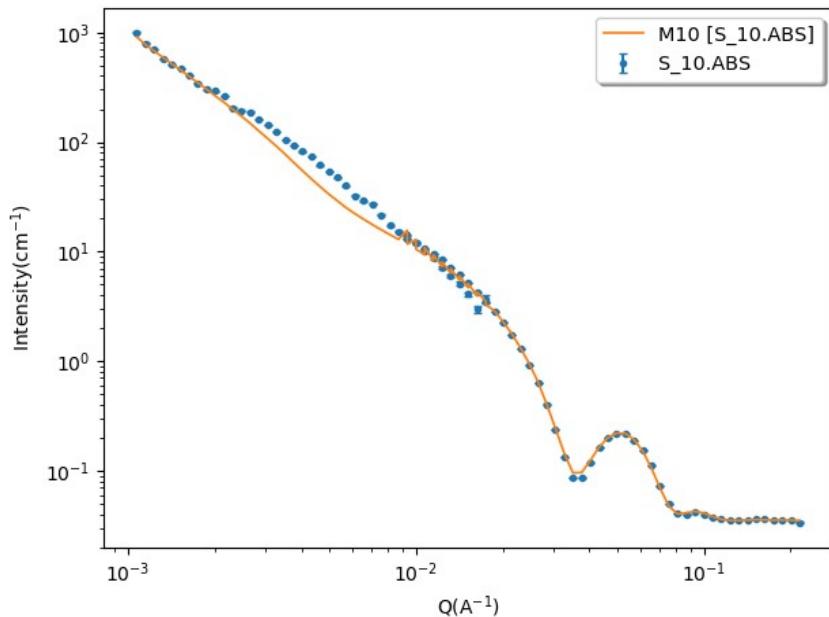
Volume Fraction (scale)	0.010202	±	7.46E-06
Background (cm^{-1})	0.034661	±	2.27E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	42.479	±	0.020146
Thickness (Å)	50.245	±	0.035216
Thickness Polydispersity	0.060529	±	0.0015919
Length (Å)	1585.4	±	4.1302
Fractal Dimension	1.6088	±	0.002997
Fitting Range	0.001072	$< q <$	0.2154

Sonicated Gel (103 min after homogenization)— Smeared Fractal Core-Shell Cylinder
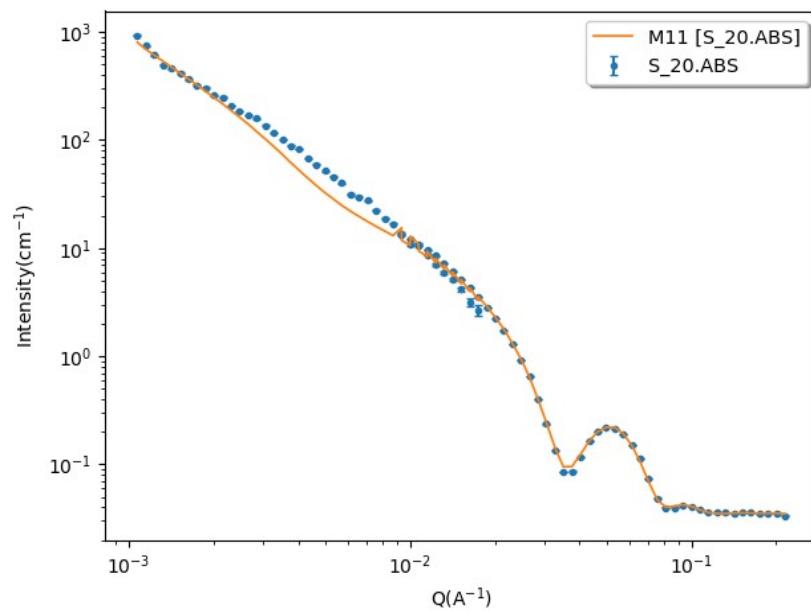
Volume Fraction (scale)	0.010225	±	7.31E-06
Background (cm^{-1})	0.034648	±	2.23E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	42.373	±	0.016559
Thickness (Å)	50.415	±	0.033041
Thickness Polydispersity	0.058014	±	0.0014928
Length (Å)	1668.7	±	4.6195
Fractal Dimension	1.5151	±	0.0031924
Fitting Range	0.001072	$< q <$	0.2154

Sonicated Gel (153 min after homogenization)— Smeared Fractal Core-Shell Cylinder
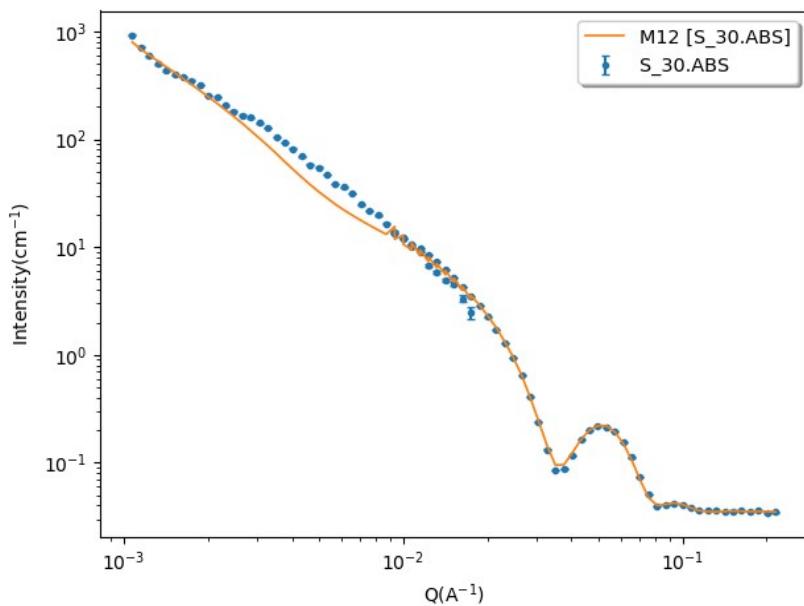
Volume Fraction (scale)	0.010275	±	7.34E-06
Background (cm^{-1})	0.034686	±	2.23E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	42.334	±	0.016494
Thickness (Å)	50.362	±	0.032984
Thickness Polydispersity	0.059662	±	0.0014556
Length (Å)	1668.5	±	4.6256
Fractal Dimension	1.5077	±	0.003187
Fitting Range	0.001072	$< q <$	0.2154

**Sonicated Gel (203 min after homogenization)— Smeared Fractal Core-Shell Cylinder**
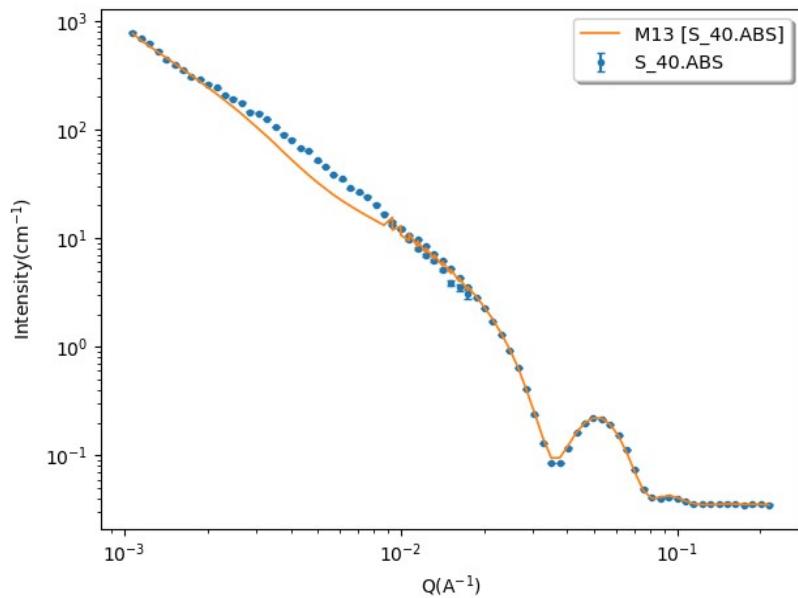
Volume Fraction (scale)	0.01026	±	7.20E-06
Background (cm^{-1})	0.034939	±	2.23E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	42.297	±	0.017078
Thickness (Å)	50.515	±	0.033174
Thickness Polydispersity	0.05114	±	0.0016694
Length (Å)	1673.7	±	4.7483
Fractal Dimension	1.4752	±	0.0032348
Fitting Range	0.001072	$< q <$	0.2154

Sonicated Gel (253 min after homogenization)— Smeared Fractal Core-Shell Cylinder
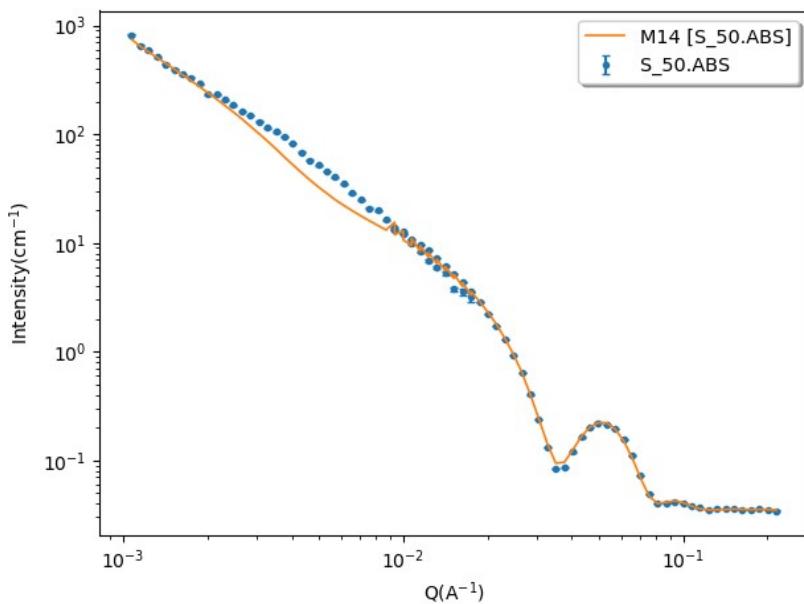
Volume Fraction (scale)	0.010285	±	7.20E-06
Background (cm^{-1})	0.034727	±	2.23E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	42.48	±	0.017546
Thickness (Å)	50.459	±	0.033381
Thickness Polydispersity	0.049631	±	0.0017297
Length (Å)	1677.2	±	4.8164
Fractal Dimension	1.4578	±	0.0032725
Fitting Range	0.001072	$< q <$	0.2154

Sonicated Gel (303 min after homogenization)— Smeared Fractal Core-Shell Cylinder
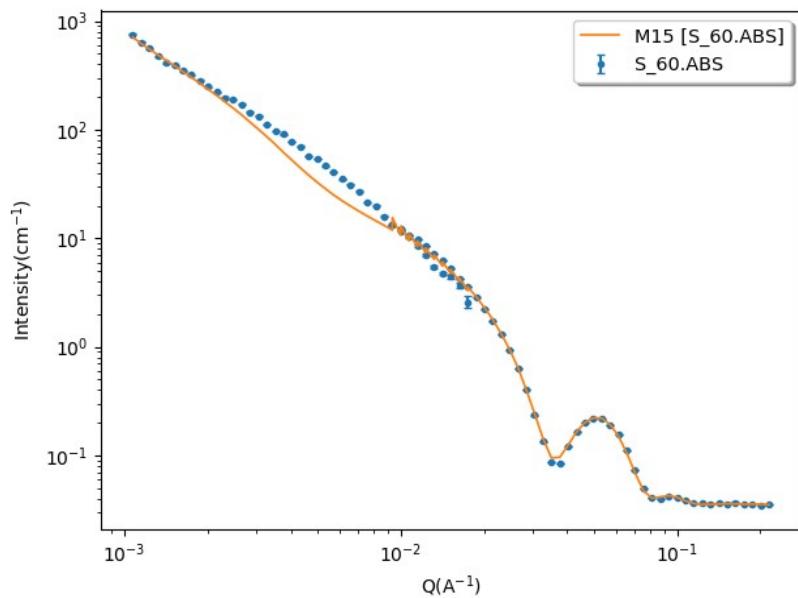
Volume Fraction (scale)	0.010245	±	7.35E-06
Background (cm^{-1})	0.034981	±	2.24E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	42.399	±	0.016409
Thickness (Å)	50.503	±	0.032807
Thickness Polydispersity	0.055041	±	0.0015583
Length (Å)	1666.4	±	4.694
Fractal Dimension	1.4289	±	0.0032894
Fitting Range	0.001072	$< q <$	0.2154

**Sonicated Gel (353 min after homogenization)— Smeared Fractal Core-Shell Cylinder**
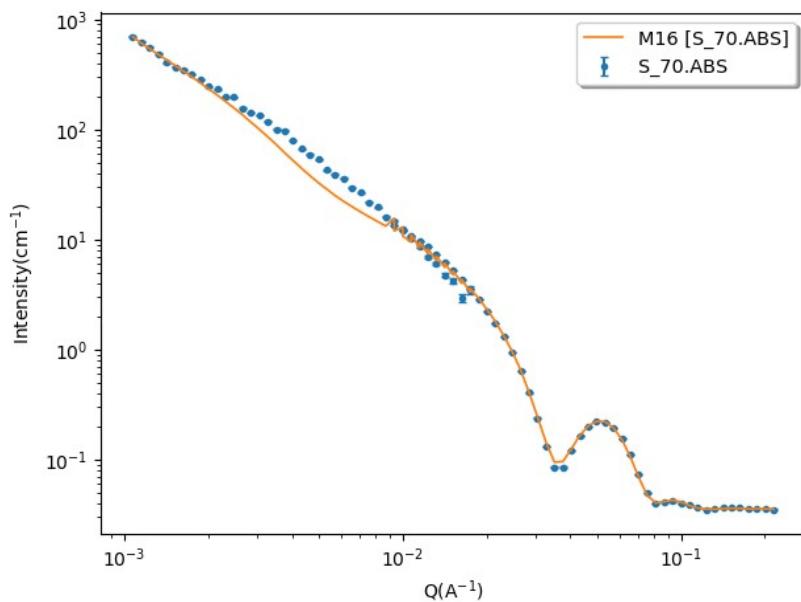
Volume Fraction (scale)	0.010238	±	7.11E-06
Background (cm^{-1})	0.034931	±	2.23E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	42.302	±	0.018695
Thickness (Å)	50.688	±	0.033681
Thickness Polydispersity	0.045606	±	0.0019032
Length (Å)	1684.5	±	4.9918
Fractal Dimension	1.4105	±	0.0033411
Fitting Range	0.001072	< q <	0.2154

Sonicated Gel (403 min after homogenization)— Smeared Fractal Core-Shell Cylinder
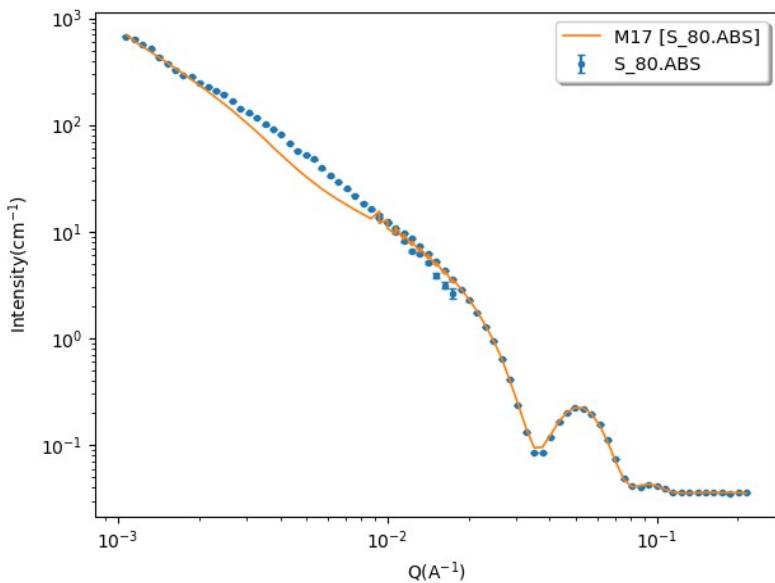
Volume Fraction (scale)	0.010299	±	7.20E-06
Background (cm^{-1})	0.035029	±	2.23E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	42.483	±	0.017188
Thickness (Å)	50.439	±	0.032972
Thickness Polydispersity	0.034947	±	0.0023811
Length (Å)	1674.5	±	4.8191
Fractal Dimension	1.4153	±	0.0033243
Fitting Range	0.001072	$< q <$	0.2154

**Sonicated Gel (453 min after homogenization)— Smeared Fractal Core-Shell Cylinder**
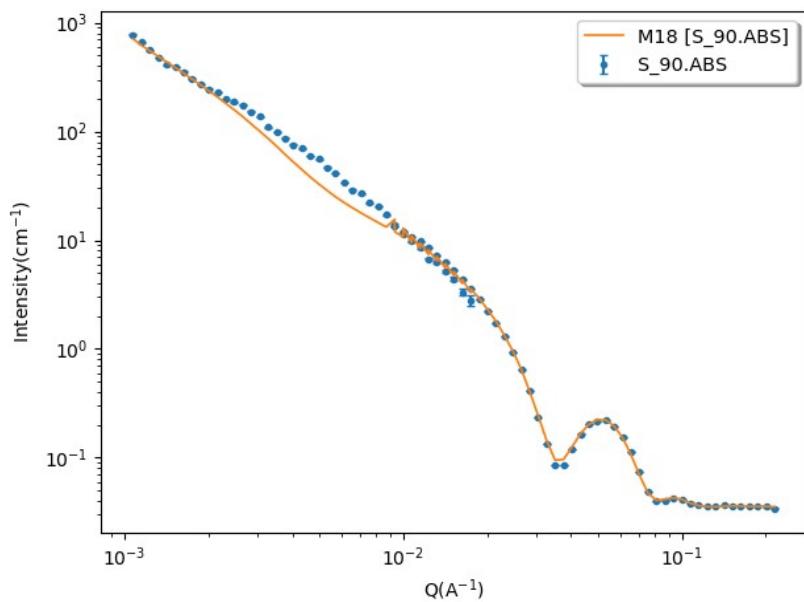
Volume Fraction (scale)	0.010288	±	7.19E-06
Background (cm^{-1})	0.034909	±	2.23E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	42.44	±	0.017675
Thickness (Å)	50.492	±	0.033431
Thickness Polydispersity	0.049002	±	0.0017507
Length (Å)	1677.9	±	4.8712
Fractal Dimension	1.4259	±	0.0033087
Fitting Range	0.001072	$< q <$	0.2154

Sonicated Gel (478 min after homogenization)— Smeared Fractal Core-Shell Cylinder
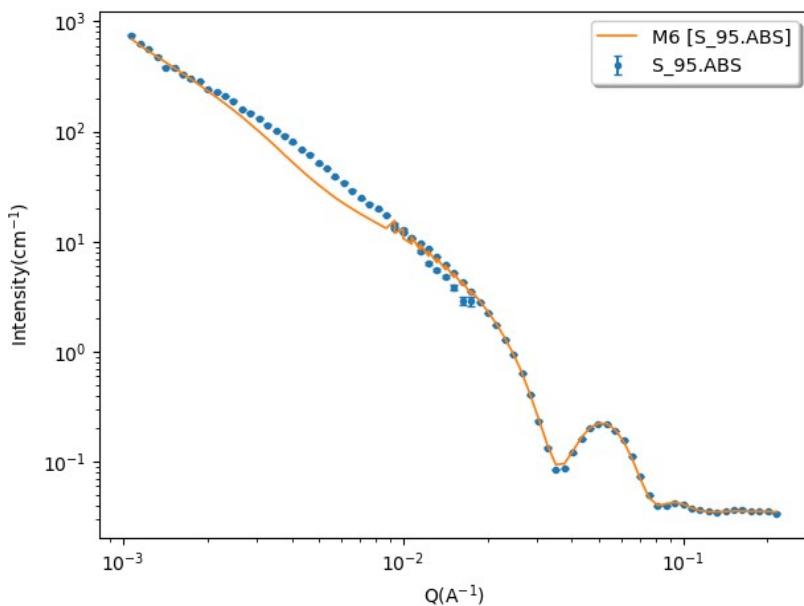
Volume Fraction (scale)	0.010346	±	7.27E-06
Background (cm^{-1})	0.034868	±	2.23E-05
Core SLD (Å^{-2})	3.16E-06	±	0
Shell SLD (Å^{-2})	1.62E-06	±	0
Solvent SLD (Å^{-2})	3.16E-06	±	0
Radius (Å)	42.562	±	0.017907
Thickness (Å)	50.288	±	0.033507
Thickness Polydispersity	0.044799	±	0.0019206
Length (Å)	1679.7	±	4.9258
Fractal Dimension	1.4045	±	0.0033337
Fitting Range	0.001072	< q <	0.2154