**Figure S1.** Typical stress-strain curves of DMA/C17.3M hydrogels under compression as the dependences of the nominal $\sigma_{\text{nom}}$ (A) and true stresses $\sigma_{\text{true}}$ (B) on the compression ratio $\lambda$. The dashed red arrow illustrates calculation of the real fracture stress for the hydrogel containing 50 mol% C17.3M from the maximum in $\sigma_{\text{true}}$ vs $\lambda$ curve. The corrected $\sigma_{\text{nom}}$ vs $\lambda$ curves up to the fracture point are shown in C.
**Fig. S2.** Gel fraction \( W_g \) and equilibrium water content (\( H_2O \% \)) of the hydrogels plotted against their hydrophobe content.

**Fig. S3.** DSC traces of the hydrogels. The type of the hydrogels and their hydrophobe content are indicated.
Fig. S4. $G'$ (filled symbols), and $G''$ (open symbols) of the hydrogels during the heating - cooling cycle between 5 and 80 °C. The type of the hydrogels and their hydrophobe content indicated. $\omega = 6.28$ rad.s$^{-1}$, $\gamma_0 = 0.001$. 
Fig. S5  $G'$ (filled symbols) and $G''$ (open symbols) of the hydrogels with 20, 30, and 50 mol\% hydrophobe at 5 and 80 °C shown as a function of the frequency $\omega$. $\gamma_0 = 0.001$. 
**Fig. S6.** Young’s modulus $E$, compressive fracture stress $\sigma_f$, fracture strain $\varepsilon_f$, and toughness $W$ of the hydrogels plotted against the fraction $f_{cry}$ of units in crystalline domains. Hydrogels: DMA/C17.3M ($\triangle$), DMA/C18A ($\blacksquare$), AAc/C18A ($\bullet$).

**Figure S7.** (A): Two successive loading / unloading tensile cycles up a maximum elongation ratio $\lambda_{max}$ of 3.5 and 4.0 for the hydrogel samples indicated. Note that between each cycle, the gel samples were immersed in water at 70 °C. 1$^{st}$ and 2$^{nd}$ cycles are shown by the solid and dashed curves, respectively.