***Supply Information***

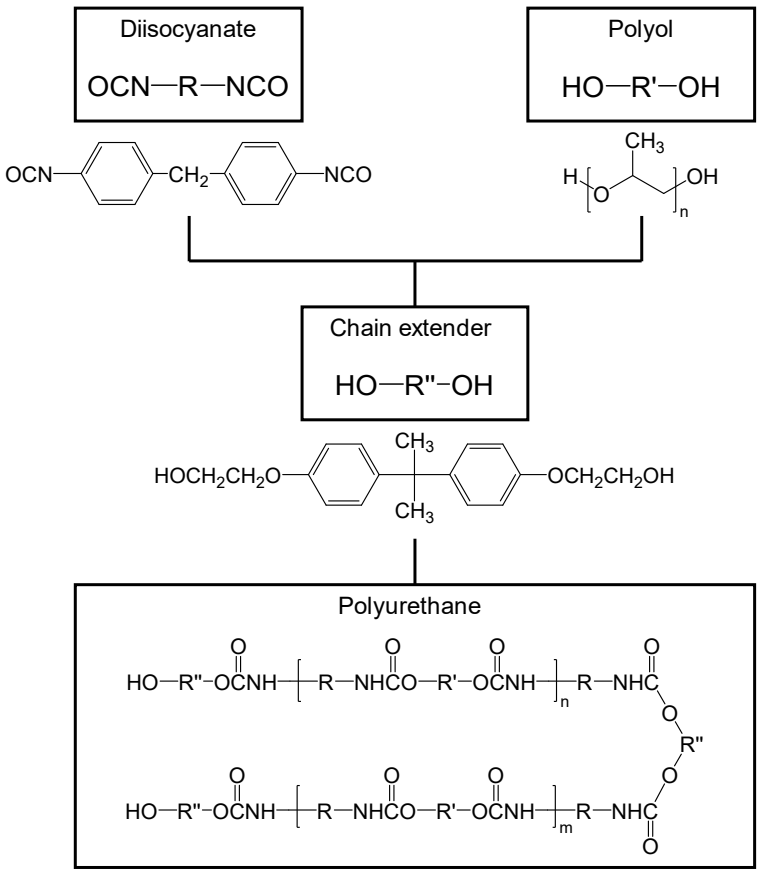


**Fig. S1** Synthetic rout and plausible chemical structure of SMP inferred from FT-IR (Fig. 1(b)) and previous patents,[26,27] where hard segments, composed of MDI and reactants of bisphenol A and ethylene oxide, connected to soft segments of polypropylene glycol.

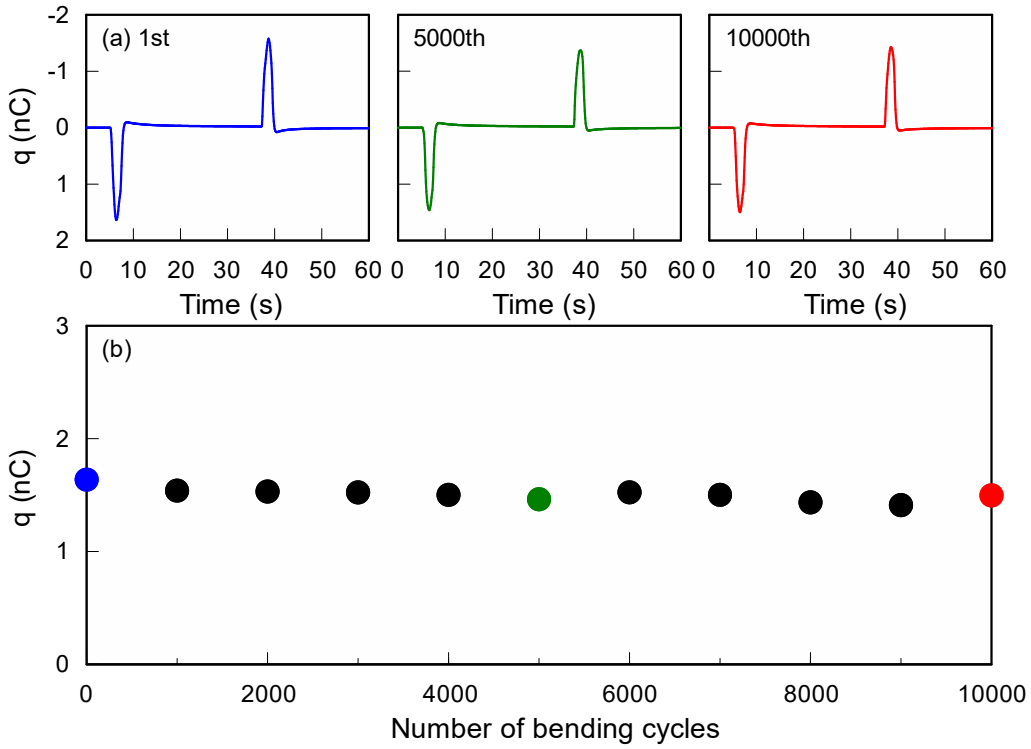


**Fig. S2** (a) Time dependence of $q$ generated at the 1st (blue), 5000th (green), and 10000th (red) bending at $d$ = 7 mm and $v$ = 3.3 mm $s^{-1}$ and (b) change in $q$ with number of bending cycles.

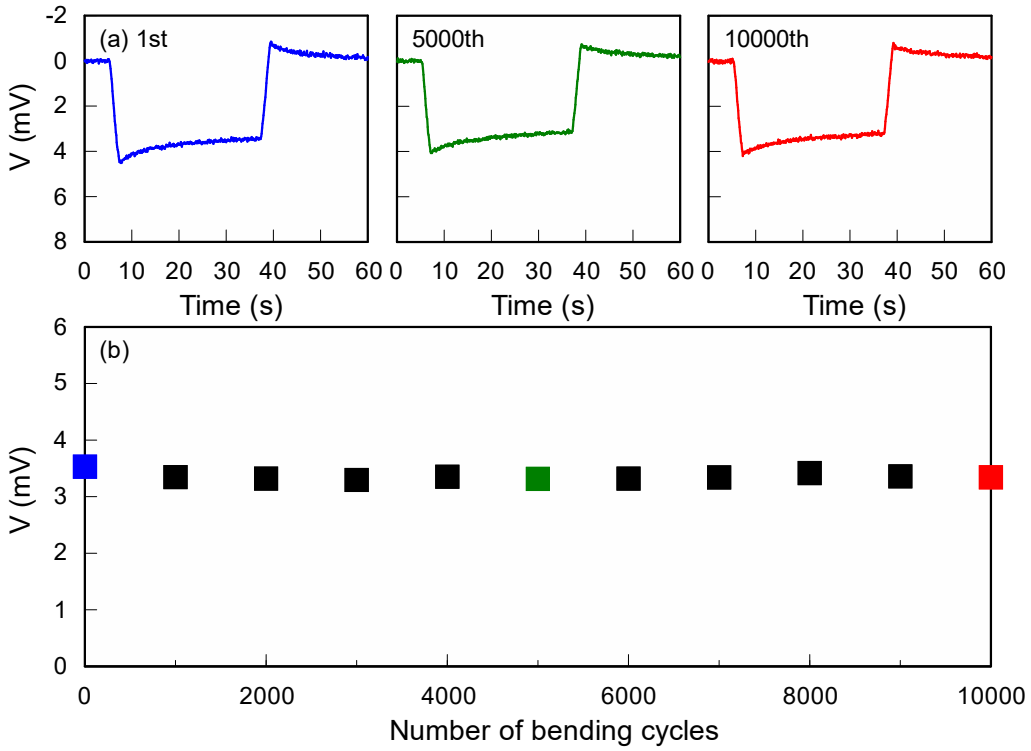


**Fig. S3** (a) Time dependence of $V$ generated at the 1st (blue), 5000th (green), and 10000th (red) bending at $d$ = 7 mm and $v$ = 3.3 mm $s^{-1}$ and (b) change in $V$ with number of bending cycles.

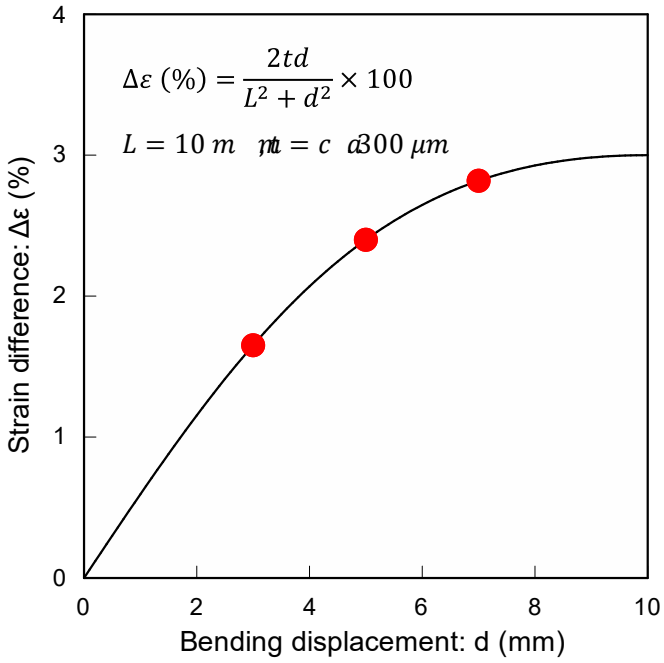


**Fig. S4** Relation between bending displacement ($d$) and strain difference ($\Delta\varepsilon$) of ionic SMP gel sensor ($L$ = 10 mm and $t$ = *ca*. 300 µm).

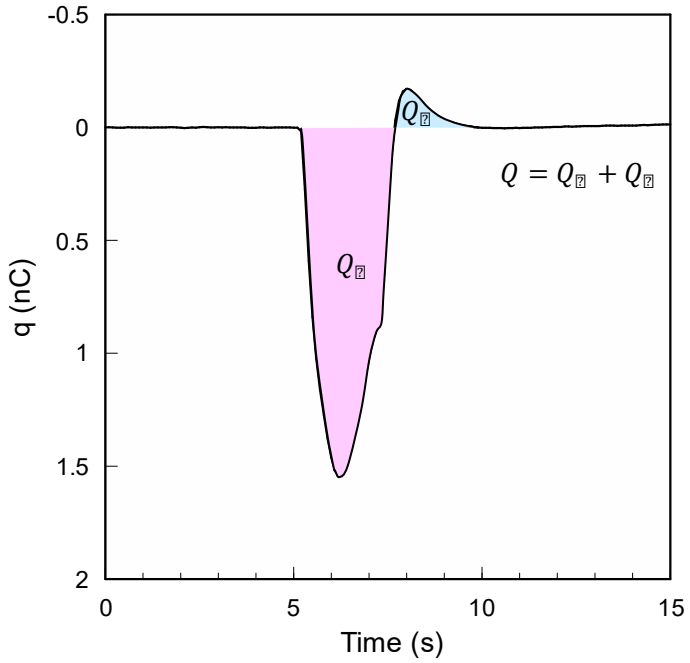


**Fig. S5** Calculations of overall net positive charge ($Q_+$), overall net negative charge ($Q_-$), and total charge ($Q = Q_+ + Q_-$) stored in the ionic SMP gel ($W_{IL}$ = 25 wt%) under bending deformation ($d$ = 7 mm and $v$ = 3.3 mm $s^{-1}$).

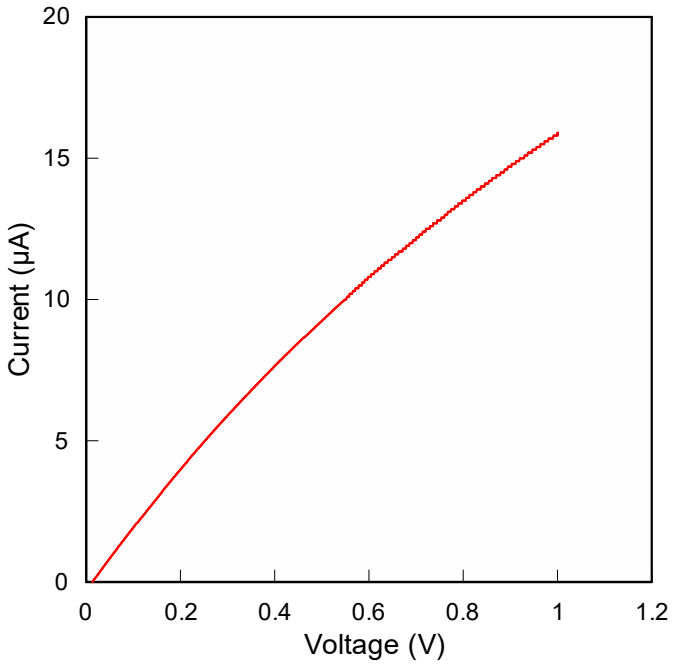


**Fig. S6** Current-voltage curve of ionic SMP gel sensor ($W_{IL}$ = 25 wt%) measured at a sweeping rate of 10 mV $s^{-1}$ with an electrochemical impedance system (1255WB, Solartron) and capacitance was evaluated by CorrView software (Solartron).