Electronic Supplementary Information (ESI)

A crown-ether-based moldable supramolecular gel with unusual mechanical properties and controllable electrical conductivity prepared by cation-mediated cross-linking

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Scheme S1. Synthetic routes for the precursors 1 and 2.



Fig. S1. Photographs of supramolecular gels taken over time.



Fig. S2 Photographs of gelation process of **1** and **2** solutions in DMSO (0.8 mL) at (A) 1:1 (20 mg of **1**, 11.68 mg of **2**), (B) 1:1.5 (20 mg of **1**, 17.52 mg of **2**), (C) 1:2 (20 mg of **1**, 23.36 mg of **2**), and (D) 1:3 (20 mg of **1**, 35.05 mg of **2**) of molar ratios.



Fig. S3 Time-dependent ¹H NMR spectra measured during the gelation of the solution of **1** (20 mg) and **2** (23.36 mg) in DMSO (0.8 mL).



Fig. S4. IR spectra of (A) 1, (B) 2 and (C) xerogel prepared with 1 and 2.



Fig. S5. (A) ¹H NMR spectrum of 1 and 2 in methanol and that measured (B) after 4 h.



Fig. S6 Photographs of the supramolecular gels prepared from the precursors (20 mg of 1 and 23.36 mg of 2) and (A) 0.25 equiv., (B) 0.5 equiv., (C) 1 equiv., and (D) 1.5 equiv. of Cs^+ (with respect to 1) in DMSO (0.8 mL).



Fig. S7 SEM images of the supramolecular gels prepared from the precursors **1** (20 mg) and **2** (23.36 mg) at 1:2 molar ratio (A) without and (B) with Cs⁺ (1 equiv. with respect to **1**).



Fig. S8 ¹H NMR spectra of DMSO solutions of 1 and 2 (2.50 mg of 1 and 2.92 mg of 2 in 0.5 ml of DMSO) in the absence and presence of various equiv. of Cs^+ .



Fig. S9 Rheology data of strain sweep tests for the supramolecular gels prepared from various molar ratios of the precursors 1 and 2 ((A) 1:1, (B) 1:1.5, (C) 1:2, and (D) 1:3).



Fig. S10 Rheology data of frequency sweep tests for the supramolecular gels prepared from various molar ratios of the precursors **1** and **2** ((A) 1:1, (B) 1:1.5, (C) 1:2, and (D) 1:3).



Fig. S11 Rheology data of continuous step strain tests measured at 0.1% and 100% for the supramolecular gels prepared from various molar ratios of the precursors **1** and **2** ((A) 1:1, (B) 1:1.5, (C) 1:2, and (D) 1:3).



Fig. S12 Rheology data of strain sweep tests for the supramolecular gels prepared from the precursors (1:2 molar ratio) in the presence of (A) 0.25 equiv., (B) 0.5 equiv., (C) 1 equiv., and (D) 1.5 equiv. of Cs^+ (with respect to 1).



Fig. S13 Rheology data of frequency sweep tests for the supramolecular gels prepared from the precursors (1:2 molar ratio) in the presence of (A) 0.25 equiv., (B) 0.5 equiv., (C) 1 equiv., and (D) 1.5 equiv. of Cs⁺ (with respect to **1**).



Fig. S14 Rheology data of continuous step strain tests measured at 0.1% and 100% for the supramolecular gels prepared from the precursors (1:2 molar ratio) in the presence of (A) 0.25 equiv., (B) 0.5 equiv., (C) 1 equiv., and (D) 1.5 equiv. of Cs^+ (with respect to 1).



Fig. S15. Rheological data of the supramolecular gel prepared with 1 equiv. of K^+ with respect to 1. ((A) Strain sweep, (B) frequency sweep, and (C) continuous step strain tests.)



Fig. S16 Photograph of the electrical circuit comprising a light-emitting diode (LED), a power source composed of two 1.5 V dry cells, and the cylinder-shaped supramolecular gel prepared from 1:2 molar ratio of **1** and **2** (cross-sectional area: 0.636 cm², length: 1.3 cm).



Fig. S17 (A) Photographs of the electrical circuits comprising the multimeter-connected **Gel-Cs**⁺ prepared with (a) 0.5 equiv., (b) 1.0 equiv., (c) 1.5 equiv., and (d) 2.0 equiv. of Cs⁺. Graphs of (B) resistance values and (C) electrical conductivities of **Gel-Cs**⁺ samples.



Fig. S18. Measurements of resistances of the four samples for calculation of their electrical conductivities. (A) Cs^+ in DMSO, (B) Cs^+ and 1 in DMSO, (C) Cs^+ and 2 in DMSO, (D) 1 and 2 in methanol.