Electronic Supplementary Information

for

Cholesterol-based Low-molecular Mass Gelators towards Smart Ionogels

Junlin Yan,^a Jing Liu,^a Ping Jing,^a Chengkun Xu,^b Jiamin Wu,^b Di Gao^{*b} and Yu Fang^{*a}

^a Key Laboratory of Applied Surface and Colloid Chemistry of Ministry of Education, School of Chemistry and Materials Science, Shaanxi Normal University, Xi'an, P. R. China, 710062

^b Department of Chemical and Petroleum Engineering, University of Pittsburgh, Pittsburgh, Pennsylvania, USA, 15261

^{*} E-mail: yfang@snnu.edu.cn, gaod@pitt.edu



1. Schematic representation of the synthesis of the cholesteryl derivatives

Scheme S1. Schematic representation of synthesis of the cholesterol-based gelators 1-3



Scheme S2. Schematic representation of synthesis of compound 4

2. Concentration-dependent rheological behaviors of 1D/IL2 ionogel

To understand the rheological properties of the ionogels, G' and G'' were measured at different concentrations and frequencies by choosing 1D/IL2 gel as an example system (To be clear, only G' is shown in Figure S1). Reference to the traces shown in Figure S1a, it is seen that the value of G' increases from 229 Pa to 2.37×10^4 Pa along with increasing the gelator concentration from 0.1% to 1.0% (w/v), and correspondingly, the yield stress changes from 15.8 Pa to 630 Pa, indicating that both the stability of the gel network and the elastic property of the gel are well dependent upon the concentration of the gelator in the solvent. Frequency sweep is an important method to detect the tolerance performance of a material to external forces. Clearly, the G' values of the gels exhibit weak dependence on frequency when the concentration exceeds 0.1% (w/v), a typical viscoelastic behavior, suggesting that the ionogel shows good tolerance to external forces (c.f. Figure S1b). However, for the system containing of 0.1% (v/w) of the gelator, the value of G' is strongly dependent upon the frequency of the scanning process, particularly when the frequency is low. The value reaches a plateau for the scanning conducted between 2 Hz and 20 Hz. Reference to the results shown in Figure S2, it is revealed that the value of G' and G''remain roughly constant and the value of G' is almost one order greater than that of G'' as the shear stress is below a typical shear stress, the so called yield stress, which is a typical rheological behavior of a gel. Combing the results from the two measurements, it may be concluded that the system is a gel, but it shows poor tolerance to external forces. In particular, it may be a highly viscous fluid at room temperature when it is exposed to a low frequency shear stress.







Figure S2. The evolution of *G*' and *G*'' as functions of the applied shear stress of the **1D**/IL2 ionogel (0.1%, v/w)

3. Effect of external magnetic field upon the magnetic ionogel

Figure S3 displays optical micrographs of dispersed magnetic particles (diameter of about 1 μ m) in IL2 and **1D**/IL2 ionogel (0.5%, *w/v*). It is clearly seen that the magnetic particles (0.2 *wt* %) are randomly distributed in IL2 in the absence of a magnetic field (Figure S3a), whereas they self-assemble and align parallel to the direction of an applied magnetic field instantly (Figure S3b). However, when the particles were doped into the ionogel, aggregates formed during heating-cooling process. Presence of a magnetic field shows little effect upon the macroscopic property of the ionogel provided the density of the magnetic particle is less than 12% (*w/w*) (Figures S3c, S3d). However, when the particle density exceeds 20% (*w/w*), the ionogel becomes liquid instantly upon treatment of the magnetic field (Figure S3e and Figure S3f).



Figure S3. Optical microscopy images of dispersed Fe₃O₄ particles (about 1 μ m) in IL2 or **1D**/IL2 ionogel (0.5%, *w/v*). (a) 0.2 wt % of magnetic particles in IL2 in the absence or (b) presence of magnetic field, (c) 0.2 wt % of Fe₃O₄ particles in ionogel under magnetic field, (d) 10 wt % of Fe₃O₄ particles in ionogel under magnetic field, (e) 20 wt % Fe₃O₄ particles in ionogel in the absence or (f) presence of magnetic field. Scale bar, 20 μ m.



4. The FTIR spectra of 1D in CDCl₃ and 1D/IL2 xerogel

Figure S4. FTIR spectra of 1D in CDCl₃ (up, 0.4%, w/v) and 1D/IL2 xerogel (bottom, 1%, w/v)

5. Concentration dependence of the gel-to-sol temperature (T_{gel}) of 1D/IL2 ionogel

All the ionogels as obtained in the present study is thermo-reversible. T_{gel} , the gel-to-sol phase transition temperature, is a good parameter to stand for the thermostability of a gel system. As an example, the T_{gel} of **1D**/ionogel was measured at different concentrations of the gelator, **1D**. It is found that the T_{gel} of the system initially increases with increasing the gelator concentration, and then remains constant (~ 74 °C)as the concentration reaches and exceeds ca. 10 mg/mL.



Figure S5. Concentration dependence of the gel-to-sol transition temperature of 1D/IL2 ionogel

6. ¹³C NMR spectra of new compounds



Figure S8. ¹³C NMR spectrum of 1 (left) and 2 (right)



Figure S9. ¹³C NMR spectrum of 3 (left) and 4 (right)