

Supporting information for

Rapid fabrication of reconstructible hydrogels by electrophoretic microbead adhesion

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1. Materials

All reagents were used as purchased without further purification. Poly(vinyl alcohol) (polymerization degree=1500–1800), 25% glutaraldehyde solution, sorbitan monooleate (Span 80), and liquid paraffin (density is 0.825-0.850 g mL⁻¹ at 20°C) were purchased from Wako Pure Chemical Industry (Osaka, Japan). Poly(sodium 4-styrenesulfonate) (PSS, M_w = 1,000,000) and 20 wt% poly(diallyldimethylammonium chloride) (PDDA, M_w = 400,000–500,000) solution were purchased from Sigma-Aldrich (USA).

2. Preparation of microbeads

PVA was dissolved in water at 10 wt%. Glutaraldehyde (1 mol% to the vinyl alcohol units) and 100 μ L of 2 M hydrochloric acid were added to 10 mL of 10 wt% PVA aqueous solution, and to this viscous solution was added 35 mL liquid paraffin, followed by vigorous stirring. 200 mg of Span 80 added into this suspension as a stabilizer, and the cross-linking reaction carried out at ambient temperature for 24 h with stirring. For the preparation of the cationic and anionic microbeads, 1 wt% PSS or PDDA were added into the reaction mixture, respectively. The PVA microbeads

were lyophilized after washing with hexane, 2-propanol, and water, consecutively. The lyophilized microbeads were separated between diameters of 100 to 125 μm by a sieving mesh, and were used to prepare the hydrogel constructs.

3. Electrophoretic adhesion of microbeads

The same amounts (10 mg) of lyophilized cationic and anionic microbeads were dispersed in 0.3 mL deionized water. When the mixed microbead powders were dispersed in water, a pasty product was formed. Electrophoresis was carried out between two platinum (Pt) electrodes. To adhere the microbeads, pasty products were held in contact with each other between the Pt electrodes, and an electric field was applied. The applied voltage was $\pm 10 \text{ V mm}^{-1}$ and the time was 5 sec in each electric field direction.