Supporting Information

Experimental

Materials. Cell matrix type I-A, 0.3% collagen solution, was purchased from Nitta Gelatin Inc. (Osaka, Japan). *N,N*-Dimethylacrylamide (DMAAm) and glycidyl methacrylate (GMA) were purchased from Fujifilm Wako Pure Chemical Industries (Osaka, Japan) and purified by passing them through alumina column. Additionally, 3-Acrylamidophenylboronic acid (APBA) was purchased from Tokyo Chemical Industries (Tokyo, Japan) and used as received. Dopamine acrylamide (DA) and 4-acrylamido fluorescein (FLAAm) were synthesised as previously described.[1,2] The Dulbecco's Modified Eagle's Medium with low glucose (DMEM-LG), Dulbecco's phosphate-buffered saline (PBS), penicillin-streptomycin, and trypsin-ethylenediaminetetraacetic acid (trypsin-EDTA) were purchased from Sigma-Aldrich (St. Louis, USA). Trypsin-EDTA was diluted to 0.05 wt% trypsin and 0.02 wt% EDTA with PBS prior to use. Cell Tracker Red CMTPX and Cell Tracker Green CMFDA were purchased from Life Technologies (Carlsbad, USA). Calcein-AM was purchased from Dojin Chemicals (Kumamoto, Japan) and ethidium homodimer was purchased from Nippon Gene (Tokyo, Japan). All other reagents were purchased from Fujifilm Wako Pure Chemical Industries and used as received.

Synthesis of polymeric glues Poly (DMAAm-*co*-GMA-*co*-APBA) (PBA-PG) and poly (DMAAm-*co*-GMA-*co*-DA) (CAT-PG) were copolymerised using free-radical polymerisation. In the case of PBA-PG, DMAAm (1.6 mL, 15.7 mmol), GMA (0.24 mL, 1.9 mmol), APBA (0.18 mg, 0.93 mmol), and AIBN (32.8 mg, 0.20 mmol), as an initiator, were dissolved in mixed solvent (*N,N*-Dimethyl formamide (DMF)/ water = 19/1, 37 mL). In the case of CAT-PG, DMAAm (1.6 mL, 15.7 mmol), GMA (0.24 mL, 1.9 mmol), DA (0.19 mg, 0.92 mmol), and AIBN (32.8 mg, 0.20 mmol) were dissolved in DMF (39 mL). Using these solutions, each polymer was polymerised according to the following procedure. The reaction solution was deoxygenated by nitrogen gas bubbling for 30 min. Polymerisation was then performed at 70°C for 22 h. After the reaction, polymer solution was poured into diethyl ether to reprecipitate the polymer. The polymer obtained was dried at 25 °C under vacuum. ^{1}H-NMR measurements showed that these polymeric glues contained epoxide group and complementary interaction sites (phenyl boronic acid or catechol moieties) (Fig. S1). Then, PBA-PG and CAT-PG were dissolved into CD_3OD and DMSO-*d*6, respectively.

Mixing test PBA-PG and CAT-PG were diluted with DMEM-LG to 10 w/v%. After mixing these solutions, the mixed solution rapidly increased in viscosity and lost its fluidity in several minutes (Fig. S2).

Cell culture and staining of cells NIH/3T3 cells and Hep G2 cells were cultured on conventional tissue culture polystyrene (TCPS) dishes in DMEM supplemented with 100 U/mL penicillin, 100 mg/mL streptomycin, and 10% calf serum (NIH/3T3) or 10% fetal bovine serum (Hep G2) at 37°C in a humidified atmosphere with 5.0% CO_2. NIH/3T3 and Hep G2 cells were stained with Cell Tracker Green CMFDA and Cell Tracker® Red CMTPX, respectively, according to the instructions provided by the manufacturer. Stained cells were recovered from TCPS dishes by treatment with diluted trypsin-EDTA solution.

Preparation of collagen gels To prepare the collagen gel without encapsulating cells, the collagen solution (Cell matrix type I-A/10xPBS/80 mM NaOH aq. = 8/1/1 (v/v)) was poured into a 2-mm thick mould composed of a glass plate and silicon sheet, and kept at 37°C for 30 min. After that, collagen gel was taken from the mould and used as prepared or cut into roughly 3-mm size squares. For the cell-laden collagen gels, the same collagen solutions were prepared as mentioned above, suspending NIH/3T3 or Hep G2 cells at 1.0×10^5 cells/mL were formed in the 5-mm thick mould on TCPS dish and kept at 37°C for 30 min.

Adhesion test Polymeric glues were used as solutions diluted with D-PBS to 1 wt%, and then the solutions, including PBA-PG and CAT-PG, were coloured green and red, respectively, using food colourings. To modify polymeric glues to collagen gels, gels were immersed in the solution at 37°C for 30 min. After the modification of polymeric glues, modified collagen gels were floated on PBS or DMEM-LG in a petri dish and contacted manually using a tweezer.

Cytotoxycity test First, 20 μL of collagen solutions containing 5.0×10^5 cells/mL of Hep G2 cells were dropped onto glass bottom dishes. After 30 min incubation at 37°C for making the collagen gel, 1 wt% of PBA-PG solution or CAT-

PG solution in D-PBS was poured onto the collagen gel containing cells formed on a dish, and additionally incubated for 30 h at 37°C. The solution was then changed to D-PBS and incubated for another 30 min to remove excess polymeric glues. After the procedure, the solution was re-changed to DMEM-LG, and the cells inside the surface-modified collagen gel were cultured for 24 h at 37°C. Then, the media was removed and cells were incubated in Live/Dead dyes (2 μM calcein-AM and 4 μM ethidium homodimer) in PBS for 30 min at 25°C. Finally, cells were observed using a confocal microscope for counting cells and comparing the amount of live and dead cells inside the surface region (a region within 200 μm from the edge of a gel) and the inner region (a region other than the surface region). The cell viability was calculated from Eq. (1)

$$cell\ viability\ (\%) = \frac{live\ cells \times 100}{live\ cells\ +\ dead\ cells} \#(1)$$

Diffusion test F-PG was composed of DMAAm, GMA, and FLAAm (DMAAm : GMA : FLAAm = 89.9 : 10 : 0.1) and was synthesised using the same procedure used for the polymeric glues. A 1 wt% fluorescent copolymer solution diluted with D-PBS was poured onto the collagen gel constructed in the mould as shown in Fig.3(b) and kept for 30 min at 37°C to confirm the diffusion of a polymeric glue to a gel.

Supporting Figures

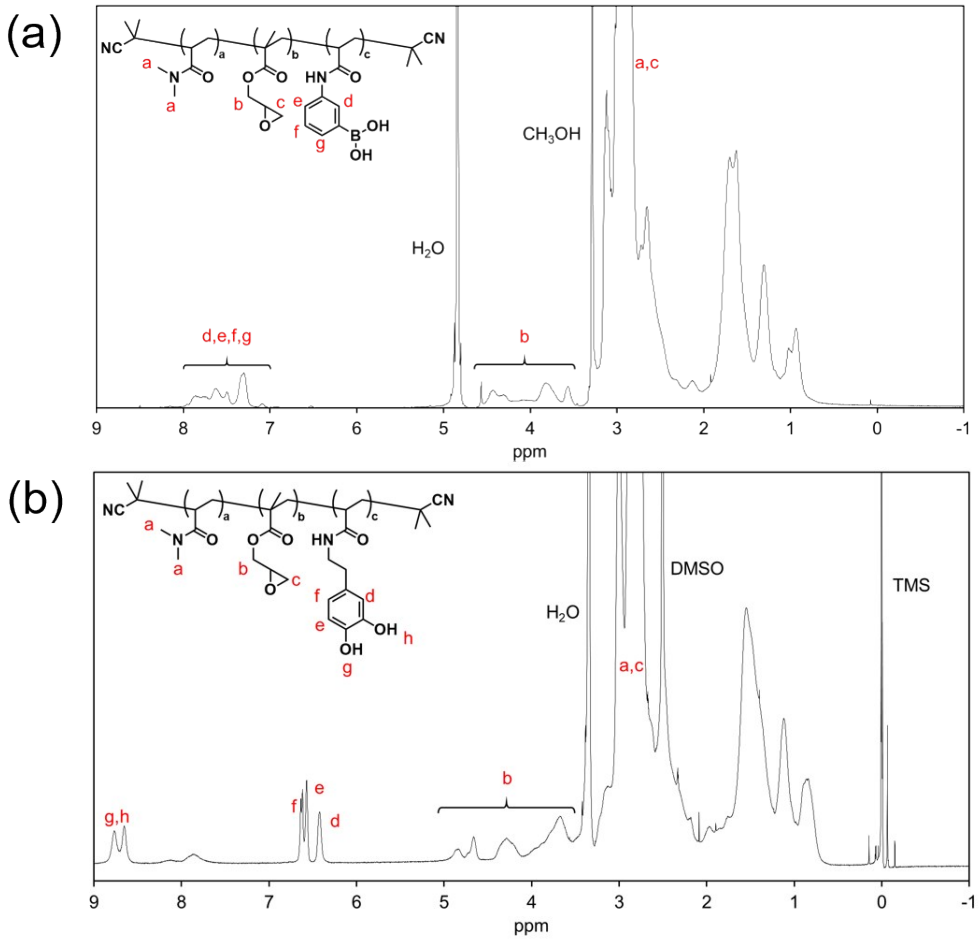


Fig. S1 ^{1}H-NMR spectra of (a) PBA-PG and (b) CAT-PG.

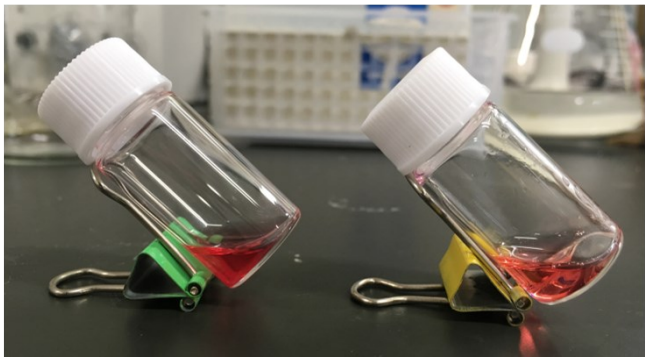
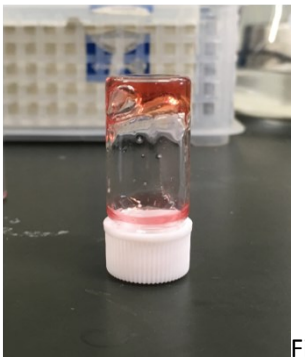
(a)  (b) 

Fig. S2 Image of PBA-PG solution (on the left in (a)) and CAT-PG solution (on the right in (a)) in DMEM-LG. After mixing these two solutions, the mixed solution converted to the gel state and lost its fluidity in a few minutes (b).

Supporting Movies

Movie S1

Adhesion between PBA-PG-modified (green) and CAT-PG-modified(red) collagen gels (PBA-PG+CAT-PG) and attachment of PBA-PG modified collagen gels(PBA-PG+PBA-PG) or CAT-PG modified collagen gels(CAT-PG+CAT-PG).

References

1 J. Nishida, M. Kobayashi and A. Takahara, *ACS Macro Lett.*, 2013, **2**, 112–115.

2 M. J. Serpe, C. D. Jones and L. A. Lyon, *Langmuir*, 2003, **19**, 8759–8764.