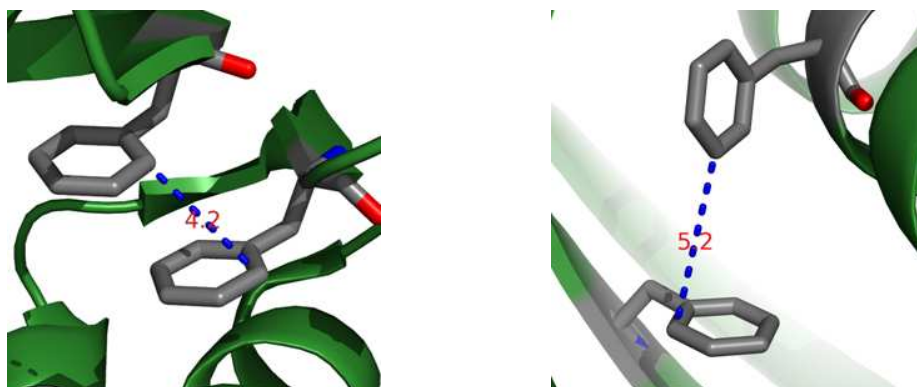**Supplementary Figure S1: Possible geometries for Phe-Phe interactions**

π-π 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.

state	α–helix	3/10-helix	β-sheet	turn	polyproline II like conformation	random coil
liquid	4.5 %	5.2 %	9.4 %	17.6 %	15.2 %	46.5 %
gel	3.5 %	3.3 %	24.9 %	14.2 %	11.1 %	42.3 %

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

state	α–helix	β-sheet	turn	disordered contribution
liquid	5 %	13 %	22 %	60 %
gel	9 %	28 %	19 %	44 %

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

$$I_{OZ}(q) = \frac{I_{OZ}(0)}{\left(1 + \xi_{OZ}^2 q^2\right)} \tag{2}$$

and the Guinier equation

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

which is frequently applied to model the scattering data of chemically cross-linked polymer gels.[2] Here ξ_{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]

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

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 ξ_{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.

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

and

$$\frac{d(t)}{2\pi} = \frac{1}{\sqrt{\frac{1}{2}\sqrt{\left(\frac{a_2}{c_2(t)}\right)} - \left(\frac{c_1(t)}{4\,c_2(t)}\right)}} \;. \tag{6}$$

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:

$$I_{total} = I_{TS} + I_{OZ} + I_G \;. \tag{7}$$

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 ξ_{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 (±1.1 nm) and radii of gyration referring to the size of the cross-linked domains of 4.6 nm.

Supporting References

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