

Supporting Information

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Shape-Memory Semicrystalline Polymeric Materials Based on Various Rubbers

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Rubber type	$\chi$ parameter	Density / $g \cdot mL^{-1}$
SBR	$0.524 - 0.285 \ \phi_2 \ ^{[S1]}$	0.935
CBR	$0.19 + 1.13 \ \phi_2  ^{[S2]}$	0.90
NR	0.39 <sup>[S3]</sup>	0.92
IIR	$0.49 + 0.25 \ \phi_2 \ ^{[S4]}$	0.92

**Table S1.** Rubber-toluene interaction parameter  $\chi$ , and rubber densities.



Figure S1. FTIR spectra of the rubbers. The spectra confirm their chemical structures.



**Figure S2.** <sup>1</sup>H NMR spectra of NR-1 and NR-2. The methine proton shows a resonance signal at 5.14 ppm (a). The signal at 2.1 ppm is due to the methylene protons (c, c') and the signal of the methyl protons appears at 1.7 ppm (b). Unsaturation degree =  $\frac{a}{(c+c')/4} = 1.0$ . <sup>1</sup>H NMR was performed at room temperature using CDCl<sub>3</sub> as the solvent with TMS as internal standard.



Figure S3. <sup>1</sup>H NMR spectrum of CBR. The methine protons shows a signal at 5.4 ppm (a). The signal at 2.1 ppm is due to the methylene protons (b). Unsaturation degree  $=\frac{a/2}{b/4} = 0.99\pm0.01$ . <sup>1</sup>H NMR was performed at room temperature using CDCl<sub>3</sub> as the solvent with TMS as internal standard.



**Figure S4.** <sup>1</sup>H NMR spectrum of SBR. The phenyl protons show signals at around 7.2 ppm (c). <sup>1</sup>H NMR was performed at room temperature using CDCl<sub>3</sub> as the solvent with TMS as internal

standard. The signals at around 5.4 and 4.9 ppm are due to the protons of 1,4-cis and 1,4-trans butadiene units (b, b'), and the vinyl units (a), respectively. The signals between 0.8 and 2.7 ppm are due to the methyl and methylene units. The spectrum of CDCl<sub>3</sub> containing TMS is shown in the right panel. The unsaturation degree was calculated by,<sup>[S5]</sup>

Unsaturation = 
$$\frac{(A+B)/2}{(A+B)/2 + C_{calib}/5}$$
(S1)

where A and B represent the signal intensity ranges from 4.3 ppm to minimum intensity point around 5.0 ppm, and from around 5.0 ppm to minimum intensity point around 6.1 ppm, respectively, and  $C_{calib}$  is given by, <sup>[S5]</sup>

$$C_{calib} = C - CD_{blank} \frac{TMS}{TMS_{blank}}$$
(S2)

where C is the intensity range from 6.1 ppm to 7.7 ppm,  $TMS_{blank}$  is the integrated signal intensity of TMS in CDCl<sub>3</sub> containing TMS,  $CD_{blank}$  is the intensity from 6.1 ppm to 7.7 ppm in CDCl<sub>3</sub> containing TMS, and TMS is the integrated signal intensity of TMS in SBR sample solution. The unsaturation degree of SBR was calculated as  $84\pm5$  %.



**Figure S5.** (a): TEM images of the c-IPN with 30 wt% NR-1. Scale bars: 500 nm and 1µm. (b): STEM image of the same c-IPN and its EDX maps. Scale bars: 2 µm.



Figure S6. Frequency ( $\omega$ ) dependences of G' (symbols), and tan  $\delta$  (curves) of c-IPNs at 25 and 65 °C. The type of rubbers indicated.  $\gamma_0 = 0.1\%$ .



**Figure S7.** The dependence of the fractures stress  $\sigma_f$  and elongation to break  $\sigma_f$  of c-IPNs on the type of the rubber. The rubber contents are indicated.



**Figure S8.** Scheme of the interconnected interpenetrating rubber and PC18A networks (c-IPN), and the individual network components.

## References

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