Supporting Information for

Supramolecular Elastomers. Particulate β-Sheet Nanocrystal-Reinforced Synthetic Elastic Networks

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1. Additional AFM images

**AFM.** Both the height and phase images obtained in the tapping mode only revealed features attributed to the microtoming process as shown in Figure S1.

![Height (left) and phase (right) images of microtomed films of 4c obtained from tapping mode AFM. Only shear banding and contours from microtoming are observed.](image)

**Figure S1.** Height (left) and phase (right) images of microtomed films of 4c obtained from tapping mode AFM. Only shear banding and contours from microtoming are observed.

![Height-adhesion overlay AFM images (500 nm x 500 nm) of 4c in QNM mode.](image)

**Figure S2.** Height-adhesion overlay AFM images (500 nm x 500 nm) of 4c in QNM mode.
2. Particle analysis based on adhesion maps from AFM

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Mean</th>
<th>Minimum</th>
<th>Maximum</th>
<th>Sigma</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total Count</td>
<td>580</td>
<td>580</td>
<td>580</td>
<td>0.00</td>
</tr>
<tr>
<td>Density</td>
<td>2000 (µm(^2))</td>
<td>2000 (µm(^2))</td>
<td>2000 (µm(^2))</td>
<td>0.00</td>
</tr>
<tr>
<td>Height</td>
<td>-0.3 (nN)</td>
<td>-0.6 (nN)</td>
<td>-0.1 (nN)</td>
<td>0.08 (nN)</td>
</tr>
<tr>
<td>Area</td>
<td>102.9 (nm(^2))</td>
<td>23.8 (nm(^2))</td>
<td>14816 (nm(^2))</td>
<td>622.8 (nm(^2))</td>
</tr>
<tr>
<td>Diameter</td>
<td>9.2 (nm)</td>
<td>5.5 (nm)</td>
<td>137.3 (nm)</td>
<td>6.8 (nm)</td>
</tr>
</tbody>
</table>
Figure S3. Particle analysis using an adhesion map. Top, Adhesion map with particles identified by NanoScope Analysis v1.2 software. Middle, Histogram of particle adhesion distribution. Bottom, Summary of analysis.

3. Energy dispersive X-ray analysis

Figure S4. EDS of Areas 1 and 2 indicated by the squares. Area 1 is contaminated by Al which is most likely the residual of EAI\textsubscript{2}Cl\textsubscript{2} catalyst for the Alder ene reaction.
4. Derivation of the relationship of \( N_{\text{agg}} \) and \( K \) according to open association model.

We rewrite the open association as the following:

\[
U_x \text{-} a + d \text{-} U_y \leftrightarrow U_x \text{-} ad \text{-} U_y
\]

where \( d \text{-} U_x \) represents all aggregates of any degree of aggregation with a reactive end group “d”, \( U_y \text{-} a \) represents all aggregates of any degree of aggregation with a reactive end group “a”, and \( U_x \text{-} ad \text{-} U_y \) represents all aggregates of any degree of aggregation and contains reacted “ad” groups. Further, the equilibrium constant of the above process is \( K \), the extent of reaction of either the reactive group “a” or “d” at equilibrium is \( p \), and the number average degree of aggregation to be \( N_{\text{agg}} \) at eq, and \( N_{\text{agg}} = (1-p)^{-1} \), and the initial concentration of the reactive groups “a” and “d” is the initial unimer concentration, \([U]_o\).

At equilibrium, the concentrations of the reactive groups “d” and “a” are both \(( [U]_o - p [U]_o ) \), and the concentration of the reacted group “ad” is \( p [U]_o \). Hence, the equilibrium constant can be expressed as:

\[
K = p [U]_o / ( [U]_o - p [U]_o )^2
\]

Substitute \( N_{\text{agg}} \) for \( p \),

\[
K = (N_{\text{agg}}^2 - N_{\text{agg}}) / [U]_o
\]

Solve for \( N_{\text{agg}} \),

\[
N_{\text{agg}} = \frac{1}{2} [1 + (1 + 4 K [U]_o)^{1/2}] \]

When \( N_{\text{agg}} \) is a relatively large number, \( N_{\text{agg}} \approx (K [U]_o)^{1/2} \)