Supporting Information:

**Experimental Section**

N-isopropylacrylamide (NIPA), \( N,N' \)-methylenebis(acrylamide) (bis), hydrogen peroxide, ammonium peroxodisulfate (APS) and \( N,N,N',N' \)-tetramethylethylenediamine (TEMED) were purchased from Wako Pure Chemical Industries, Ltd. An oil-soluble dye, Yellow AB, was obtained from Tokyo Kasei Kogyo Co., Ltd. All the chemicals were used without further purification. DGI was synthesized by essentially the same procedures as those in the previous work\[^{[16]}\]. The crude product was purified by silica gel column (Silica Gel 60N, KANTO Chemical Co., Inc., Tokyo). The final reaction product was checked by element analysis to contain 64.39 % C (64.48 % theoretical), 9.83 % H (9.74 % theoretical). Ionic surfactant SDS with the purity of higher than 99.0 % was obtained from MP Biomedicals, Inc., Germany. Millipore deionized water bubbled by argon for 15 min was used. The schematic procedure for the synthesis of PDGI-PNIPA gels are depicted in S-Figure 1. In a typical experiment, PDGI-PNIPA hybrid gels were prepared as follows: the iridescent mesophase of DGI were prepared with the DGI and SDS concentrations of 2.03 and 0.03 wt %, respectively. The mesophase of DGI and SDS were kept at 55 °C for one day to obtain the iridescent color since the Krafft point of DGI is 43 °C. The iridescent sample was then polymerized with hydrogen peroxide as an initiator by UV radiation for 15 min. The molecular weight of PDGI was checked by gel permeation chromatography to be \( M_w= 80,000 \), \( M_w/M_n=2.07 \). After polymerization, the iridescent color remained even for a long time at room temperature. 210 mg NIPA, 7.2 mg APS and 8 mg bis were mixed together and dissolved into 1 g of deionized water. The above solution was then mixed with 4 g of polymerized DGI mesophase (where PDGI is 1.63 wt % and SDS is 1/40 molar ratio respect to DGI) after cooling down to room temperature. After 3 h standing, 15 µl TEMED was added and the mixture was kept at 4 °C for 1 h to complete polymerization. The obtained hybrid gels turn turbid after polymerization, which may be ascribed to some extent of micro phase separation occurs in this system. This phase separation between PDGI bilayer membranes and PNIPA gels may facilitate the releasing of water during the shrinking process.

The samples used for shrinking and swelling experiments were prepared in capillaries with the inner diameter of 1.3 mm. PNIPA gels were similarly synthesized without PDGI. After preparation, all the samples were immersed in deionized water for a week to remove the added SDS and unreacted monomers, etc.. For the samples prepared in the capillaries, shrinking and swelling procedures were repeated several times in order to take out the samples.

The samples prepared in the capillaries were cut into about 10 mm long for shrinking and swelling tests. The volumes of these gels were calculated by \( V = \pi D^2 L/4 \), where \( D \) and \( L \) are the diameter and length of the cylindrical gels, respectivley. The samples for polariscope experiments were cut from the bulk gels of PDGI-PNIPA and PNIPA, respectively. The samples were put on a slide glass and observed between two
polarizers crossed each other. Laser confocal microscopy experiment was carried out at Nikon Imaging Center of Hokkaido University. The PDGI-PNIPA gel was cut and put on a slide glass. The images were observed and recorded by Nikon Eclipse TE 2000-E Spectral Imaging Confocal Microscopy at 100-times magnification with oil immersion.

S-Figure 1. Two steps synthesis of PDGI-PNIPA hybrid gel. DGI monomers and PDGI in water shows iridescent color because of the multi-bilayer onion-like structures. After polymerization of NIPA with cross-linker, PNIPA gel networks were synthesized in the space of the compartments between the onion-like PDGI bilayer membranes.

FF-TEM image for DGI onion-like vesicles

S-Figure 2. Freeze Fracture TEM image of iridescent DGI solution, the onion-like bilayer membranes with the diameter of about 20 µm can be clearly seen.
The calculation of the vacant space in DGI vesicle system

DGI forms lamellar sheets at low SDS concentration but lamellar vesicles at high SDS concentration (see Ref. 1). Taking the interplanar distance, \(d\), between DGI bilayer membranes to be \(d_1=220\) nm (1.63 wt \% DGI, 1/800 molar ratio of SDS to DGI, lamellar sheets) and \(d_2=180\) nm (1.63 wt \% DGI, 1/40 molar ratio of SDS to DGI, onion-like lamellar vesicles, the same conditions as that used in this work after mixed with NIPA monomers) into account, we can calculate the weight fraction of DGI in both lamellar sheets and vesicles solutions as follows:

\[
c_1 = \frac{s \cdot \rho_s \cdot \delta}{s \cdot \rho_s \cdot \delta + s \cdot \rho_w \cdot (d_1 - \delta)}
\]

\[
c_2 = \frac{s \cdot \rho_s \cdot \delta}{s \cdot \rho_s \cdot \delta + s \cdot \rho_w \cdot (d_2 - \delta) + \frac{R}{N} \cdot \rho_w}
\]

\[c_1 = c_2, \quad R/N = d_1 - d_2 = 40\, \text{nm}\]

where, \(c_1\) and \(c_2\) are the weight fractions of DGI in water; \(\rho_s\) and \(\rho_w\) the densities of surfactant layer and water, respectively; \(\delta\) the thickness of bilayer membranes; \(N\) the average number of bilayer membranes; \(R\) the average distance between the onion-like vesicles.

Since the size of the vesicle is in several tens of \(\mu m\), while the interplanar distance between the bilayers is less than 200 nm, we can take the curved bilayer membranes as flat ones to make the following calculation. Then the vacant volume is:

\[
V\% = \frac{R \cdot s}{N \cdot d \cdot s} = 40 / 220 = 18.2\%.
\]
of the total gel. The gel segmented by the PGDI bilayers is not a small part, but higher than 80%.

The following images show the whole shrinking process of this PDGI-PNIPA hybrid gel. The PDGI-PNIPA gel, together with PNIPA gel, was put on the hot heating stage at 50 °C. The shrinkage of PDGI-PNIPA gel occurs and ends within 15 seconds, demonstrating the super-rapid shrinking behavior.

Elapsed time: 0 s

Elapsed time: 2 s
Elapsed time: 5 s

Elapsed time: 6 s
Elapsed time: 20 s

Elapsed time: 25 s
Elapsed time: 30 s

Elapsed time: 35 s
Elapsed time: 40 s

Elapsed time: 43 s
Elapsed time: 50 s