## Autonomous self-healing polyisoprene elastomer with high modulus and good toughness based on synergy of dynamic ionic crosslinks and highly disordered crystals

Yohei Miwa,<sup>1,†,\*</sup> Mayu Yamada,<sup>1</sup> Yu Shinke,<sup>2</sup> and Shoichi Kutsumizu<sup>1</sup>

<sup>1</sup>Department of Chemistry and Biomolecular Science, Faculty of Engineering, Gifu

University, Yanagido, Gifu 501-1193, Japan.

<sup>2</sup>The Yokohama Rubber Co., Ltd., Hiratsuka, 254-8601, Japan.

<sup>†</sup>PRESTO, Japan Science and Technology Agency.

\*Author to whom correspondence should be addressed.

E-mail: y\_miwa@gifu-u.ac.jp

**Table of Contents** 

## <Supporting Figures>

Fig. S1. <sup>13</sup>C-NMR spectrum for *cis*-PI used in this work.

**Fig. S2.** FT–IR spectra for the indicated samples. Bands at 1708 cm<sup>-1</sup> and 1585 cm<sup>-1</sup> are assigned to v(C=O) of carboxy and  $v(O-C-O^{-})$  of sodium carboxylate, respectively.

Fig. S3. First heating and first cooling processes of CPI/TPI blend containing 20wt% of TPI.

Eq. S1. Calculation of penetration depth of infrared rays in the ATR FT-IR measurement



Fig. S1. <sup>13</sup>C-NMR spectrum for *cis*-PI used in this work.



**Fig. S2.** FT–IR spectra for the indicated samples. Bands at 1708 cm<sup>-1</sup> and 1585 cm<sup>-1</sup> are assigned to v(C=O) of carboxy and  $v(O - C - O^{-})$  of sodium



**Fig. S3.** First heating and first cooling processes of CPI/TPI blend containing 20wt% of TPI.

$$d_p = \frac{\lambda}{2\pi \sqrt{n_1^2 \sin^2 \theta_1 - n_2^2}} \qquad (S1)$$

*d*<sub>p</sub>: Penetration Depth

- $\lambda$ : Wavelength of Infrared
- $\theta_1$ : Incidence Angle (= 45°)
- $n_1$ : Refractive Index of Diamond (= 2.42)
- $n_2$ : Refractive Index of PI (= 1.51)

Eq. S1. Calculation of penetration depth of infrared rays in the ATR FT-IR measurement