SUPPLEMENTARY INFORMATION

Using small angle neutron scattering to explore porosity, connectivity and accessibility, towards optimised hierarchical solid acid catalysts

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Physicochemical and textural characterisation

Powder XRD

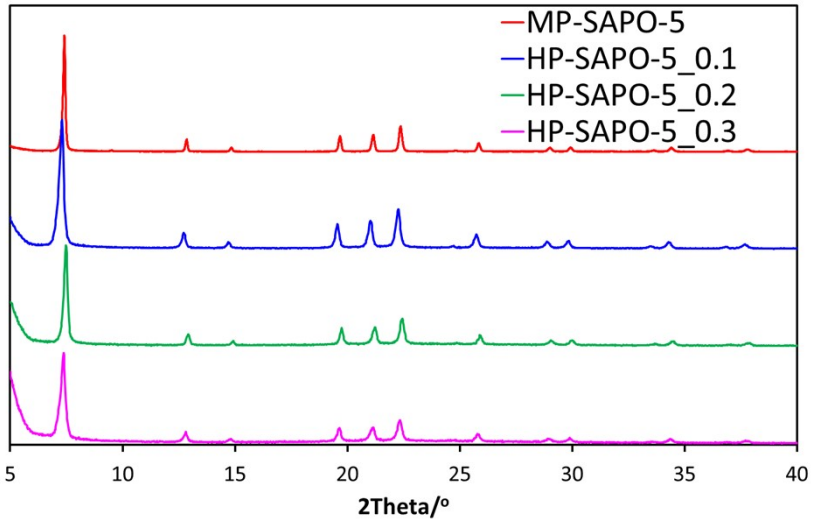


Figure S1: Powder X-ray diffraction pattern of the four SAPO-5 species, confirming phase pure AlPO-5 has been formed.

ICP

Table S1: ICP analysis of microporous and hierarchical SAPO-5.

System	Al (wt%)	P (wt%)	Si (wt%)	(mol P + mol Si) / mol Al
MP-SAPO-5	21.9	24.1	1.4	1.02
HP-SAPO-5_0.1	16.1	17.8	6.1	1.33
HP-SAPO-5_0.2	19.5	15.5	9.8	1.18
HP-SAPO-5_0.3	19.9	15.0	11.2	1.20

SEM images

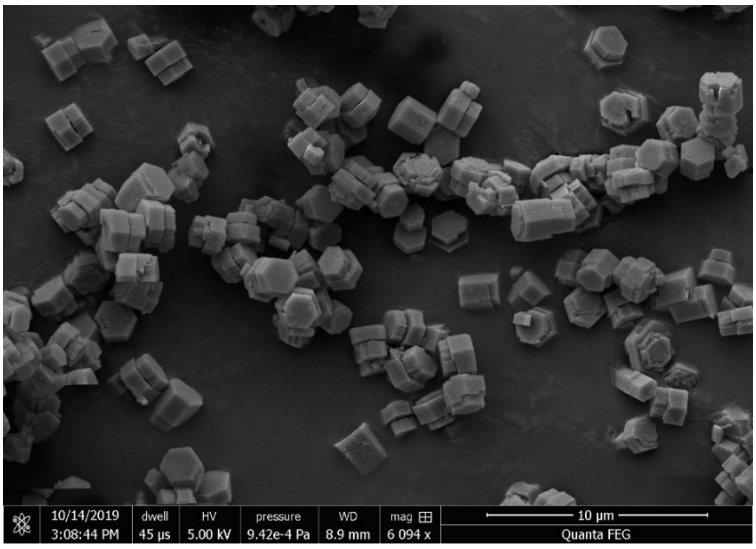


Figure S2: SEM image showing the crystalline nature of MP-SAPO-5.

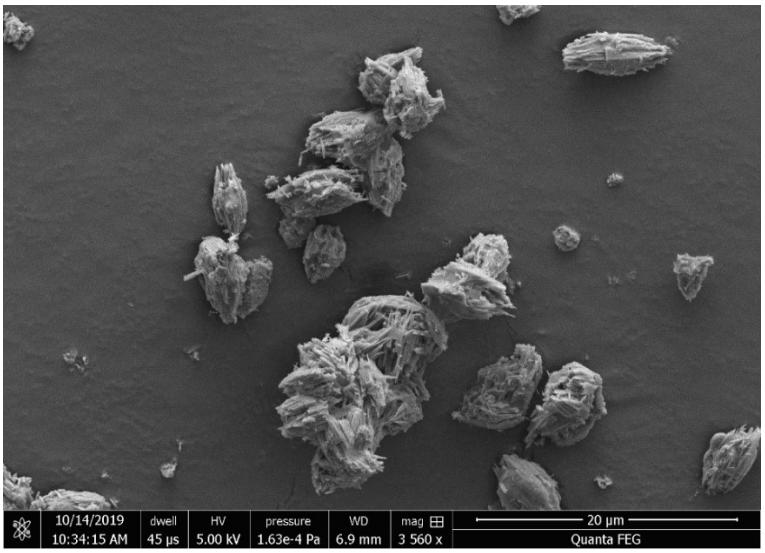


Figure S3: SEM image showing the rough non-uniform surface of HP-SAPO-5_0.1.

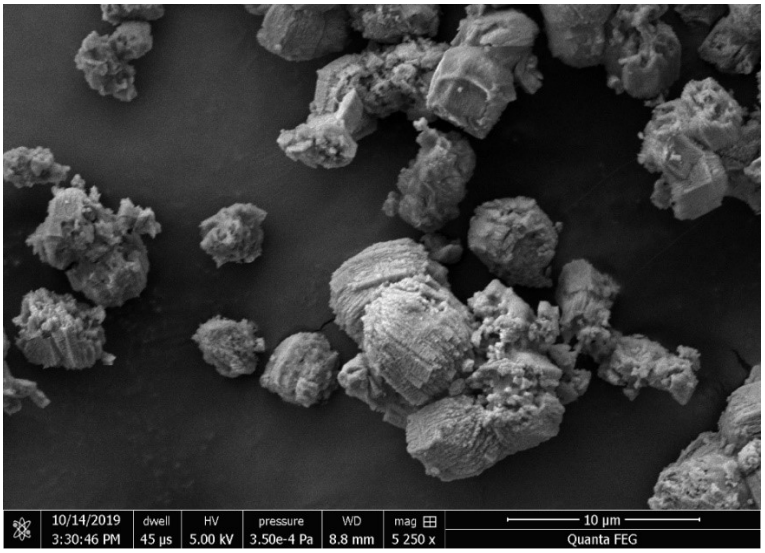


Figure S4: SEM image showing the rough non-uniform surface of HP-SAPO-5_0.2.

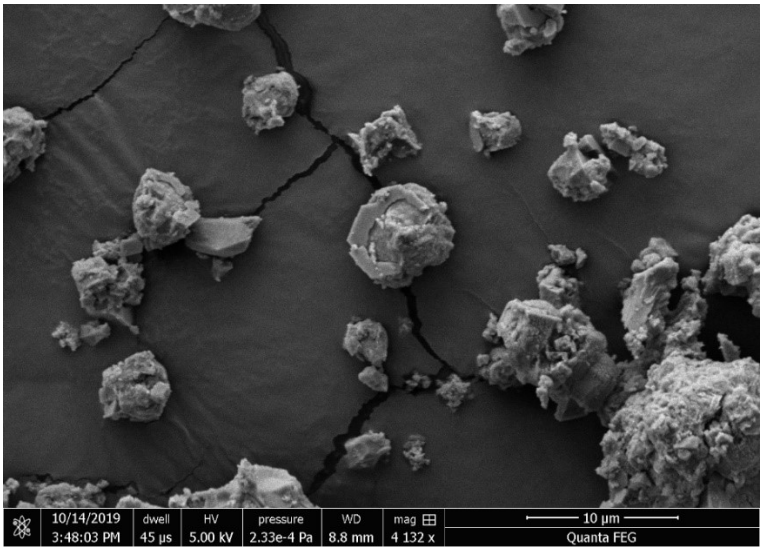
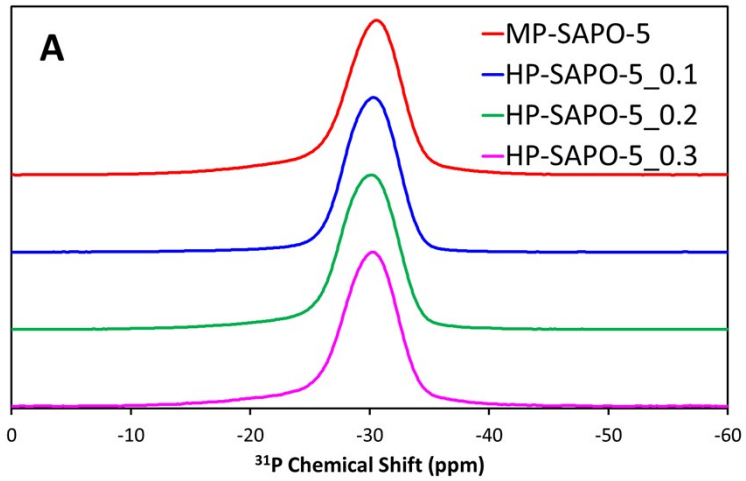
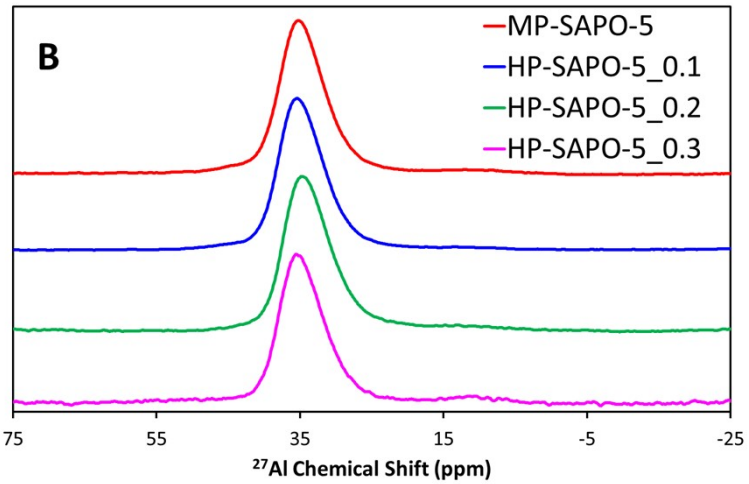


Figure S5: SEM image showing the rough non-uniform particles of HP-SAPO-5_0.3.

Solid State NMR





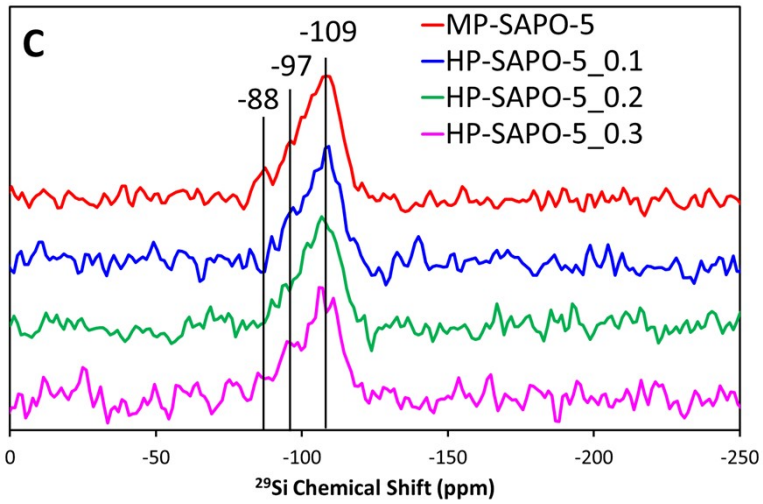


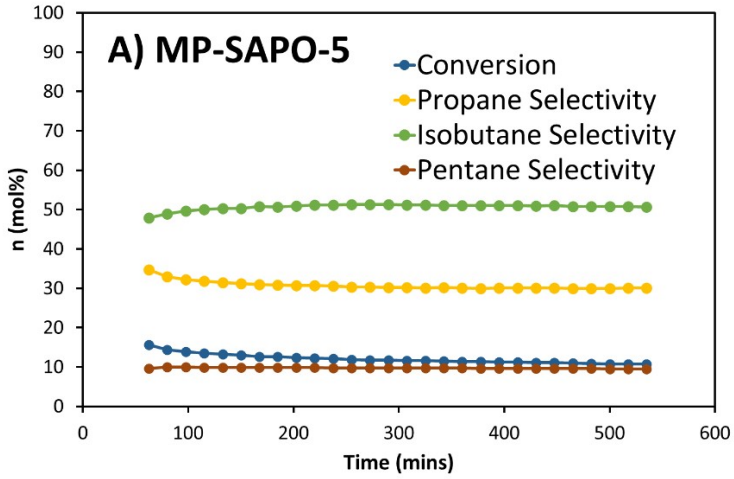
Figure S6: Showing 1D solid state NMR data of the four SAPO-5 samples, focussing on the ^{31}P (A), ^{27}Al (B) and ^{29}Si (C) nuclei. Further details can be found in the main text in the experimental methods.

NH_3-TPD data

Table S2: Summary of the NH_3-TPD data characterising acid site quantity.

Sample	Chemisorbed NH_3 (µmol/g_{sample})	Desorption Maxima (°C)
MP-SAPO-5	188	375
HP-SAPO-5_0.1	258	389
HP-SAPO-5_0.2	326	384
HP-SAPO-5_0.3	245	369

n-Butane isomerisation catalysis data



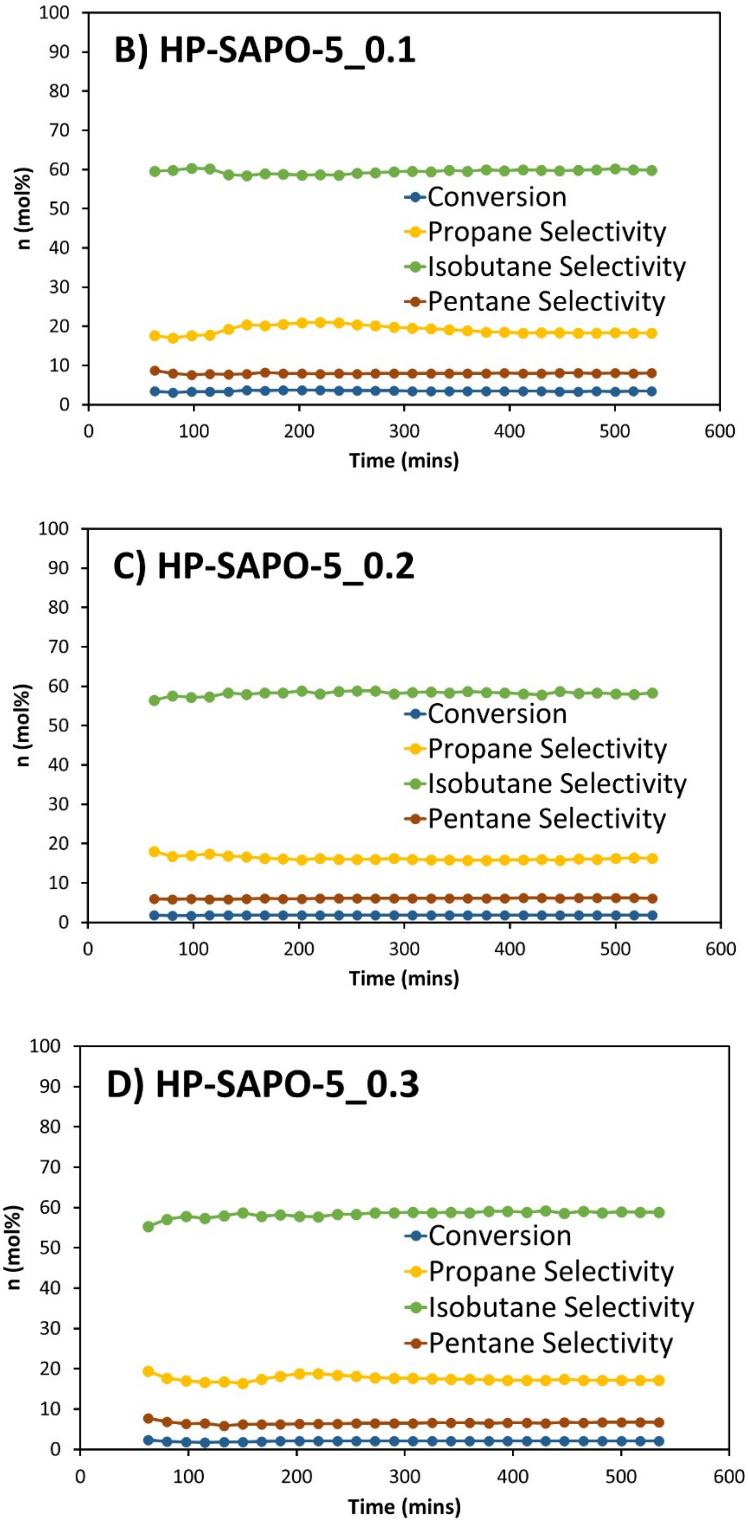


Figure S7: On-stream catalytic data showing the activity of the four SAPO-5 systems (A-D) for the n-butane isomerisation reaction. Reaction conditions: 5 ml/min flow of 10% n-butane in nitrogen at 300 °C, 0.3 g of catalyst.

Ethanol dehydration catalysis data

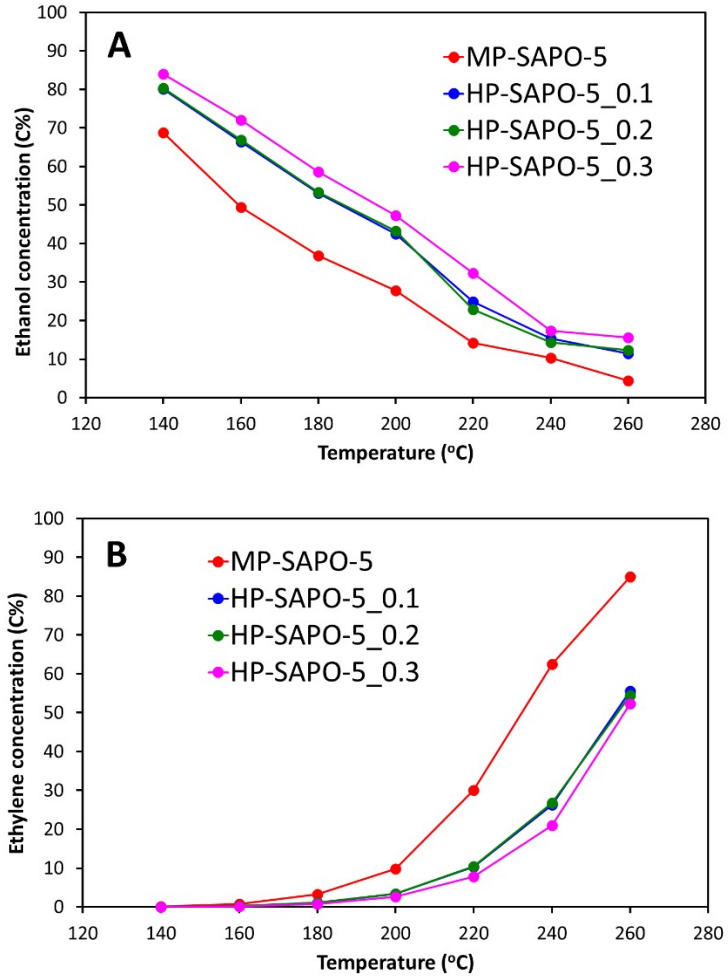


Figure S8: Showing the ethanol dehydration data with the C% of Ethanol (A) and Ethylene (B) in the output stream. Reaction conditions: 25 mL/min N_2 carrier gas, 0.3 g of catalyst, 10 µL/min flow of 10 vol% heptane in ethanol.

Small angle neutron scattering model fitting for MP-SAPO-5

Table S3: Full fitting parameters and values, with associated uncertainties, achieved with different models on MP-SAPO-5, towards optimal fitting.

Parameter	2G+P[a]	2G+F[b]	2G+F PD[c]	2G+P+F[d]	2G+P+F PD[e]
Goodness of fit					
Reduced χ^2	407.6	190.1	123.5	6.912	4.724
General parameters					
Overall Scale (Fixed)	1	1	1	1	1
Background, cm^{-1}	0.021 ± 1.5x10^{-4}	-0.074 ± 0.044	-0.114 ± 0.064	0.016 ± 1.7x10^{-4}	0.016 ± 1.8x10^{-4}
1st Gaussian peak, Bragg peak, 100					
Scale, **A**	0.146	0.144	0.153	0.143	0.142

(> 0)	± 0.001	± 0.001	± 0.001	± 0.001	± 0.001
Position, $Å^{-1}$ (0.50 to 0.55)	0.524 ± $1.5x10^{-4}$	0.522 ± 0.002	0.523 ± $1.5x10^{-4}$	0.524 ± $1.6x10^{-4}$	0.524 ± $1.6x10^{4}$
Width[f], $Å^{-1}$	0.021 ± $1.9x10^{-4}$	0.026 ± $2.1x10^{-4}$	0.023 ± $1.9x10^{-4}$	0.023 ± $1.9x10^{-4}$	0.023 ± $1.9x10^{-4}$
2nd Gaussian peak, Bragg peak, 110					
Scale, **B** (> 0)	0.037 ± 0.020	0.096 ± 0.043	0.137 ± 0.064	0.011 ± 0.001	0.010 ± 0.001
Position, $Å^{-1}$ (0.90 to 0.95)	0.905 ± 0.003	0.900 ± 0.008	0.900 ± 0.009	0.910 ± 0.004	0.911 ± 0.005
Width[f], $Å^{-1}$	0.003 ± 0.001	0.709 ± 0.188	0.908 ± 0.238	0.028 ± 0.005	0.032 ± 0.005
Power Law					
Scale, **C** (> 0)	$4.995x10^{-5}$ ± $2.935x10^{-7}$	-	-	$1.067x10^{-6}$ ± $1.652x10^{-9}$	$1.057x10^{-6}$ ± $1.893x10^{-9}$
Power (3 to 4)	3.189 ± 0.001	-	-	4.000 ± $5.8x10^{-9}$	4.000 ± $5.9x10^{-9}$
Fractal					
Scale, **D** (0 to 1)	-	0.024 ± 0.001	0.021 ± 0.001	0.005 ± $2.1x10^{-5}$	0.005 ± $5.0x10^{-5}$
Radius, Å	-	10.003 ± 0.062	4.680 ± 0.386	21.561 ± 0.109	14.437 ± 0.507
Fractal dimension (2 to 3)	-	2.896 ± 0.003	2.906 ± 0.001	2.676 ± 0.010	2.542 ± 0.013
Correlation length, Å	-	1080.6 ± 63.3	817.0 ± 17.0	52.9 ± 0.8	61.8 ± 1.3
SLD Block, 10^{-6} $Å^{-2}$ (Fixed)	-	0	0	0	0
SLD Solvent, 10^{-6} $Å^{-2}$ (Fixed)	-	4.1	4.1	4.1	4.1
Radius Polydispersity[g] (0 to 1)	-	-	0.521 ± 0.029	-	0.300 ± 0.013

a) 2G+P = 2Gaussian Peaks and a Power Law; $Intensity_{2G+P}$ = Background + A.$Intensity_{Gaussian1}$ + B.$Intensity_{Gaussian2}$ + C.$Intensity_{PowerLaw}$. A-D here (and in all equations associated with this table) are scalar quantities that are proportional to the volume fraction of that component in the model. Only the intensity terms are functions of Q. Links to the individual models are found at the end of the ESI.

b) 2G+F = 2Gaussian Peaks and a Fractal, with no polydispersity; $Intensity_{2G+F}$ = Background + A.$Intensity_{Gaussian1}$ + B.$Intensity_{Gaussian2}$ + D.$Intensity_{Fractal}$.

c) 2G+F PD = 2Gaussian Peaks and a Fractal, with polydispersity; $Intensity_{2G+F\ PD}$ = Background + A.$Intensity_{Gaussian1}$ + B.$Intensity_{Gaussian2}$ + D.$Intensity_{Fractal+PD}$.

d) 2G+P+F = 2Gaussian Peaks, a Power Law and a Fractal, with no polydispersity; $Intensity_{2G+P+F}$ = Background + A.$Intensity_{Gaussian1}$ + B.$Intensity_{Gaussian2}$ + C.$Intensity_{PowerLaw}$ + D.$Intensity_{Fractal}$.

e) 2G+P+F PD = 2Gaussian peaks, a Power Law and a Fractal with polydispersity; $Intensity_{2G+P+F\ PD}$ = Background + A.$Intensity_{Gaussian1}$ + B.$Intensity_{Gaussian2}$ + C.$Intensity_{PowerLaw}$ + D.$Intensity_{Fractal+PD}$.

f) One standard deviation = FWHM/2.354.

g) HWHM of Lognormal distribution at the median radius divided by the median radius.

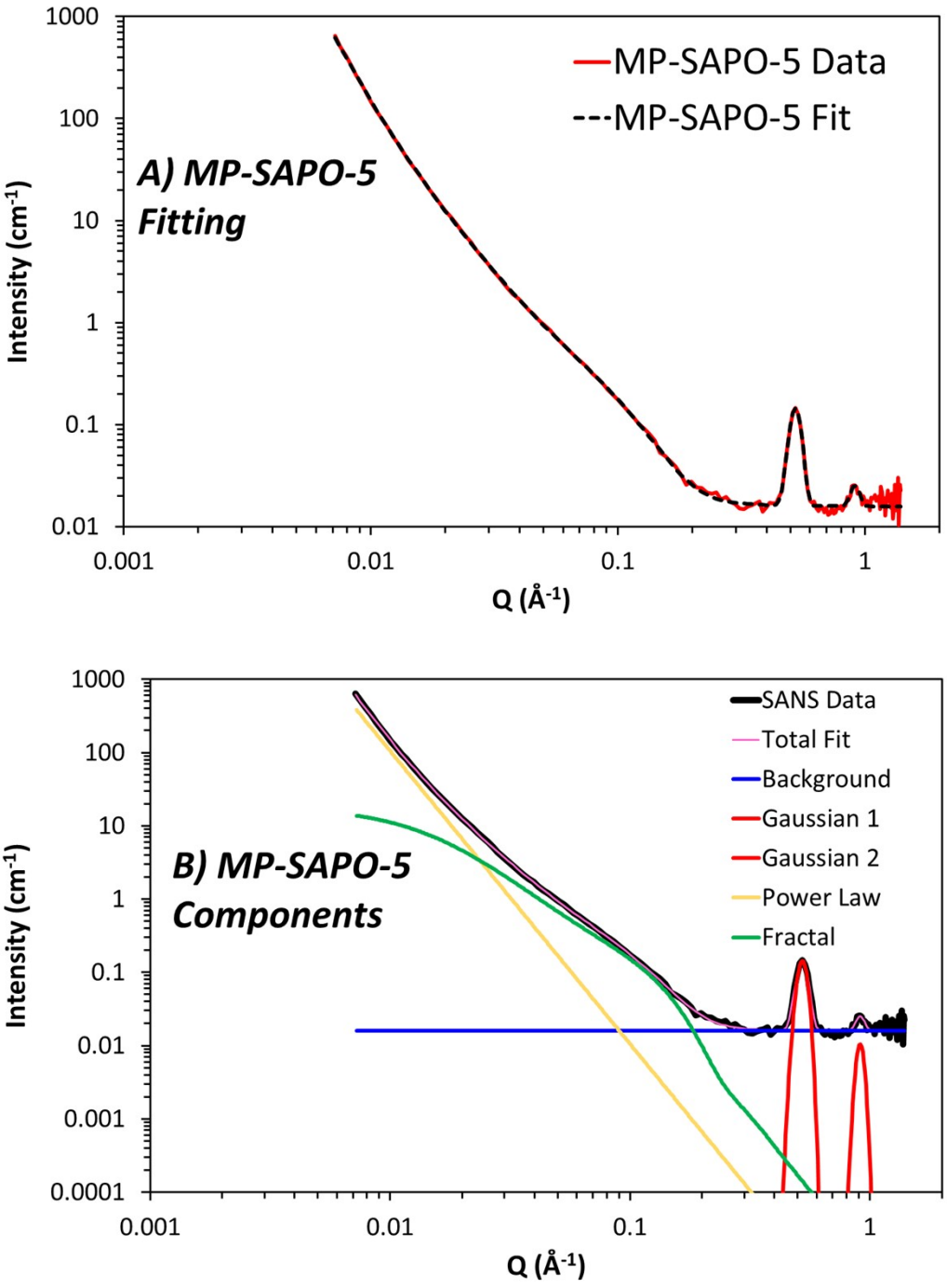


Figure S9: A) MP-SAPO-5 SANS data fitted with a 2 Gaussian peak, Power Law and Fractal aggregate (with Polydisersity) model, and B) Showing the individual contributions to the fit.

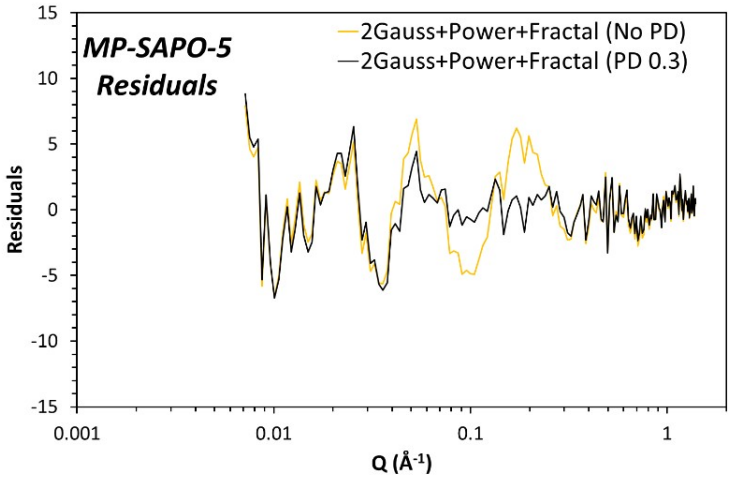


Figure S10: Residual plots after fitting the MP-SAPO-5 system with 2 Gaussian peak, Power Law and Fractal aggregate model, with and without radius polydispersity on the fractal pore size.

Small angle neutron scattering model fitting for HP-SAPO-5 systems

2 Gaussians, Power Law and Fractal

Table S4: Full fitting parameters and values, with associated uncertainties, achieved with a model including 2 Gaussians, Power Law and Fractal for the three HP-SAPO-5 systems.

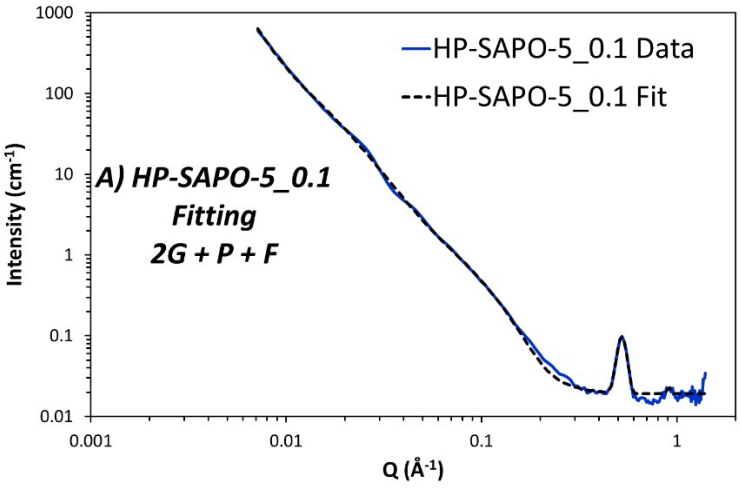
Parameter	HP-SAPO-5_0.1	HP-SAPO-5_0.2	HP-SAPO-5_0.3
Goodness of fit			
Reduced χ^2	140.8	158.7	175.6
General parameters			
Overall Scale (Fixed)	1	1	1
Background, cm^{-1}	0.019 ± $1.8x10^{-4}$	0.023 ± $1.8x10^{-4}$	0.025 ± $1.7x10^{4}$
1st Gaussian peak, Bragg peak, 100			
Scale, **A** (> 0)	0.087 ± 0.001	0.077 ± 0.001	0.067 ± 0.001
Position, $Å^{-1}$ (0.50 to 0.55)	0.523 ± $2.5x10^{-4}$	0.523 ± $2.9x10^{-4}$	0.524 ± $3.2x10^{-4}$
Width[a], $Å^{-1}$	0.023 ± $2.9x10^{-4}$	0.022 ± $3.4x10^{-4}$	0.022 ± $3.7x10^{-4}$
2nd Gaussian peak, Bragg peak, 110			
Scale, **B** (> 0)	0.0047 ± 0.003	0.005 ± 1.721	0.001 ± 43598000
Position, $Å^{-1}$ (0.90 to 0.95)	0.913 ± 0.009	0.927 ± 1.058	0.928 ± 202820000
Width[a], $Å^{-1}$	0.017 ± 0.013	0.004 ± 0.817	0.001 ± 20059000
Power law			
Scale, **C**	$9.095x10^{-7}$	$8.125x10^{-7}$	$7.955x10^{-7}$

(> 0)	± 2.3x10^{-9}	± 2.2x10^{-9}	± 2.2x10^{-9}
Power (3 to 4)	4.000 ± 6.5x10^{-9}	4.000 ± 6.4x10^{-9}	4.000 ± 6.4x10^{-9}
Fractal			
Scale, **D** (0 to 1)	0.019 ± 3.5x10^{-5}	0.022 ± 4.3x10^{-5}	0.024 ± 5.5x10^{-5}
Radius, Å	14.902 ± 0.049	13.905 ± 0.048	12.284 ± 0.047
Fractal dimension (2 to 3)	2.969 ± 0.003	2.931 ± 0.002	2.872 ± 0.002
Correlation length, Å	60.088 ± 0.181	59.412 ± 0.171	61.159 ± 0.180
SLD Block, 10^{-6} Å^{-2} (Fixed)	0	0	0
SLD Solvent, 10^{-6} Å^{-2} (Fixed)	4.1	4.1	4.1
Radius Polydispersity[b] (Fixed)	0.3	0.3	0.3

a) One standard deviation = FWHM/2.354

b) HWHM of Lognormal distribution at the median radius divided by the median radius.

The model here is described by 2Gaussian peaks, a Power Law and a Fractal with polydispersity; $Intensity_{2G+P+F\ PD}$ = Background + A.$Intensity_{Gaussian1}$ + B.$Intensity_{Gaussian2}$ + C.$Intensity_{PowerLaw}$ + D.$Intensity_{Fractal+PD}$. A-D here are scalar quantities that are proportional to the volume fraction of that component in the model. Only the intensity terms are functions of Q. Links to the individual models are found at the end of the ESI.



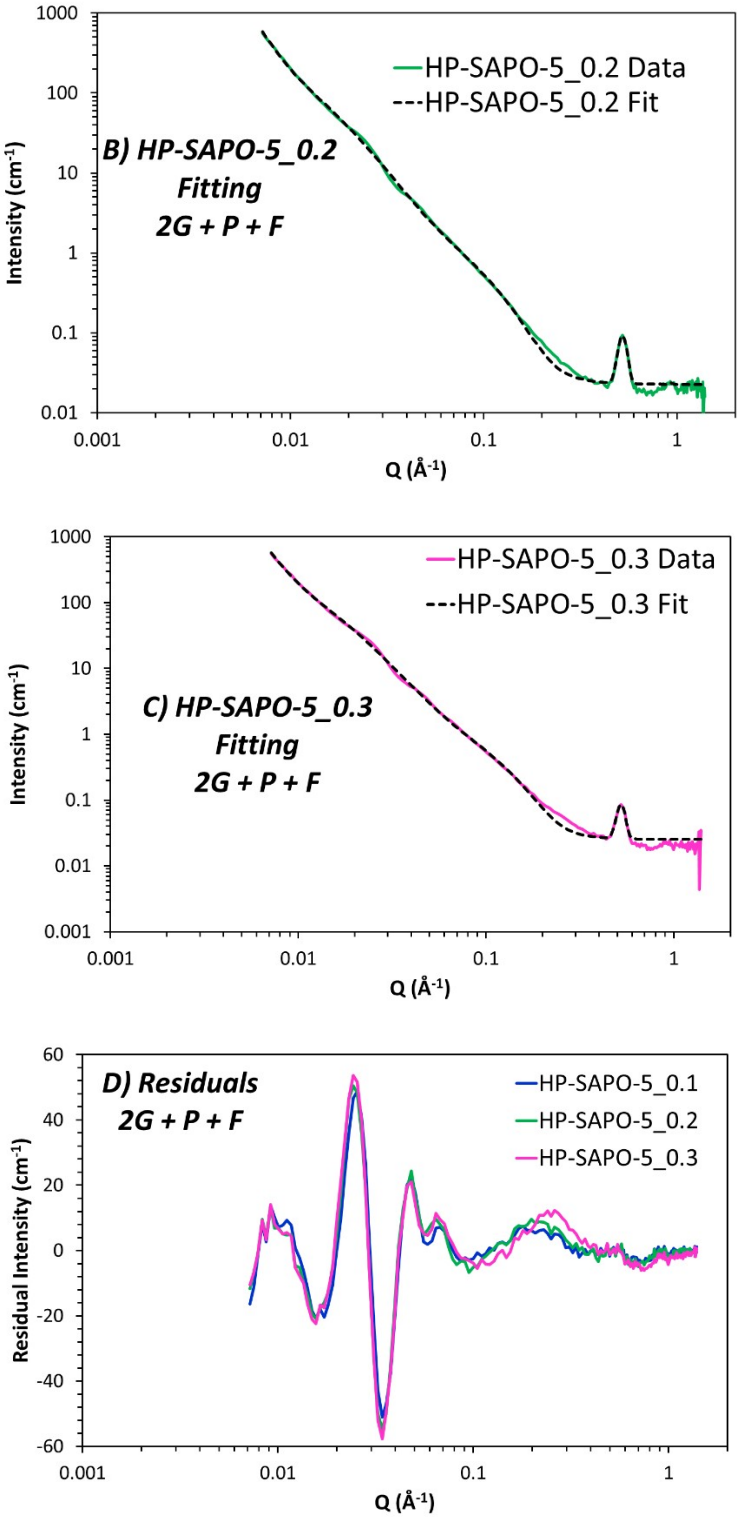


Figure S11: Showing SANS fits (A-C) for HP-SAPO-5_0.1, HP-SAPO-5_0.2 and HP-SAPO-5_0.3 (respectively) to a model which sums 2 Gaussian peaks a Power Law and a Fractal with Polydispersity (2G + P + F), as defined in Table S4. D) Shows the residual intensity from these models, serving as a goodness of fit.

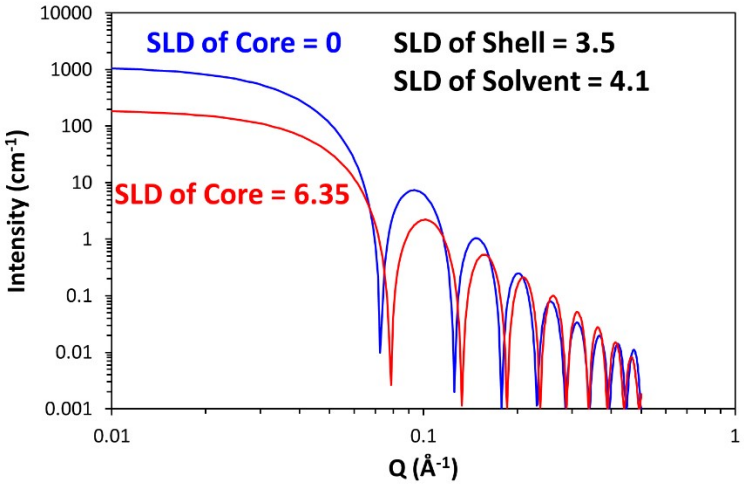


Figure S12: Showing the variation in local maxima with a core-shell sphere equation based on a fixed SLD of solvent and shell, but varying the core SLD (all in 10^{-6} Å^{-2}). Scale = 0, Background = 0.001 cm^{-1}, Radius = 50 Å, Thickness = 10 Å.

4 Gaussians, Power Law and Fractal

Table S5: Fitting parameters and values, with associated uncertainties, achieved with a model including 4 Gaussians, Power Law and Fractal for the three HP-SAPO-5 systems.

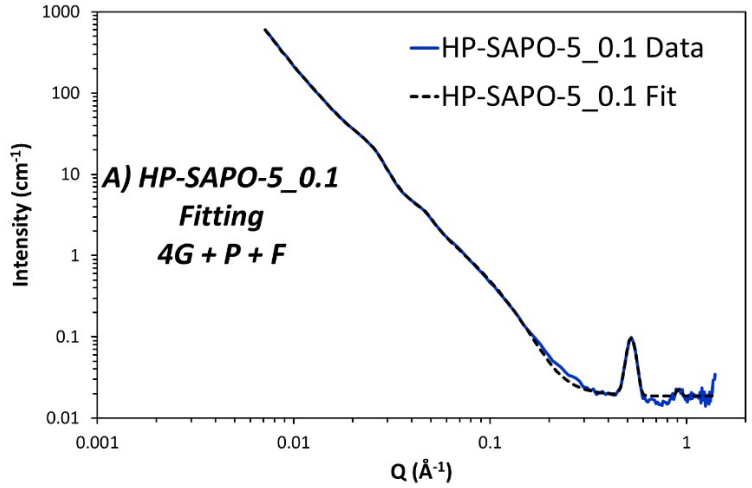
Parameter	HP-SAPO-5_0.1	HP-SAPO-5_0.2	HP-SAPO-5_0.3
Goodness of fit			
Reduced χ^2	7.829	10.253	17.390
General parameters			
Overall Scale (Fixed)	1	1	1
Background, cm^{-1}	0.019 ± 1.7x10^{-4}	0.020 ± 2.3x10^{-4}	0.024 ± 1.6x10^{-4}
1st Gaussian peak, Bragg peak, 100			
Scale, **A** (> 0)	0.087 ± 0.001	0.078 ± 0.001	0.067 ± 0.001
Position, Å^{-1} (0.50 to 0.55)	0.523 ± 2.5x10^{-4}	0.524 ± 2.9x10^{-4}	0.524 ± 3.2x10^{-4}
Width[a], Å^{-1}	0.024 ± 2.9x10^{-4}	0.023 ± 3.4x10^{-4}	0.023 ± 3.1x10^{-4}
2nd Gaussian peak, Bragg peak, 110			
Scale, **B** (> 0)	0.005 ± 0.002	0.004 ± 0.002	5.696x10^{-5} ± 1.4x10^{-4}
Position, Å^{-1} (0.90 to 0.95)	0.913 ± 0.008	0.926 ± 0.010	0.913 ± 0.016
Width[a], Å^{-1}	0.020 ± 0.011	0.026 ± 0.013	0.001 ± 1.5x10^{-4}
3rd Gaussian peak, Hexagonal close packing diffraction			
Scale, **C** (> 0)	7.962 ± 0.072	9.510 ± 0.095	9.209 ± 0.051

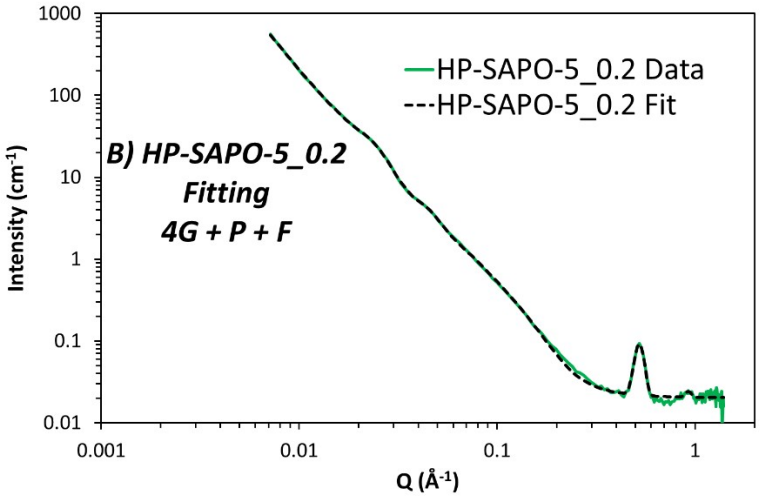
Position, $Å^{-1}$	0.023 $\pm 5.0x10^{-5}$	0.022 $\pm 5.2x10^{-5}$	0.023 $\pm 3.0x10^{-5}$
Width[a], $Å^{-1}$	0.004 $\pm 6.1x10^{-5}$	0.004 $\pm 6.2x10^{-5}$	0.004 $\pm 2.7x10^{-5}$
4^{th} Gaussian peak, Hexagonal close packing diffraction			
Scale, **D** (> 0)	0.574 ± 0.012	0.576 ± 0.013	0.627 ± 0.008
Position, $Å^{-1}$	0.046 $\pm 1.3x10^{-4}$	0.046 $\pm 1.4x10^{-4}$	0.045 $\pm 8.0x10^{-5}$
Width[a], $Å^{-1}$	0.004 $\pm 1.7x10^{-4}$	0.004 $\pm 1.8x10^{-4}$	0.005 $\pm 8.4x10^{-5}$
Power law			
Scale, **E** (> 0)	$8.145x10^{-6}$ $\pm 1.4x10^{-6}$	$1.432x10^{-4}$ $\pm 1.3x10^{-5}$	$1.815x10^{-5}$ $\pm 2.1x10^{-7}$
Power (3 to 4)	3.544 ± 0.033	3.000 ± 0.018	3.381 ± 0.002
Fractal			
Scale, **F** (0 to 1)	0.017 $\pm 1.8x10^{-4}$	0.014 $\pm 3.5x10^{-4}$	0.021 $\pm 6.2x10^{-5}$
Radius, Å	14.347 ± 0.058	12.858 ± 0.062	11.499 ± 0.045
Fractal dimension (2 to 3)	2.619 ± 0.010	2.325 ± 0.021	2.524 ± 0.003
Correlation length, Å	110.88 ± 2.36	130.10 ± 9.73	98.81 ± 0.67
SLD Block, 10^{-6} $Å^{-2}$ (Fixed)	0	0	0
SLD Solvent, 10^{-6} $Å^{-2}$ (Fixed)	4.1	4.1	4.1
Radius Polydispersity[b] (Fixed)	0.3	0.3	0.3

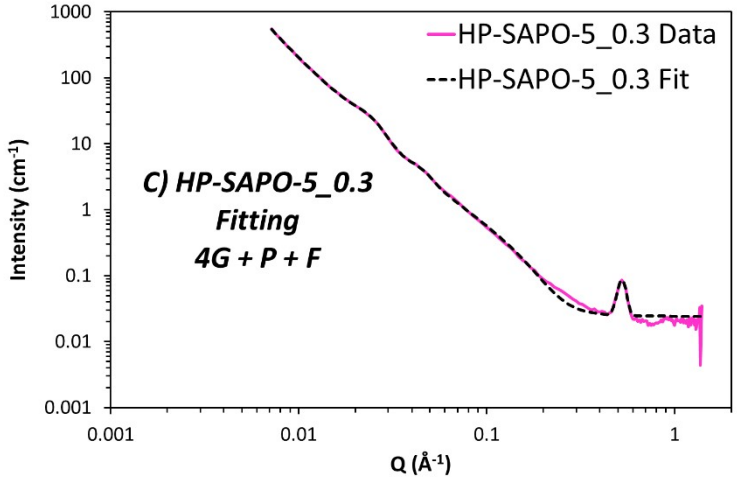
a) One standard deviation = FWHM/2.354

b) HWHM of Lognormal distribution at the median radius divided by the median radius.

The model here is described by 4Gaussian peaks, a Power Law and a Fractal with polydispersity; $Intensity_{4G+P+F\ PD}$ = Background + A.$Intensity_{Gaussian1}$ + B.$Intensity_{Gaussian2}$ + C.$Intensity_{Gaussian3}$ + D.$Intensity_{Gaussian4}$ + E.$Intensity_{PowerLaw}$ + F.$Intensity_{Fractal+PD}$. A-F here are scalar quantities that are proportional to the volume fraction of that component in the model. Only the intensity terms are functions of Q. Links to the individual models are found at the end of the ESI.


HP-SAPO-5_0.1 Data
HP-SAPO-5_0.1 Fit
A) HP-SAPO-5_0.1
Fitting
4G + P + F
Intensity (cm-1)
Q (Å-1)
1000
100
10
1
0.1
0.01
0.001
0.01
0.1
1


HP-SAPO-5_0.2 Data
HP-SAPO-5_0.2 Fit
B) HP-SAPO-5_0.2
Fitting
4G + P + F
Intensity (cm-1)
Q (Å-1)
1000
100
10
1
0.1
0.01
0.001
0.01
0.1
1


HP-SAPO-5_0.3 Data
HP-SAPO-5_0.3 Fit
C) HP-SAPO-5_0.3
Fitting
4G + P + F
Intensity (cm-1)
Q (Å-1)
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0.01
0.1
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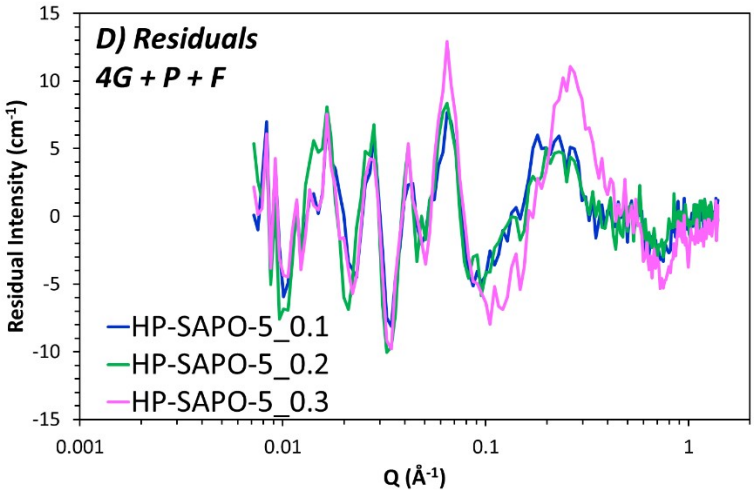


Figure S13: Showing SANS fits (A-C) for HP-SAPO-5_0.1, HP-SAPO-5_0.2 and HP-SAPO-5_0.3 (respectively) to a model which sums 4 Gaussian peaks a Power Law and a Fractal with Polydispersity (4G + P + F), as defined in Table S5. D) Shows the residual intensity from these models, serving as a goodness of fit.

4 Gaussians, Power Law, Fractal and Sphere

Table S6: Fitting parameters and values, with associated uncertainties, achieved with a model including 4 Gaussians, Power Law, Fractal and a Sphere for the three HP-SAPO-5 systems.

Parameter	HP-SAPO-5_0.1	HP-SAPO-5_0.2	HP-SAPO-5_0.3
Goodness of fit			
Reduced χ^2	3.435	5.035	5.660
General parameters			
Overall Scale (Fixed)	1	1	1
Background, cm^{-1}	0.017 ± $1.4x10^{-4}$	0.020 ± $1.5x10^{-4}$	0.021 ± $4.4x10^{-6}$
1st Gaussian peak, Bragg peak, 100			
Scale, **A** (> 0)	0.087 ± 0.001	0.078 ± 0.001	0.069 ± 0.001
Position, $Å^{-1}$ (0.50 to 0.55)	0.523 ± $2.5x10^{-4}$	0.524 ± $3.0x10^{-4}$	0.524 ± $3.2x10^{-4}$
Width[a], $Å^{-1}$	0.024 ± $2.2x10^{-4}$	0.023 ± $3.5x10^{-4}$	0.025 ± $3.5x10^{-4}$
2nd Gaussian peak, Bragg peak, 110			
Scale, **B** (> 0)	0.006 ± 0.001	0.004 ± 0.002	0.312 ± nan
Position, $Å^{-1}$ (0.90 to 0.95)	0.914 ± 0.007	0.925 ± 0.010	0.934 ± nan
Width[a], $Å^{-1}$	0.027 ± 0.002	0.027 ± 0.013	0.001 ± nan
3rd Gaussian peak, Hexagonal close packing diffraction			

Scale, **C** (> 0)	8.095 ± 0.035	8.997 ± 0.145	9.252 ± 0.115
Position, $Å^{-1}$	0.023 ± $2.7x10^{-5}$	0.023 ± $5.8x10^{-5}$	0.023 ± $5.2x10^{-5}$
Width[a], $Å^{-1}$	0.004 ± $2.5x10^{-5}$	0.004 ± $8.1x10^{-5}$	0.004 ± $7.0x10^{-5}$
4^{th} Gaussian peak, Hexagonal close packing diffraction			
Scale, **D** (> 0)	0.634 ± 0.008	0.695 ± 0.020	0.695 ± 0.017
Position, $Å^{-1}$	0.045 ± $8.1x10^{-5}$	0.044 ± $2.3x10^{-4}$	0.044 ± $1.8x10^{-4}$
Width[a], $Å^{-1}$	0.005 ± $6.7x10^{-5}$	0.004 ± $2.2x10^{-4}$	0.005 ± $2.0x10^{-4}$
Power law			
Scale, **E** (> 0)	$1.913x10^{-6}$ ± $9.1x10^{-9}$	$2.602x10^{-6}$ ± $7.6x10^{-7}$	$9.258x10^{-6}$ ± $2.0x10^{-6}$
Power (3 to 4)	3.823 ± 0.001	3.754 ± 0.054	3.511 ± 0.040
Fractal			
Scale, **F** (0 to 1)	0.015 ± $4.0x10^{-5}$	0.015 ± $1.7x10^{-4}$	0.016 ± $2.0x10^{-4}$
Radius, Å	18.054 ± 0.058	18.696 ± 0.155	15.761 ± 0.115
Fractal dimension (2 to 3)	2.805 ± 0.002	2.815 ± 0.011	2.675 ± 0.012
Correlation length, Å	94.79 ± 0.34	84.55 ± 2.27	88.78 ± 2.29
SLD Block, 10^{-6} $Å^{-2}$ (Fixed)	0	0	0
SLD Solvent, 10^{-6} $Å^{-2}$ (Fixed)	4.1	4.1	4.1
Radius Polydispersity[b] (Fixed)	0.3	0.3	0.3
Sphere[c]			
Scale, **G** (> 0)	0.008 ± $1.0x10^{-4}$	0.013 ± $2.3x10^{-4}$	0.019 ± $2.9x10^{-4}$
SLD, 10^{-6} $Å^{-2}$ (Fixed)	0	0	0
SLD Solvent, 10^{-6} $Å^{-2}$ (Fixed)	4.1	4.1	4.1
Radius, Å	12.351 ± 0.070	12.116 ± 0.118	8.785 ± 0.087

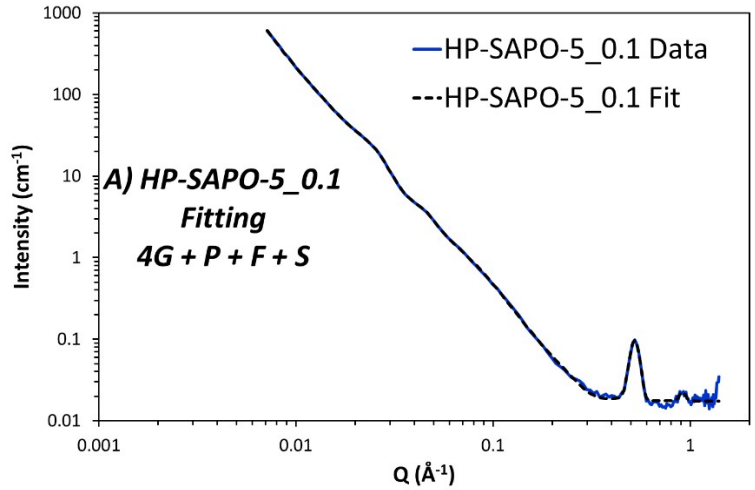
a) One standard deviation = FWHM/2.354

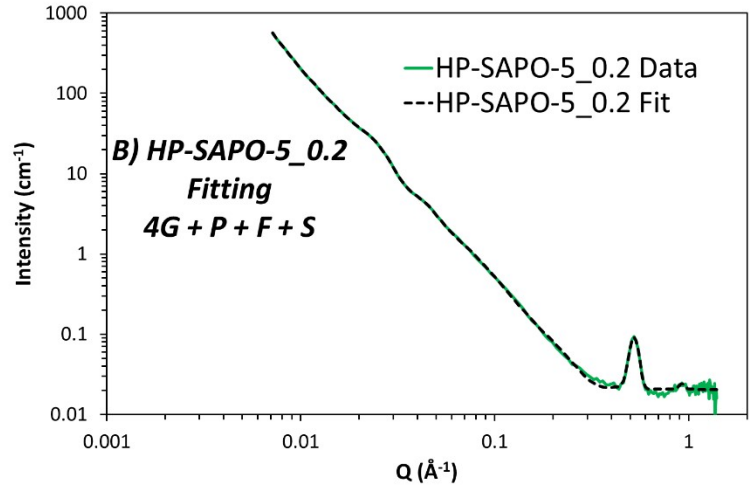
b) HWHM of Lognormal distribution at the median radius divided by the median radius

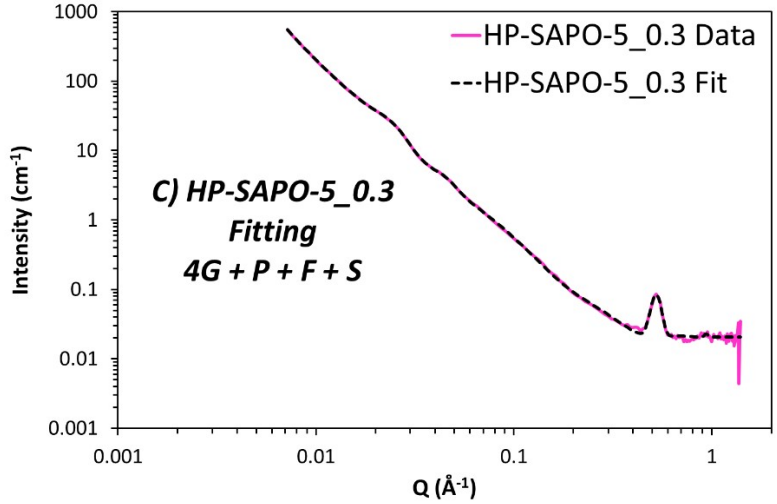
c) Polydispersity was not used for the radius of the sphere.

The model here is described by 4Gaussian peaks, a Power Law, a Fractal with polydispersity and a sphere; $Intensity_{4G+P+F+S\ PD}$ = Background + A.$Intensity_{Gaussian1}$ + B.$Intensity_{Gaussian2}$ + C.$Intensity_{Gaussian3}$ + D.$Intensity_{Gaussian4}$ + E.$Intensity_{PowerLaw}$ + F.$Intensity_{Fractal+PD}$ + G.$Intensity_{Sphere}$. A-G here are scalar

quantities that are proportional to the volume fraction of that component in the model. Only the intensity terms are functions of Q. Links to the individual models are found at the end of the ESI.







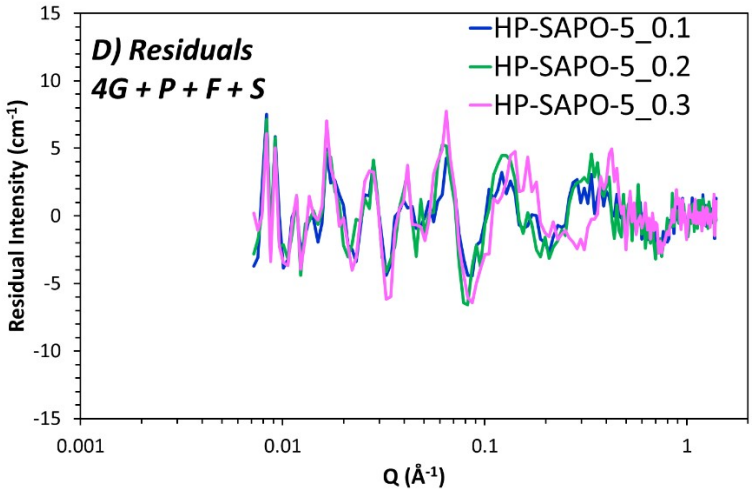


Figure S14: Showing SANS fits (A-C) for HP-SAPO-5_0.1, HP-SAPO-5_0.2 and HP-SAPO-5_0.3 (respectively) to a model which sums 4 Gaussian peaks a Power Law and a Fractal with Polydispersity and a Sphere (4G + P + F + S), as defined in Table S6. D) Shows the residual intensity from these models, serving as a goodness of fit.

4 Gaussians, Power Law, Fractal and Core-Shell Sphere

Table S7: Fitting parameters and values, with associated uncertainties, achieved with a model including 4 Gaussians, Power Law, Fractal and a Core-Shell Sphere for the three HP-SAPO-5 systems.

Parameter	HP-SAPO-5_0.1	HP-SAPO-5_0.2	HP-SAPO-5_0.3
Goodness of fit			
Reduced χ^2	3.141	4.778	4.896
General parameters			
Overall Scale (Fixed)	1	1	1
Background, cm^{-1}	0.017 ± $1.5x10^{-4}$	0.020 ± $2.1x10^{-4}$	0.021 ± $1.3x10^{-4}$
1st Gaussian peak, Bragg peak, 100			
Scale, **A** (> 0)	0.088 ± 0.007	0.078 ± 0.001	0.069 ± 0.001
Position, $Å^{-1}$ (0.50 to 0.55)	0.523 ± $2.6x10^{-4}$	0.523 ± $2.9x10^{-4}$	0.524 ± $3.1x10^{-4}$
Width[a], $Å^{-1}$	0.024 ± $2.2x10^{-4}$	0.0238 ± $3.5x10^{-4}$	0.026 ± $2.9x10^{-4}$
2nd Gaussian peak, Bragg peak, 110			
Scale, **B** (> 0)	0.007 ± 0.001	0.004 ± 0.001	0.002 ± $2.8x10^{-4}$
Position, $Å^{-1}$ (0.90 to 0.95)	0.918 ± 0.006	0.925 ± 0.010	0.909 ± 0.010
Width[a], $Å^{-1}$	0.025 ± 0.002	0.030 ± 0.011	0.018 ± 0.001
3rd Gaussian peak, Hexagonal close packing diffraction			

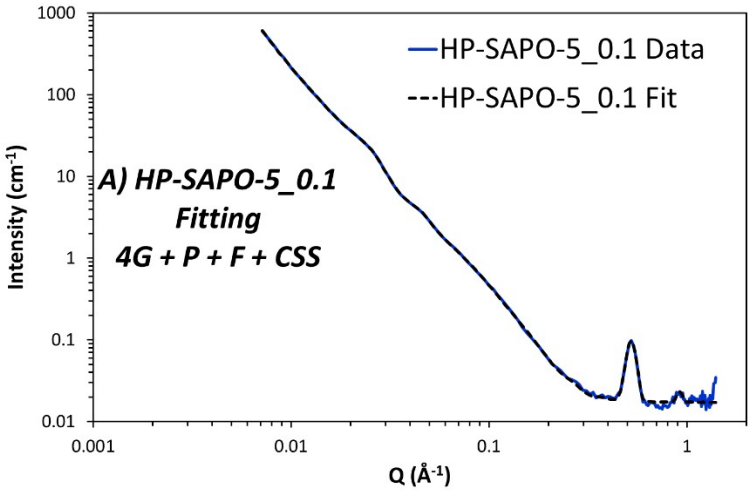
Scale, **C** (> 0)	8.337 ± 0.045	9.280 ± 0.105	9.483 ± 0.039
Position, $Å^{-1}$	0.023 ± $3.3x10^{-5}$	0.023 ± $5.4x10^{-5}$	0.023 ± $2.5x10^{-5}$
Width[a], $Å^{-1}$	0.004 ± $3.2x10^{-5}$	0.004 ± $6.4x10^{-5}$	0.004 ± $2.3x10^{-5}$
4th Gaussian peak, Hexagonal close packing diffraction			
Scale, **D** (> 0)	0.631 ± 0.008	0.654 ± 0.015	0.699 ± 0.010
Position, $Å^{-1}$	0.045 ± $8.3x10^{-5}$	0.044 ± $1.4x10^{-4}$	0.044 ± $8.1x10^{-5}$
Width[a], $Å^{-1}$	0.005 ± $7.6x10^{-5}$	0.005 ± $1.7x10^{-4}$	0.005 ± $6.7x10^{-5}$
Power law			
Scale, **E** (> 0)	$7.212x10^{-7}$ ± $3.1x10^{-9}$	$6.608x10^{-7}$ ± $7.4x10^{-9}$	$1.009x10^{-6}$ ± $5.4x10^{-9}$
Power (3 to 4)	4.000 ± 0.001	4.000 ± $6.2x10^{-9}$	3.918 ± 0.001
Fractal			
Scale, **F** (0 to 1)	0.017 ± $5.3x10^{-5}$	0.020 ± $1.6x10^{-4}$	0.023 ± $9.5x10^{-5}$
Radius, Å	11.263 ± 0.045	10.031 ± 0.117	7.425 ± 0.041
Fractal dimension (2 to 3)	2.815 ± 0.002	2.740 ± 0.007	2.679 ± 0.002
Correlation length, Å	102.07 ± 0.35	102.09 ± 1.82	103.79 ± 0.32
SLD Block, 10^{-6} $Å^{-2}$ (Fixed)	0	0	0
SLD Solvent, 10^{-6} $Å^{-2}$ (Fixed)	4.1	4.1	4.1
Radius Polydispersity[b] (Fixed)	0.3	0.3	0.3
Core-Shell Sphere[c]			
Scale, **G** (> 0)	0.031 ± $2.3x10^{-4}$	0.036 ± 0.004	0.039 ± $3.9x10^{-4}$
Radius, Å	23.472 ± 0.069	22.596 ± 0.715	20.057 ± 0.073
Shell Thickness, Å	20.061 ± 0.109	20.658 ± 0.531	17.306 ± 0.085
SLD Core, 10^{-6} $Å^{-2}$ (Fixed)	0	0	0
SLD Shell, 10^{-6} $Å^{-2}$ (Fixed)	3.5	3.5	3.5
SLD Solvent, 10^{-6} $Å^{-2}$ (Fixed)	4.1	4.1	4.1

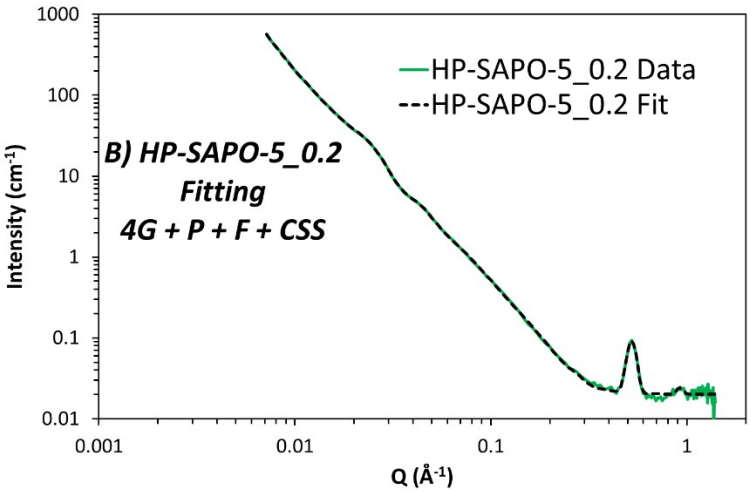
a) One standard deviation = FWHM/2.354

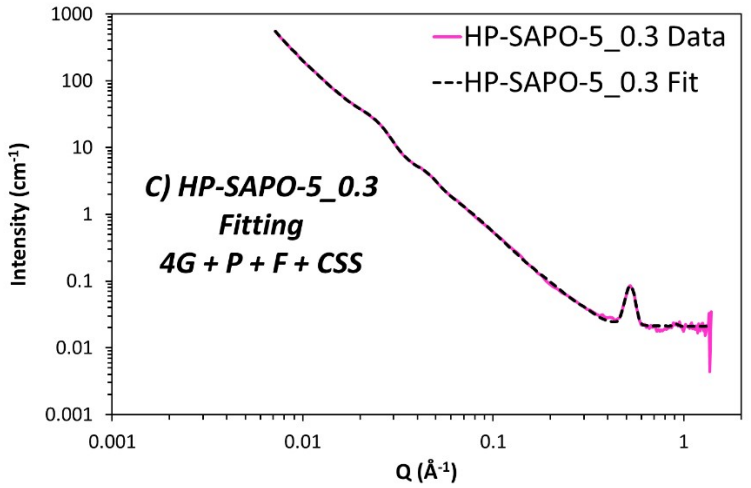
b) HWHM of Lognormal distribution at the median radius divided by the median radius

c) Polydispersity was not used for either the radius or the thickness for the Core-Shell Sphere equation.

The model here is described by 4Gaussian peaks, a Power Law, a Fractal with polydispersity and a Core-shell sphere; $Intensity_{4G+P+F+CSS\ PD}$ = Background + A.$Intensity_{Gaussian1}$ + B.$Intensity_{Gaussian2}$ + C.$Intensity_{Gaussian3}$ + D.$Intensity_{Gaussian4}$ + E.$Intensity_{PowerLaw}$ + F.$Intensity_{Fractal+PD}$ + G.$Intensity_{CoreShellSphere}$. A-G here are scalar quantities that are proportional to the volume fraction of that component in the model. Only the intensity terms are functions of Q. Links to the individual models are found at the end of the ESI.







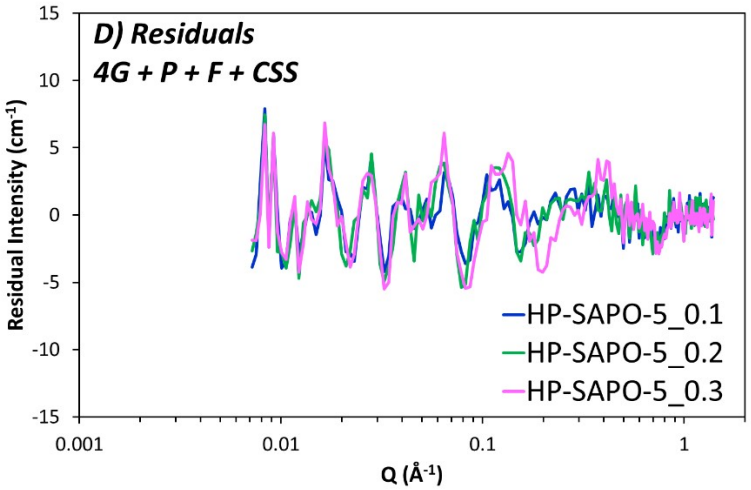
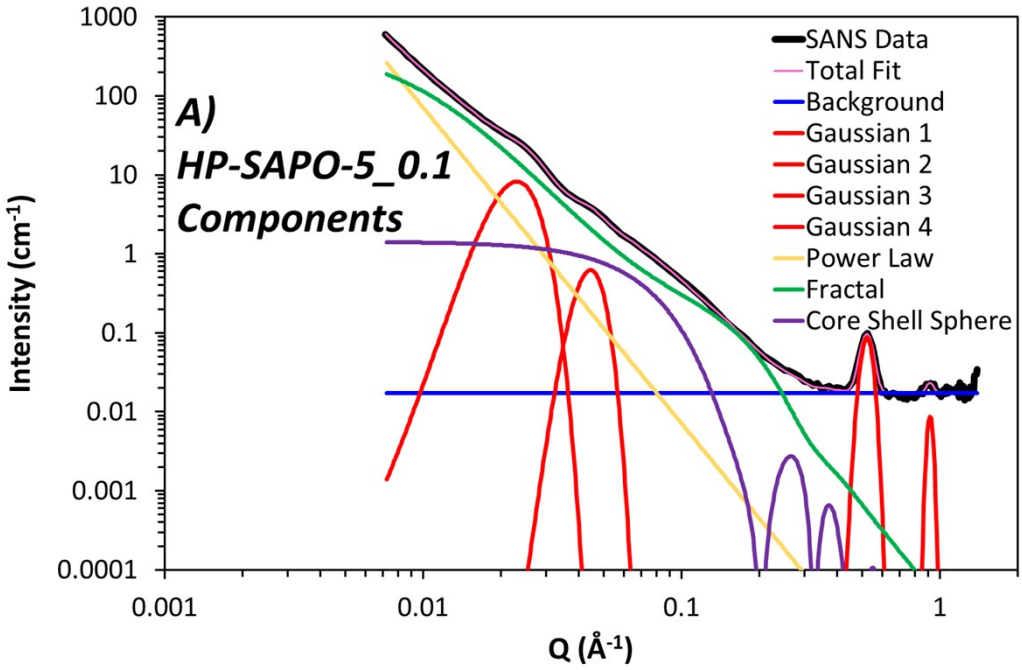


Figure S15: Showing SANS fits (A-C) for HP-SAPO-5_0.1, HP-SAPO-5_0.2 and HP-SAPO-5_0.3 (respectively) to a model which sums 4 Gaussian peaks a Power Law and a Fractal with Polydispersity and a Core-shell Sphere (4G + P + F + CSS), as defined in Table S7. D) Shows the residual intensity from these models, serving as a goodness of fit.



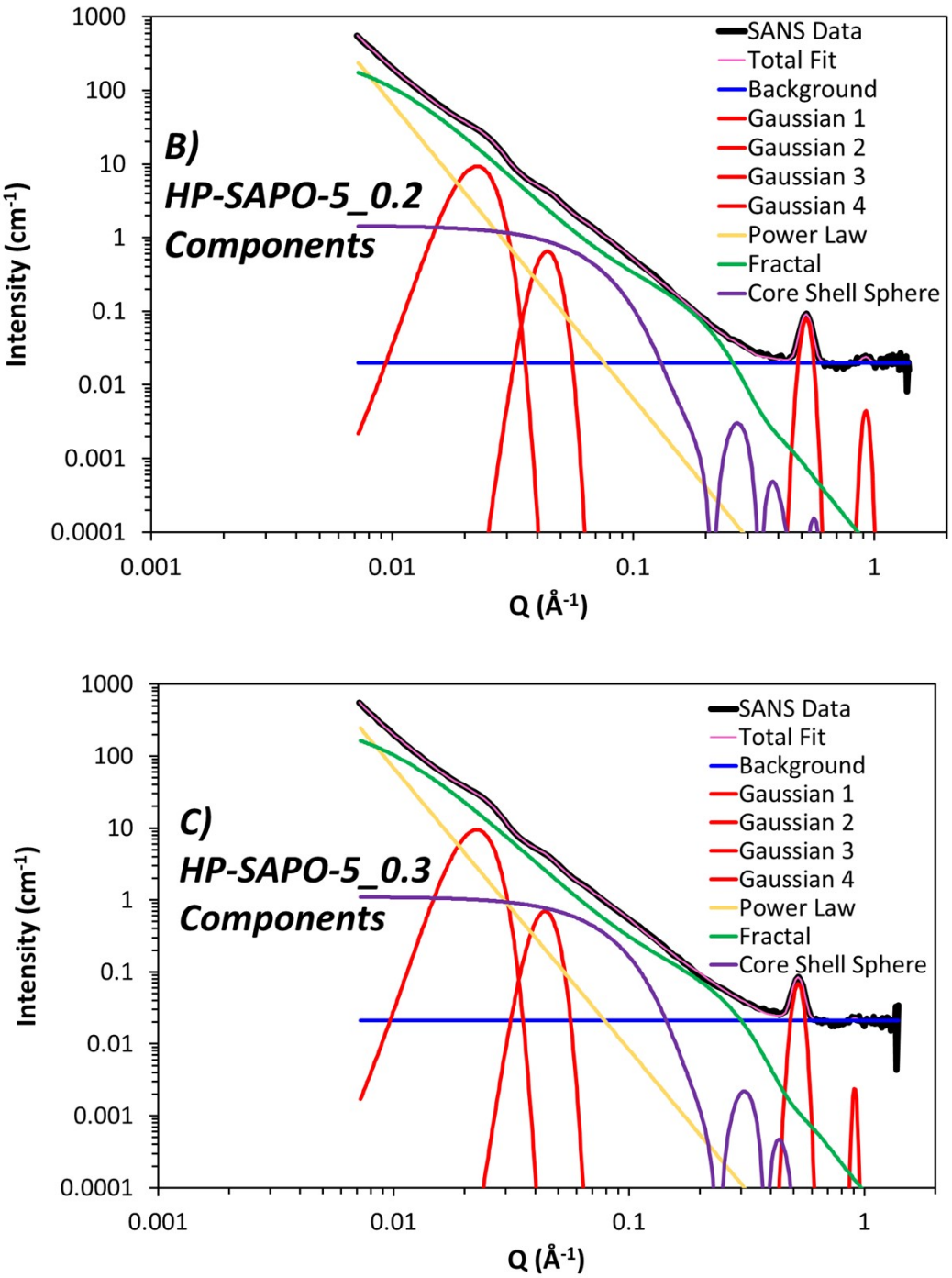


Figure S16: Showing the contribution from each component for the 4 Gaussian, Power Law, Fractal and Core-Shell Sphere fitted model for A) HP-SAPO-5_0.1, B) HP-SAPO-5_0.2 and C) HP-SAPO-5_0.3, for the SANS fit outlined in Table S7.

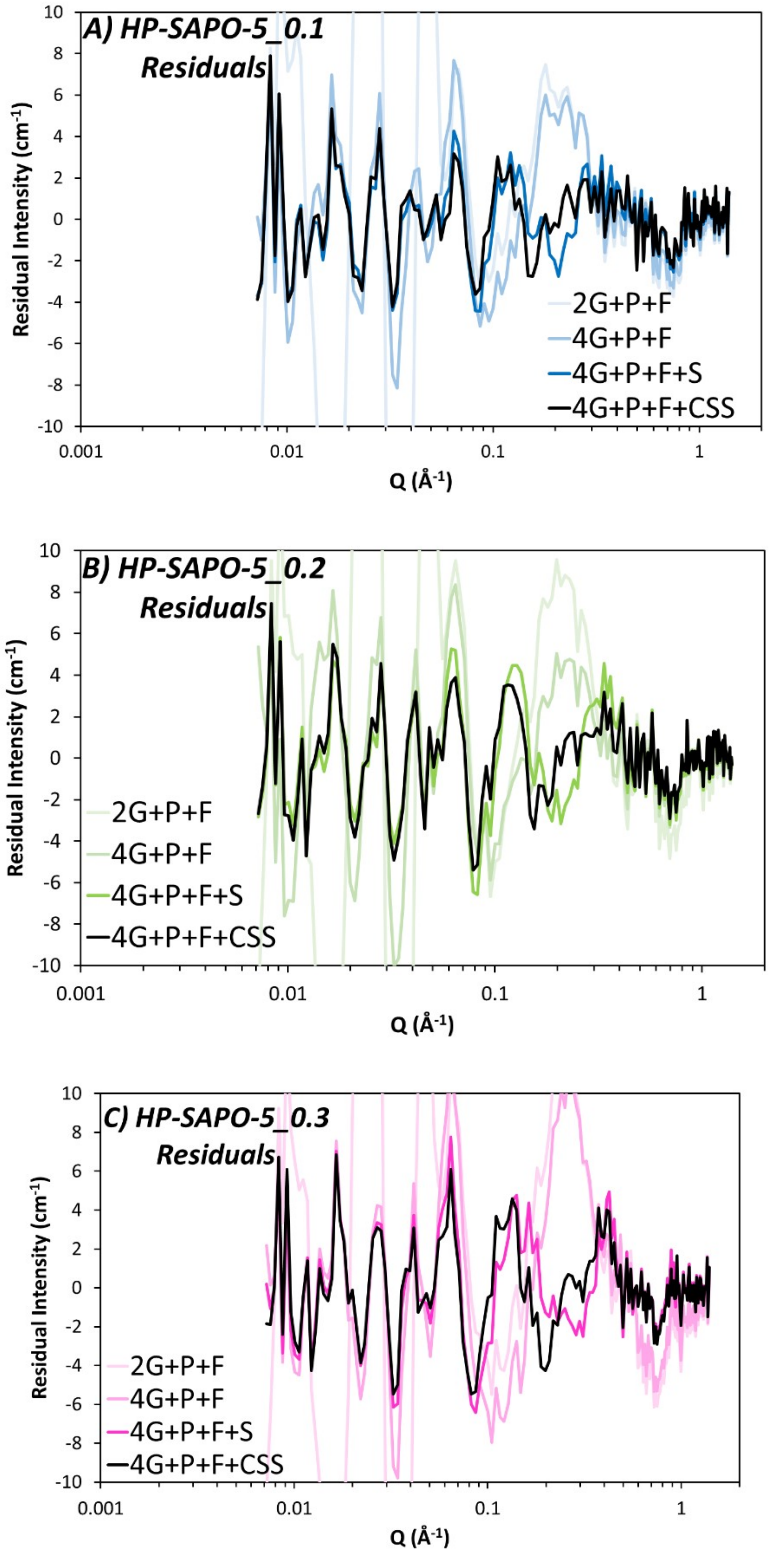


Figure S17: Comparing the residuals for A) HP-SAPO-5_0.1, B) HP-SAPO-5_0.2 and C) HP-SAPO-5_0.3, for a variety of SANS fits outlined in Tables S4 to S7.

Descriptions of Model Terms

Gaussian Peak:	http://www.sasview.org/docs/user/models/gaussian_peak.html
Power Law:	http://www.sasview.org/docs/user/models/power_law.html
Fractal:	http://www.sasview.org/docs/user/models/fractal.html
Sphere:	http://www.sasview.org/docs/user/models/sphere.html
Core-Shell Sphere:	http://www.sasview.org/docs/user/models/core_shell_sphere.html