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Supporting Information

Mechanical Properties of Structurally-Defined Magnetoactive Polymer (Co)networks

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Biphasic, poroelastic theory

The biphasic poroelastic theory, developed initially by Biot and applied later by Mow and co-workers, was employed to simulate the response of the polymer networks of the current study [1, 2].

The total stress tensor is given by the sum of the stresses of the solid and fluid phase: $\boldsymbol{\sigma}^{tot} = \boldsymbol{\sigma}^{s} + \boldsymbol{\sigma}^{f}$ where $\boldsymbol{\sigma}^{s}, \boldsymbol{\sigma}^{f}$ are the Cauchy stress tensors of the solid and fluid phase, respectively. The solid stress is calculated by the strain energy density function, *W*, as

$$\boldsymbol{\sigma}^{s} = J^{-1}\mathbf{F}\,\frac{\partial W}{\partial \mathbf{F}} \tag{1}$$

where *J* is the determinant of the deformation gradient tensor **F**, *W* is given by Eq. 1 from the main manuscript and $\mathbf{P} = \partial W / \partial \mathbf{F}$ is the 1st Piola-Kirchhoff stress tensor.

The fluid is assumed to be ideal, i.e., $\boldsymbol{\sigma}^{f} = -p\delta_{ij}$, with *p* the fluid pressure. According to the biphasic theory the linear momentum balance is written as:

$$\nabla \cdot \boldsymbol{\sigma}^{tot} = 0 \Rightarrow \nabla \cdot \left(\boldsymbol{\sigma}^{s} - p\mathbf{I}\right) = 0 \tag{2}$$

and the mass balance is given by the equation:

$$\nabla \cdot \left(\mathbf{v}^{s} + \mathbf{v}^{f}\right) = 0 \tag{3}$$

where $\boldsymbol{v}^{s}$ is the velocity of the solid phase, $\mathbf{v}^{f}$ is the fluid velocity given by Darcy's law: $\mathbf{v}^{f} = -K\nabla p$, with *K* the hydraulic conductivity of the porous material. The hydraulic conductivity is related to the permeability, *k*, of a porous material through the relationship

$$K = \frac{k}{\mu} \tag{4}$$

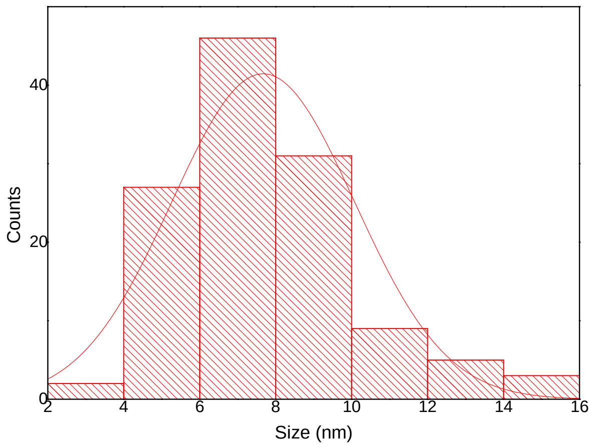
with μ being the viscosity of the fluid.

The boundary conditions employed to simulate the unconfined compression experiment are shown in Supplementary Fig. 2. A detailed description of the implementation of the biphasic model in COMSOL can be found in [3].

Supplementary Figures and Tables

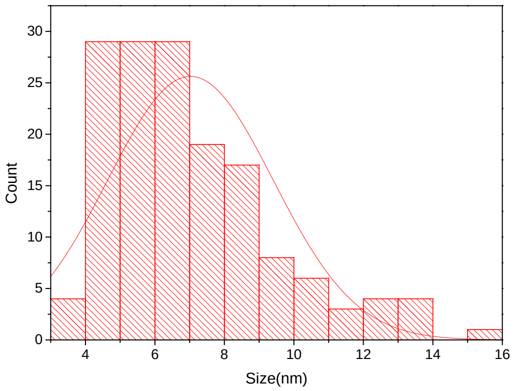
Suppelmentary Figure 1. Size distribution of the Fe_3O_4 nanoparticles embedded within the polymer network matrix based on the TEM analysis.

DMAEMA (10%)



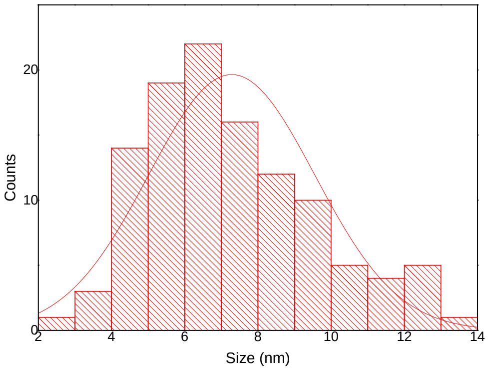
	Ntotal	Mean	Standard Deviation	Minimum	Maximum
B	123	7.67095	2.40601	2.83083	15.453

DMAEMA/BuMA (10%)



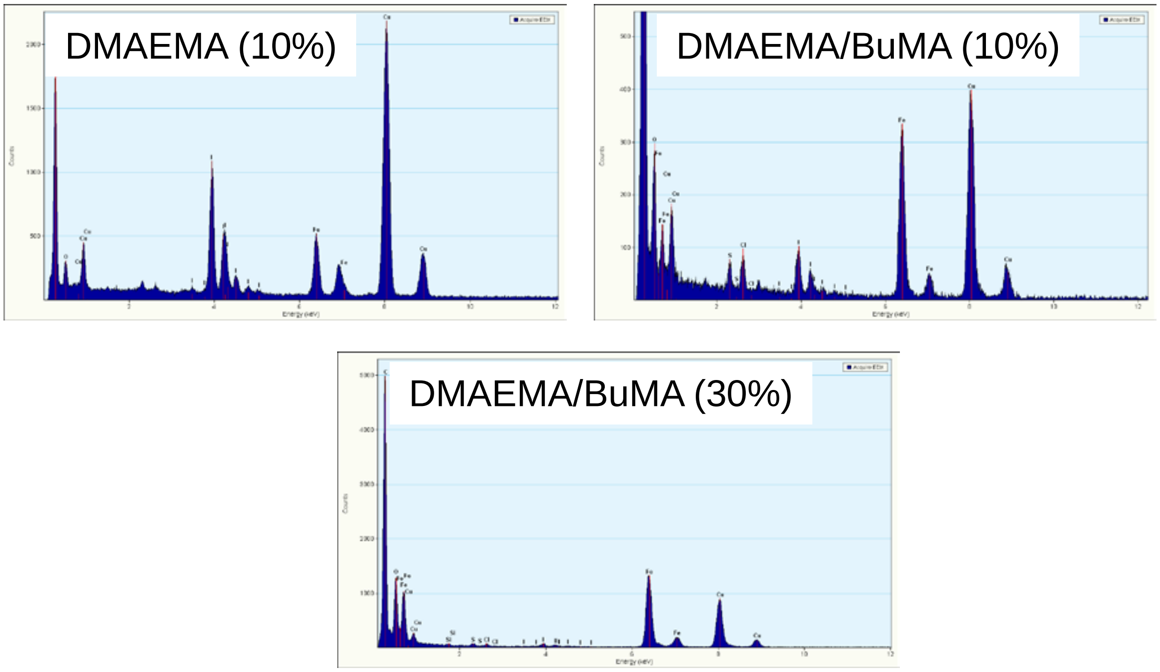
	Ntotal	Mean	Standard Deviation	Minimum	Maximum
B	153	7.02204	2.38228	3.30346	15.06

DMAEMA/BuMA (30%)

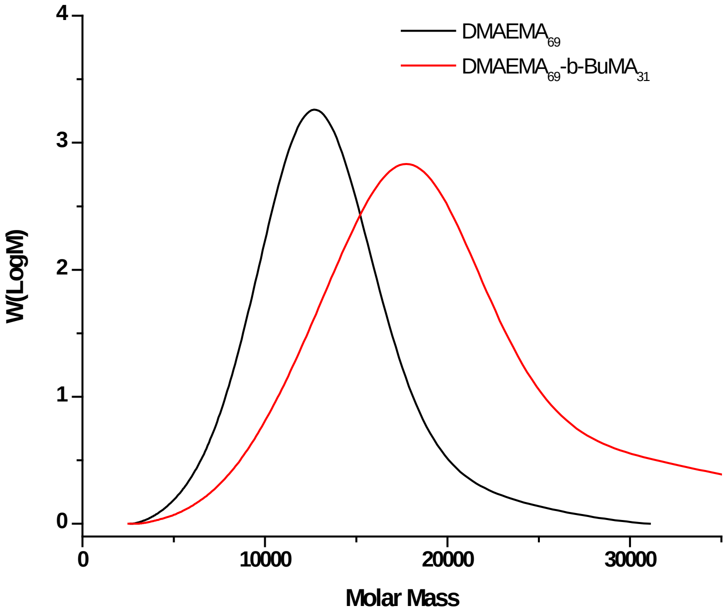


	Ntotal	Mean	Standard Deviation	Minimum	Maximum
B	112	7.28253	2.27419	2.80371	13.1406

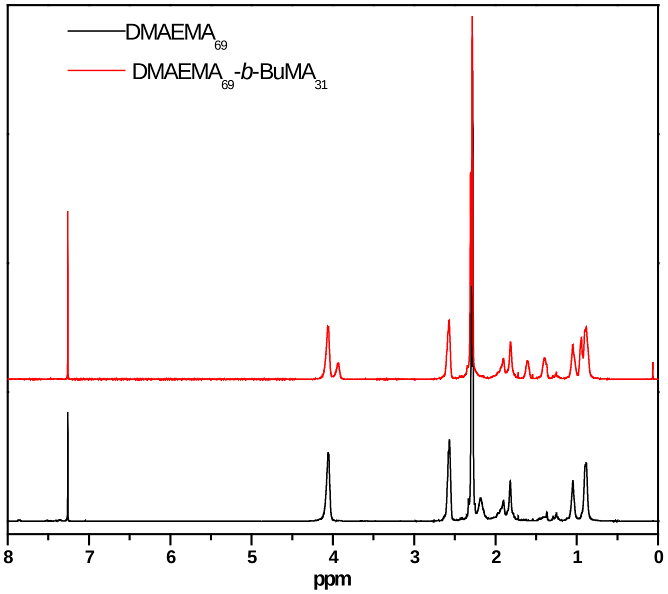
Supplementary Figure 2. EDX spectra of the nanocomposite (co)networks.



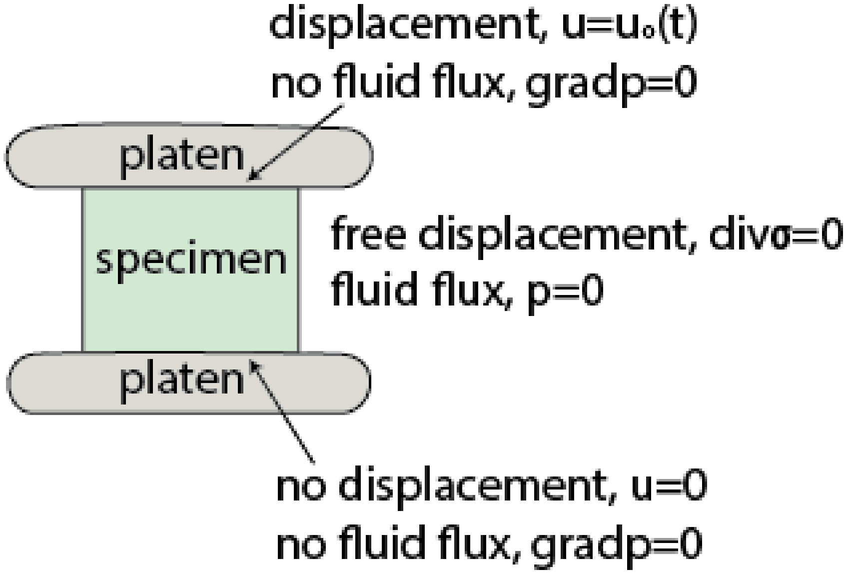
Supplementary Figure 3. Size Exclusion Chromatography (SEC) eluorgrams of $DMAEMA_{69}$ and $DMAEMA_{69}$-b-$BuMA_{31}$ linear precursors to the (co)networks.



Supplementary Figure 4. ^{1}H NMR spectra of the DMAEMA$_{69}$ homopolymer and the DMAEMA$_{69}$-b-BuMa$_{31}$ block copolymer, linear precursors to the networks.



Supplementary Figure 5. Boundary conditions employed for the simulation of the unconfined, stress relaxation experiment. At the top side of the specimen, there is no fluid flux (pressure gradient is zero) and the displacement, u, that determines the four cycles of the stress relaxation experiment is applied. At the bottom side, there is no displacement and no fluid flux. At the sides, the specimen is allowed to deform freely (divergence of stress tensor is zero) and there is fluid flux (fluid pressure is zero).



Supplementary Table 1. Statistical analysis for the Young's Modulus of the different groups.

p-test for DMAEMA homopolymer networks	
Tested pairs	**p-value**
0%-10%	0.011
0%-30%	0.0003
10%-30%	0.004

p-test for DMAEMA-b-BuMA diblock copolymer networks	
Tested pairs	**p-value**
0%-10%	0.006
0%-30%	0.018
10%-30%	0.051

P-Test between the two polymeric groups	
Tested pairs	**p-value**
0%-0%_b	0.004
10%-10%_b	0.008
30%-30%_b	0.035

References

1. V. C. Mow, S. C. Kuei, W. M. Lai and C. G. Armstrong, *J Biomech Eng-T Asme*, 1980, 102, 73-84.
2. Biot, *J Appl Phys*, 1941, 12, 155-164.
3. R. L. Spilker, J. C. Nickel and L. R. Iwasaki, *Ann Biomed Eng*, 2009, 37, 1152-1164.