Supporting Information

A hierarchical scattering function for silica-filled rubbers under deformation:
Effect of the initial cluster distribution.

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Fitting parameters for sample A

γ (%)	$\xi_V\ (nm)$	$\xi_H\ (nm)$	$\zeta_V\ (nm)$	$\zeta_H\ (nm)$	$D_m(V)$	$D_m(H)$
0	9.6 ± 0.2	22.7 ± 0.1	287 ± 6	362 ± 6	3 ± 0.2	2.6 ± 0.1
10	10.4 ± 0.1	23.4 ± 0.1	299 ± 6	354 ± 6	3 ± 0.2	2.5 ± 0.1
20	10.4 ± 0.1	22.6 ± 0.1	350 ± 4	265 ± 4	3 ± 0.2	2.5 ± 0.1
30	11.7 ± 0.2	21.7 ± 0.1	371 ± 9	290 ± 5	3 ± 0.1	2.5 ± 0.1
40	13.8 ± 0.1	20.2 ± 0.2	388 ± 6	278 ± 5	3 ± 0.1	2.5 ± 0.1
50	14.8 ± 0.2	19.8 ± 0.1	421 ± 5	274 ± 5	2.9 ± 0.1	2.5 ± 0.1
60	17.1 ± 0.1	17.6 ± 0.2	465 ± 3	220 ± 3	2.8 ± 0.1	2.5 ± 0.1

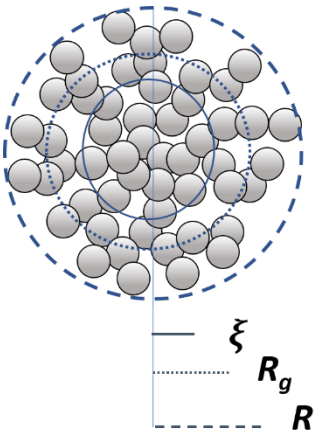
70	19.3 ± 0.2	16.0 ± 0.2	460 ± 3	201 ± 3	2.7 ± 0.1	2.6 ± 0.1
80	20.6 ± 0.1	16.9 ± 0.2	463 ± 3	202 ± 3	2.7 ± 0.1	2.6 ± 0.1
90	23.0 ± 0.2	15.4 ± 0.2	474 ± 4	214 ± 4	2.6 ± 0.1	2.6 ± 0.1
100	24.2 ± 0.2	16.3 ± 0.2	514 ± 4	207 ± 4	2.6 ± 0.1	2.5 ± 0.1
		Unload				
γ (%)	$\xi_V\ (nm)$	$\xi_H\ (nm)$	$\zeta_V\ (nm)$	$\zeta_H\ (nm)$		
100	24.2 ± 0.2	16.3 ± 0.2	514 ± 4	207 ± 4	2.6 ± 0.1	2.5 ± 0.2
90	22.9 ± 0.1	16.5 ± 0.2	487 ± 3	206 ± 4	2.6 ± 0.1	2.5 ± 0.1
80	20.8 ± 0.2	17.2 ± 0.2	438 ± 3	213 ± 4	2.7 ± 0.1	2.6 ± 0.1
70	19.2 ± 0.1	17.2 ± 0.2	419 ± 4	219 ± 4	2.7 ± 0.1	2.6 ± 0.1
60	17.5 ± 0.1	18.9 ± 0.2	397 ± 3	211 ± 4	2.7 ± 0.1	2.5 ± 0.1
50	15.3 ± 0.1	19.1 ± 0.2	387 ± 3	222 ± 4	2.8 ± 0.1	2.6 ± 0.1
40	14.7 ± 0.1	19.6 ± 0.2	366 ± 4	238 ± 4	2.8 ± 0.1	2.5 ± 0.1
30	12.9 ± 0.1	20.6 ± 0.2	359 ± 4	247 ± 4	2.9 ± 0.1	2.6 ± 0.1
20	11.7 ± 0.1	22.2 ± 0.2	346 ± 3	231 ± 4	3 ± 0.1	2.6 ± 0.1
10	10.4 ± 0.1	23.6 ± 0.2	298 ± 7	358 ± 4	3 ± 0.1	2.6 ± 0.1
0	9.7 ± 0.1	23.2 ± 0.2	293 ± 6	365 ± 4	3 ± 0.1	2.6 ± 0.1

Fitting parameters for Sample B

γ (%)	ξ_V (nm)	ξ_H (nm)	ζ_V (nm)	ζ_H (nm)	$D_m(V)$	$D_m(H)$
0	18.0 ± 0.1	10.4 ± 0.1	399 ± 5	290 ± 5	2.7 ± 0.1	3 ± 0.1
10	21.4 ± 0.1	10.6 ± 0.1	389 ± 5	284 ± 5	2.7 ± 0.1	3 ± 0.1
20	23.0 ± 0.1	10.5 ± 0.1	418 ± 5	273 ± 5	2.6 ± 0.1	3 ± 0.1
30	24.1 ± 0.1	9.9 ± 0.1	391 ± 5	265 ± 5	2.6 ± 0.1	3 ± 0.1
40	25.0 ± 0.1	10.0 ± 0.1	381 ± 5	258 ± 5	2.6 ± 0.1	3 ± 0.1
50	26.0 ± 0.2	10.5 ± 0.2	396 ± 5	256 ± 5	2.6 ± 0.1	2.7 ± 0.1
60	26.2 ± 0.1	10.7 ± 0.1	389 ± 5	251 ± 5	2.6 ± 0.1	2.7 ± 0.1
70	26.8 ± 0.2	10.1 ± 0.1	409 ± 5	249 ± 4	2.6 ± 0.1	2.7 ± 0.1
80	27.3 ± 0.2	10.7 ± 0.1	352 ± 4	261 ± 4	2.6 ± 0.1	2.7 ± 0.1
90	27.4 ± 0.1	10.1 ± 0.1	340 ± 4	254 ± 4	2.6 ± 0.1	2.7 ± 0.1
100	27.5 ± 0.2	10.9 ± 0.1	356 ± 4	259 ± 4	2.6 ± 0.1	2.7 ± 0.1
		Unload				
γ (%)	ξ_V (nm)	ξ_H (nm)	ζ_V (nm)	ζ_H (nm)		
100	27.5 ± 0.1	10.9 ± 0.1	356 ± 4	259 ± 4	2.6 ± 0.1	2.7 ± 0.1
90	26.2 ± 0.1	10.8 ± 0.1	346 ± 4	247 ± 4	2.5 ± 0.1	3 ± 0.1
80	24.7 ± 0.2	9.7 ± 0.1	343 ± 5	255 ± 5	2.5 ± 0.1	3 ± 0.1
70	23.9 ± 0.1	10.1 ± 0.1	345 ± 4	248 ± 5	2.5 ± 0.1	3 ± 0.1
60	23.7 ± 0.1	9.7 ± 0.1	334 ± 4	256 ± 5	2.5 ± 0.1	3 ± 0.1
50	24.3 ± 0.1	10.0 ± 0.1	340 ± 5	251 ± 5	2.5 ± 0.1	3 ± 0.1
40	24.1 ± 0.1	9.7 ± 0.1	336 ± 5	259 ± 5	2.5 ± 0.1	3 ± 0.1
30	24.2 ± 0.1	10.0 ± 0.1	344 ± 5	271 ± 4	2.5 ± 0.1	3 ± 0.1
20	22.9 ± 0.1	10.1 ± 0.1	340 ± 5	298 ± 5	2.5 ± 0.1	3 ± 0.1
10	19.6 ± 0.1	9.7 ± 0.2	353 ± 5	307 ± 5	2.6 ± 0.1	3 ± 0.1

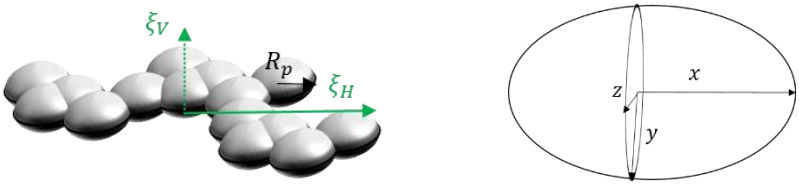
0	21.0 ± 0.1	11.1 ± 0.2	365 ± 5	314 ± 5	2.6 ± 0.1	3 ± 0.1

Estimation of aggregation number per cluster (N_{agg})



Schematic representation of an isotropic cluster and characteristic parameters: Fractal dimension (ξ), Radius of gyration (R_g) and radius (R).

In our investigation, an aggregation number for the anisometric case is estimated for the sample A at $\gamma = 0$ and along the two perpendicular directions, as represented in the scheme below.



$R_p\ (particles) \approx 8\ nm$

$\xi_H \approx 27\ nm$ $\quad D_m(H) \approx 2.6$ (corresponding to the long x axis)

$\xi_V \approx 10\ nm$ $\quad D_m(V) \approx 3$ (corresponding to the short y axis, assuming y=z)

The parameter ξ was obtained in two directions. We assume here that the two short axes in the ellipsoidal clusters are equal.

$$R_g = \sqrt{\frac{D_m \cdot (D_m + 1)}{2}} \cdot \xi$$

$$R = \sqrt{\frac{5}{3}} \cdot R_g$$

$R_H \approx 75\ nm,$
$R_V \approx 31\ nm$

From this we can compute a fictive aggregate number for a cluster with the respective radii, following:

$$N_{agg} = \left(\frac{R}{R_p}\right)^{D_m}$$

The results is $N_{agg} \approx 340$ corresponding to the longest axis (H) and $N_{agg} \approx 60$ for the short axis (V).

If the clusters would be isotropic then the number of particles (N_p) would correspond to:

$$N_p = \sqrt[3]{N_{agg}}$$

Along the three directions X, Y and Z.

With the N_{agg} calculated before we estimate then for the example of sample A at $\gamma = 0$:

$N_p(H) \approx 7$

$N_p(V) \approx 4$ (along the two short directions)

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