Tough and elastic thermoplastic organogels and elastomers made of semi-crystalline polyolefin-based block copolymers

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Supplemental Information

Correction for the volume fraction of the mineral oil
Figure S1. (a) Nominal stress versus nominal strain curves of the gel, the dried gel and the gel after accounting for the volume fraction of the mineral oil (in grey). (b) Evolution of the maximal tangent modulus as a function of the maximal true strain of the gel (open squares), the dried gel (open circles) and the gel after correction for the volume fraction of the mineral oil.
(filled squares) and (c) Young’s modulus as a function of the maximal true strain of the gel (open squares), the dried gel (open circles) and the gel after correction for the volume fraction of the mineral oil (filled squares).

**Total viscous true stress**

The total relaxing true stress is set up of two parts representing an elastic strain (modulus $E_r$) and a viscous deformation described by the Eyring law of viscosities:

$$\dot{\varepsilon}_H = \frac{\dot{\sigma}_r}{E_r} + \dot{\varepsilon}_0 \sinh \frac{\sigma_r}{\sigma_0} = 0 \quad \text{Eq S1}$$

or

$$\frac{d}{dt} \frac{\sigma_r}{\sigma_0} = \frac{l}{\tau_r} \sinh \frac{\sigma_r}{\sigma_0} \quad \text{Eq S2}$$

with

$$\tau_r^{-1} = \dot{\varepsilon}_0 E_r \frac{E_r}{\eta_0} \quad \text{Eq S3}$$

$\tau_r$ denotes the relaxation time which would be found in the Newtonian limit of very low stresses.

The differential equation (Eq S2) can be solved

$$\frac{\sigma_r(t)}{\sigma_0} = 2 \tanh \left[ \tanh \frac{\sigma_r(0)}{2\sigma_0} \exp \left( -\frac{t}{\tau_r} \right) \right] \quad \text{Eq S4}$$

The experimental curve representing the stress decay $\Delta \sigma$ during the relaxation tests is given by

$$\Delta \sigma = \sigma(0) - \sigma(t) = \sigma_r(0) - \sigma_r(t) = \sigma_r(0) - 2\sigma_0 \tanh \left[ \tanh \frac{\sigma_r(0)}{2\sigma_0} \exp \left( -\frac{t}{\tau_r} \right) \right] \quad \text{Eq S5}$$

Fitting the experimental data (Figure S2-b) obtained from the relaxation tests by this expression (Eq S5) yields the three parameters $\sigma_0$, $\tau_r$ and $\sigma_r(0)$. The total amount of viscous stress is given
by $\sigma(0)$. 

Figure S2. (a) True stress versus true strain curves obtained from monotonic tensile tests on the neat polymer before relaxation at given strains. (b) True stress relaxation $\Delta\sigma(t)$ of the neat polymer after stretching to the given strains.

**Time allowed to the sample to relax at zero load**
Figure S3. Time given to the sample to recover at zero load. (a) For the dried gel and (b) for the gel.

Syndiotactic polypropylene crystalline planes from WAXS data
Figure S4. WAXS intensity profiles obtained from the meridional scans and equatorial scans of the WAXS patterns corrected by the air and the thickness of the sample and after the removal of the amorphous halo. (a) Intensity profiles obtained from the meridional scans of sPP18_dg, before and after deformation, (b) intensity profiles obtained from the equatorial scans of sPP18_dg, before and after deformation, (c) profiles obtained from the meridional scans of sPP18_n, sPP18_dg and sPP18_g after deformation, (d) intensity profiles obtained from the equatorial scans of sPP18_n, sPP18_dg and sPP18_g after deformation.
Figure S5. Azimuthal scans of the WAXS images at $q$~14 nm$^{-1}$. (a) sPP18_n, (b) sPP18_dg and (c) sPP18_g.

**Determination of the radius of the crystalline fibrils**
Figure S6. Peak intensity as a function of $q$ of azimuthal scans done at increasing $q$ values on the SAXS pattern recorded at the maximal true strain of 2.8 during the step cycle test performed on sPP18_dg; definition of $q^*$. 

**Ruland’s method for the analysis of the diffraction streak:**

We used the method proposed by Ruland\textsuperscript{56} to analyze the length and the width of the streak as a function of $q$ to obtain information regarding the average length $l_f$ of the fibrils and the fibril misorientation $\theta$ from the fiber axis. $B_{obs}$ is the full width at half maximum of the azimuthal profiles of the equatorial streak fitted with a Lorentzian function. The average fibril length $l_f$ and the average misorientation $\theta$ have been obtained from the slope and the y-intercept of the $B_{obs}=f(1/q)$ and $qB_{obs}=f(q)$ curves (see Eq S6 and Eq S7).

$$qB_{obs} = \frac{2\pi}{l_f} + q\theta$$  \hspace{2cm} \text{Eq S6}$$

and

$$B_{obs} = \frac{2\pi}{l_f} \frac{1}{q} + \theta$$  \hspace{2cm} \text{Eq S7}$$
The values of the average length $l_f$ and of the average misorientation $\theta$ reported are the average values between the two values obtained using both equations (Eq S6 and Eq S7).