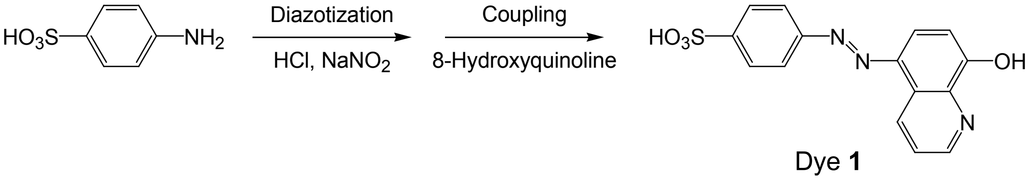
Supplementary Information

Materials. The chemicals used in the synthesis, 8-hydroxyquinoline, sulfanilic acid, γ-cyclodextrin, HCl (37%), $NaNO_2$, sodium acetate, HEPES (4-(2-hydroxyethyl)-1-piperazineethanesulfonic acid), D_2O, and DMSO-d_6, were of laboratory reagent grade.

Dye 1 synthesis. Sulfanilic acid (1.221 g, 7.06 mmol) was dissolved in water (20 mL), and HCl (37 %, 1.47 mL) was added while stirring. The temperature was lowered to 0-5 ^{o}C, and a 5% $NaNO_2$ aqueous solution (7.06 mmol, 10 mL) was added dropwise. The solution was stirred for an hour at 0-5 ^{o}C to complete the diazotization process. 8-Hydroxyquinoline (1.025 g, 7.06 mmol) in 0.5 N NaOH (16 mL) was added dropwise for 1 hr and stirring continued at room temperature overnight. After the reaction, the pH was adjusted to 7.5~8.0 using a dilute sodium acetate solution. The dye precipitate was then collected by filtration. The crude product was recrystallized three times in water (50 mL) and then dried in a vacuum oven (yield 35 %). 1H NMR (500 MHz, DMSO-d6): 9.3 (d, 1H), 9.0 (m, 1H), 9 (d, 2H), 7.8 (d, 2H), 7.7 (q, 2H), 7.2 (d, 1H).



Scheme S1. Synthesis of dye **1**.

Characterization of hydrogel.

POM measurement. Before loading the sample, both the slide and cover glasses were washed with acetone twice and dried at 90 °C (2 minutes). Several drops of warmed hydrogel were applied between two slides, and this was observed using a DFC295 digital camera (Leica).

2D-ROESY and HR-MAS. The 2D-ROESY spectrum was measured on a Bruker Avance II 900 (900 MHz, Korea Basic Science Institute), with a relaxation delay of 2 sec and a ROESY spinlock time of 250 ms. HR-MAS NMR experiments were recorded on a Varian UI500 (500 MHz, Korea Basic Science Institute) at a sample spinning speed of 2300 Hz. All NMR measurements were carried out at 298 K using D_2O solvent.

SAXS. Small-angle X-ray scattering experiments were performed on a multi-purpose X-ray diffractometer (X'pert Pro MPD, PANalytical, Korea Basic Science Institute) with Cu Ka radiation (λ=0.15406 nm, 3 KW). All measurements were made at a fixed temperature of 25 °C.

SEM. The gel samples of the γ-CD-dye **1**-LiCl system were dried overnight under a vacuum. After sputter-coating the samples with gold, SEM images were obtained with a Hitachi S-4800 scanning electron microscope.

TEM. For visualization by TEM, a sample was prepared by placing one drop of the solution onto a carbon-coated copper grid, and this was then dried overnight at room temperature. The measurements were taken by Field Emission Transmission Electron Microscopy (JEM-2100F, Jeol).

FT-IR. Samples of the dried gel and the inclusion complex of γ-CD and dye **1** were measured by an Avatar 370 FT-IR spectrometer. All measurements were made after the samples were vacuum-dried for a week.

ICD. Induced circular dichroism (ICD) spectra were recorded on a Jasco J-815 in a HEPES buffer solution (pH 7.5) with a 0.1 mm path length and a 1 nm spectral bandwidth at room temperature. All measurements were made at 25 ^{o}C.

Rheological Measurement. Rheological measurements were conducted using an Advanced Rheometric Expansion System (ARES) (Rheometric Scientific, USA) equipped with a parallel-plate fixture having a radius of 12.5 mm and a gap size of 1.0 mm. Before loading the sample, two plates were covered with a sandpaper in order to remove a wall slippage between the material and the plates. Before each measurement, a fresh sample was applied and stood still for 10 minutes, as a measure to ensure complete relaxation and temperature equilibrium. All measurements were made at 20 °C, if not otherwise stated.

Ionic Conductivity Measurement. Conductivity measurements were performed using a conductivity meter (Accumet, XL 20) at room temperature. Two conductivity probes (Accumet, cell constants 1.0 cm^{-1} and 10 cm^{-1}) were properly selected depending on the conductivity values of samples. The probes were calibrated with calibration standards (Fisher Scientific) before measurements. After each use the probes were cleaned with nitric acid, rinsed with deionized water and dried.

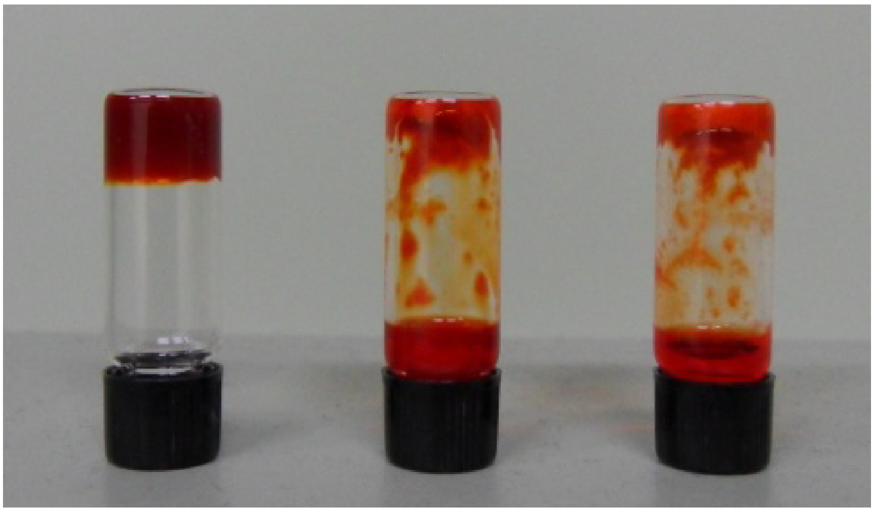


Figure S1. Photographic images of the inclusion complexes with different CDs (γ-CD, α-CD, β-CD, from left to right). All concentrations of dye **1**, CDs, and LiCl are fixed at 60 mM.

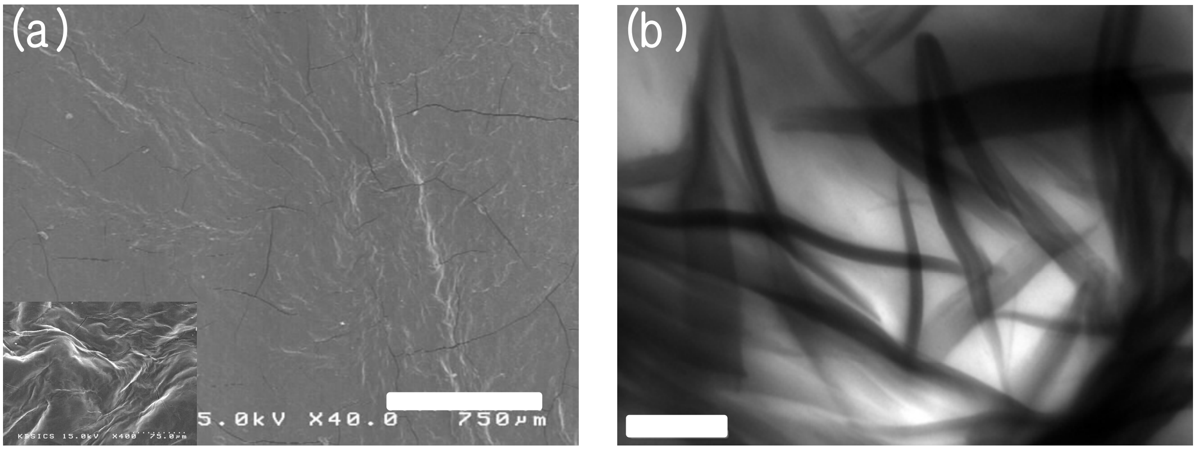


Figure S2. (a) SEM images at lower and higher (inset) magnifications, and (b) TEM image of the hydrogel. The concentrations of dye **1**, CDs, and LiCl are fixed at 60 mM. Scale bars are 750 μm (left) and 100 nm (right), respectively.

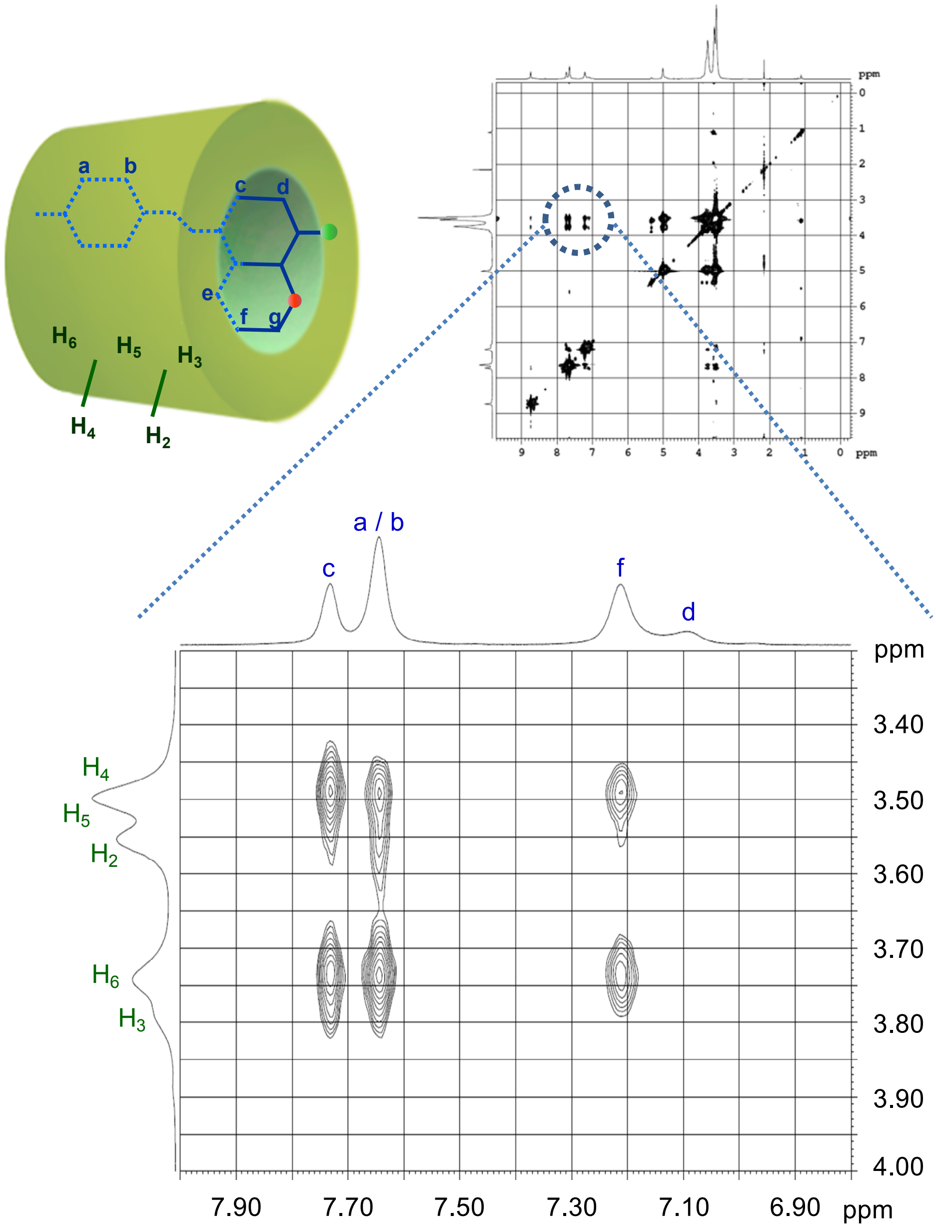


Figure S3. ROESY spectrum (spinlock time: 250 ms) of the hydrogel. The concentrations of dye **1**, γ-CD, and LiCl are fixed at 60 mM. All measurements were made using D_2O as a solvent at 25 °C. The possible binding mode of the inclusion complex is also shown.

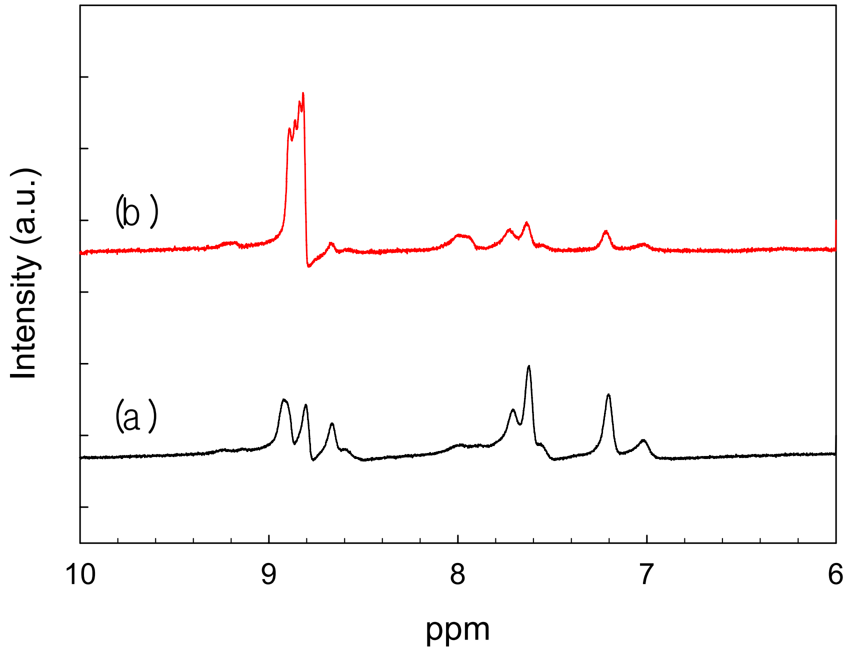


Figure S4. ^{1}H HR-MAS NMR spectra of the complexes in the (a) absence and (b) presence of LiCl. The concentrations of dye **1**, CDs, and LiCl are fixed at 60 mM. Measurements were made using D_2O as a solvent at 25 °C.

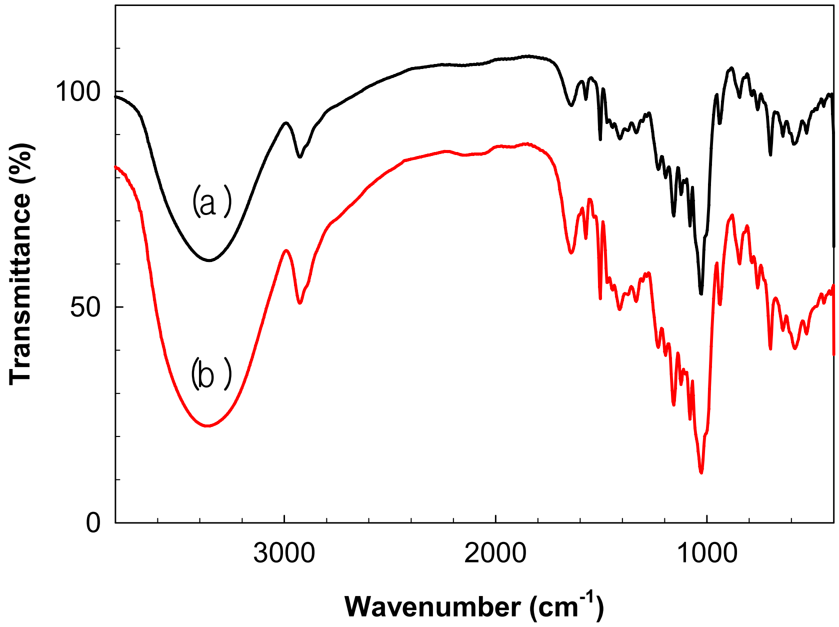


Figure S5. FT-IR spectra in the regions of 3800 ~ 400 cm^{-1} of the complex in the (a) absence and (b) presence of LiCl. The concentrations of dye **1**, CDs, and LiCl are fixed at 60 mM. Measurements were made after vacuum-drying the samples for a week.

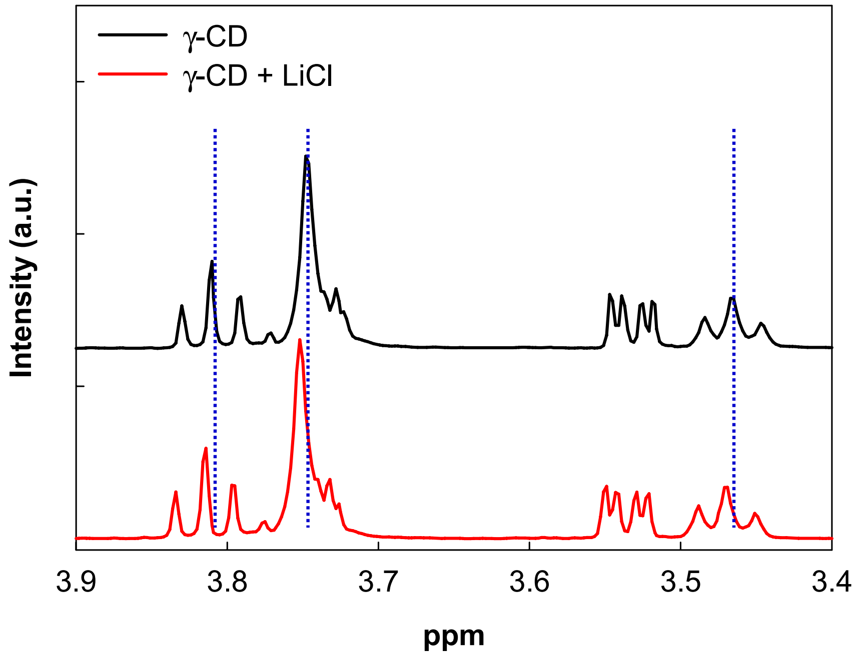


Figure S6. Partial ^{1}H-NMR spectra of the complex in the (a) absence and (b) presence of LiCl. The concentrations of all chemicals were fixed at 60 mM. Measurements were made at 25 °C using D_2O as a solvent.

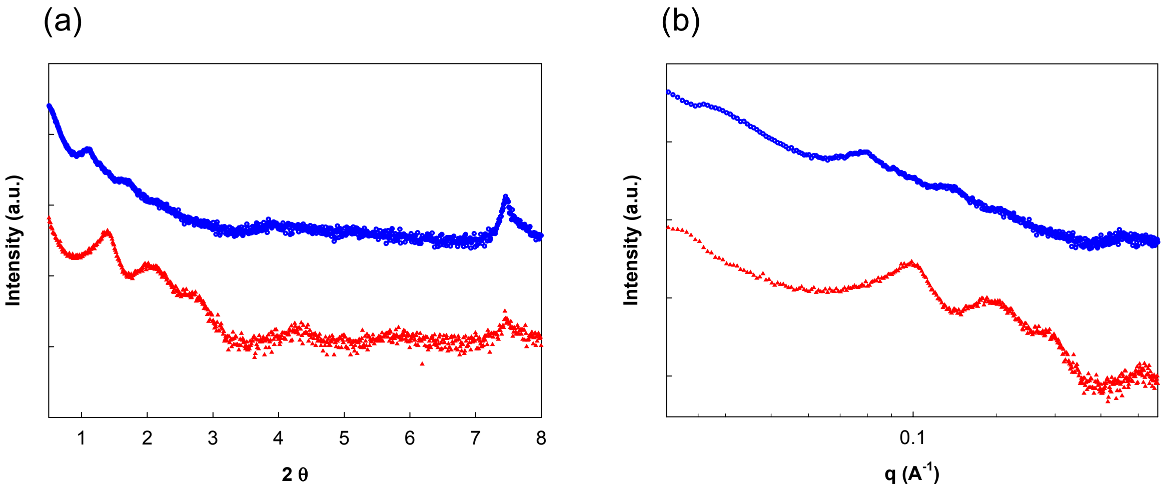


Figure S7. (a) Small-angle X-ray scattering (SAXS) data of the complexes in the range of 2θ with different LiCl concentrations of 1.2 mM (red, lower) and 60 mM (blue, upper), and (b) SAXS data of the complexes in regard to the *q* vector. The complexes consist of dye **1** (60 mM) and γ-CD (60 mM) in both cases.

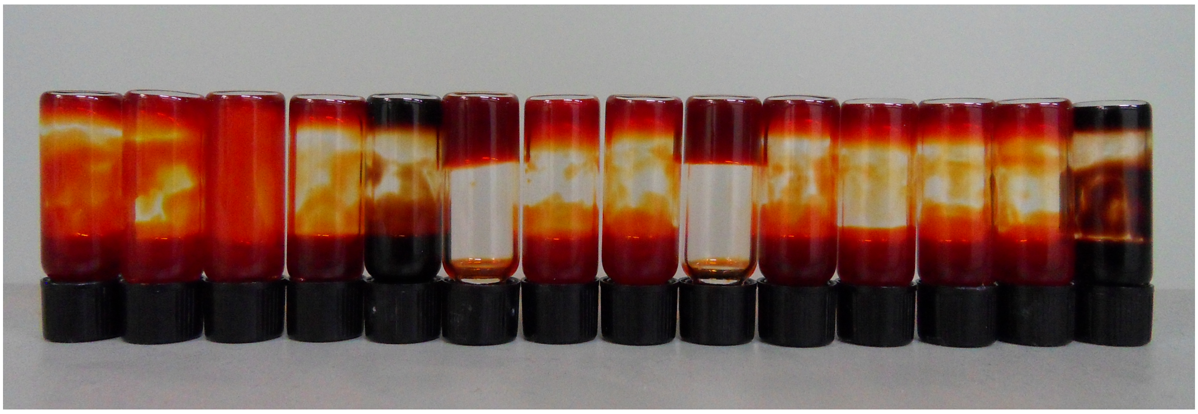


Figure S8. Photographic images, upon the addition of metal ions, of the complex. The concentrations of dye **1**, γ-CD, and metal ions are fixed at 60 mM. The order of the addition of metal ions was as follows: none, Al^{3+}, Cr^{3+}, Cu^{2+}, Fe^{3+}, Li^{+}, Mg^{2+}, Pb^{2+}, Ca^{2+}, Ni^{2+}, Zn^{2+}, Cd^{2+}, Hg^{2+}, and Ru^{3+} (left~right). All solutions were prepared in HEPES buffer solution (pH 7.5). Calcium shows an effect similar to that of lithium, but only at lower temperature.