Supplementary Figure S1: Possible geometries for Phe-Phe interactions

\(\pi-\pi\) stacking interactions with parallel displaced (left) or T-shaped (right) geometries. Examples are generated from the pdb entry 1GYB\(^1\) using Pymol. The excimer emission band (Figure 1C inset) suggests similar Phe-Phe interactions to occur within Nsp1\(^{2-601}\) hydrogels.

Supplementary Table S2: Secondary structural elements of Nsp1\(^{2-601}\) in the liquid and gel states as monitored by circular dichroism spectroscopy and analyzed by CDSSTR.

<table>
<thead>
<tr>
<th>state</th>
<th>(\alpha)-helix</th>
<th>3/10-helix</th>
<th>(\beta)-sheet</th>
<th>turn</th>
<th>polyproline II like conformation</th>
<th>random coil</th>
</tr>
</thead>
<tbody>
<tr>
<td>liquid</td>
<td>4.5 %</td>
<td>5.2 %</td>
<td>9.4 %</td>
<td>17.6 %</td>
<td>15.2 %</td>
<td>46.5 %</td>
</tr>
<tr>
<td>gel</td>
<td>3.5 %</td>
<td>3.3 %</td>
<td>24.9 %</td>
<td>14.2 %</td>
<td>11.1 %</td>
<td>42.3 %</td>
</tr>
</tbody>
</table>

Supplementary Table S3: Secondary structural elements of Nsp1\(^{2-601}\) in the liquid and gel states as monitored by FTIR spectroscopy.

<table>
<thead>
<tr>
<th>state</th>
<th>(\alpha)-helix</th>
<th>(\beta)-sheet</th>
<th>turn</th>
<th>disordered contribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>liquid</td>
<td>5 %</td>
<td>13 %</td>
<td>22 %</td>
<td>60 %</td>
</tr>
<tr>
<td>gel</td>
<td>9 %</td>
<td>28 %</td>
<td>19 %</td>
<td>44 %</td>
</tr>
</tbody>
</table>
Supplementary Text S4: The structural hierarchy of the hydrogel meshwork

For an evaluation of the average mesh size in the gels, an overall fit function has been developed. This comprises the Ornstein-Zernike equation

\[ I_{OZ}(q) = \frac{I_{OZ}(0)}{1 + \xi_{OZ}^2 q^2} \]  
(2)

and the Guinier equation

\[ I_G(q) = I_G(0) \exp \left[ \frac{-R_G^2 q^2}{3} \right] \]  
(3)

which is frequently applied to model the scattering data of chemically cross-linked polymer gels.\(^2\) Here \(\xi_{OZ}\) is the correlation length which is attributed to the pore size of the meshwork and \(R_G\) the radius of gyration which characterizes the condensed nodes. Given that the observation of a scattering peak in gels points to the existence of a microphase separation we modelled this contribution by the Teubner-Strey equation.\(^3\)

\[ I(t) \propto \frac{1}{a_2 + c_1(t) q^2 + c_2(t) q^4} \]  
(4)

Although this equation has been originally deviated to explain the scattering pattern of microemulsions it has be shown to equally well produce satisfactory fits\(^4\) to sponge-like hydrogels.

Here the parameters \(a_2\), \(c_1\), and \(c_2\) are employed to extract the correlation length \(\xi_{TS}\) of the protein-rich domains and the repeat distance \(d(t)\). The latter denotes the average distance between two protein-rich or protein-poor domains.

\[ \xi(t) = \frac{1}{\sqrt{\frac{1}{2} \left( \frac{a_2}{c_2(t)} \right) + \left( \frac{c_1(t)}{4 c_2(t)} \right)}} \]  
(5)

and

\[ d(t) = \frac{1}{\frac{1}{2} \sqrt{\left( \frac{a_2}{c_2(t)} \right) - \left( \frac{c_1(t)}{4 c_2(t)} \right)}} \]  
(6)

Taking these elements into consideration, it follows that the overall fit function \(I_{total}\) covering the high- and low-q-range of the experimental curve comprises three terms:

\[ I_{total} = I_{TS} + I_{OZ} + I_G. \]  
(7)
From the observation of Figure 4D, it can be seen that this function renders an adequate description of the experimental data ($R^2 = 0.9996$). Hence the morphology of the hydrogel can be described by two microscopically separated volume fractions of protein-rich and protein-poor domains with a correlation length of $\xi_{TS}=16.5$ nm and a repeat distance of $d=40$ nm. On a smaller length scale a meshwork exists with average pore sizes of 2.7 nm ($\pm 1.1$ nm) and radii of gyration referring to the size of the cross-linked domains of 4.6 nm.

Supporting References