Supporting Information

Effects of pH on structure and mechanical property

of dried pH-responsive latex particles

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Gel permeation chromatography (GPC)

GPC analysis was performed using a refractive index (RI) detector equipped with a Shodex Ohpak SB-G guard column and SB–804 HQ column (bead size = 10 μm, exclusion limit: $M_n = 1.0 \times 10^6$, pullulan) working at 40 °C under a flow rate of 0.6 mL/min.   0.3 M Na_2SO_4 aqueous solution containing 0.5 M acetic acid was used as an eluent.   The molecular weight of the sample polymer was calibrated with standard poly(2-vinylpyridine) samples of 6 different molecular weights ranging from 5.70×10^3 to 3.16×10^5 g/mol.

Fast Fourier transform analysis

A fast Fourier transform (FFT) analysis of the SEM image of the dried particulate materials was conducted using ImageJ software (National Institutes of Health, USA).

Table S1.   Volume-average (D_v) and number-average (D_n) diameters and diameter distributions data for the PDEA-PS latex particles used in this study

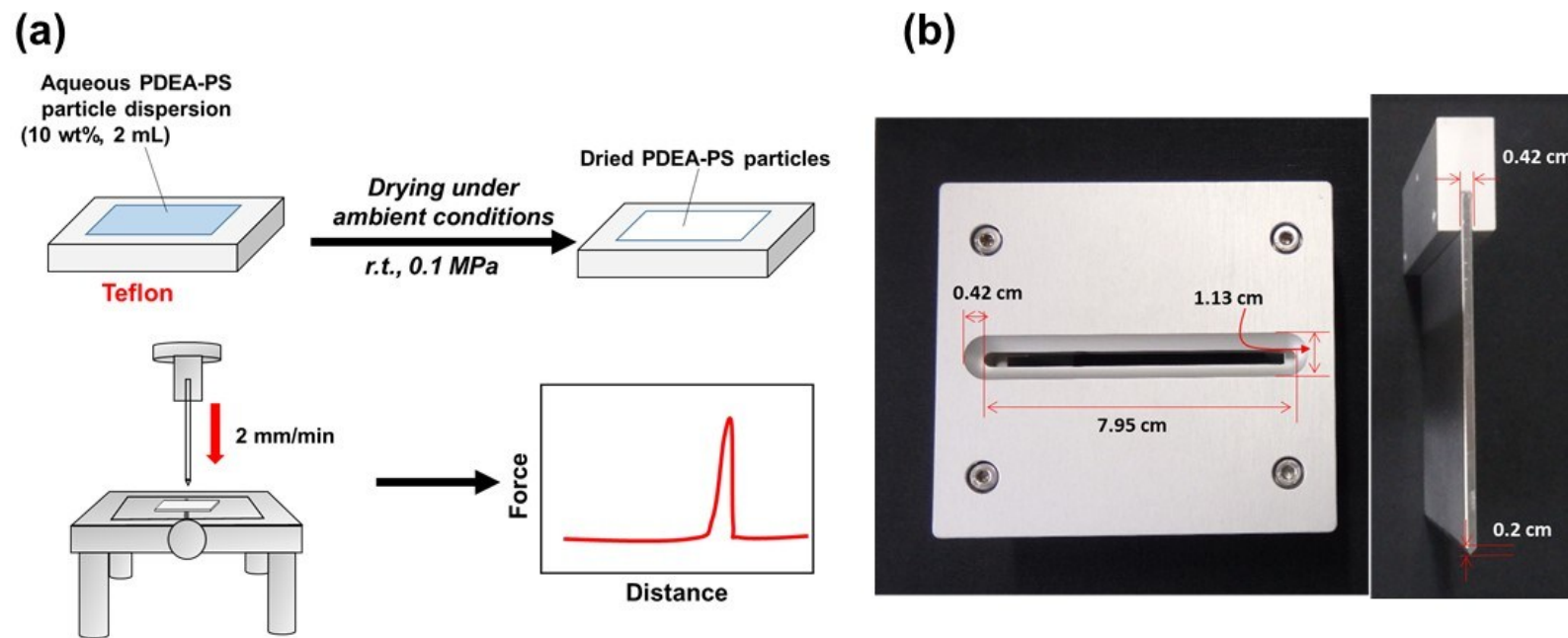|	Media	D_v / μm	D_n / μm	D_w / D_n
PDEA-PS	IPA	2.50±0.63	2.24	1.01
PDEA-PS	water	2.59±0.77	2.20	1.01

Figure S1.   (a) Scheme for the fracture test of the samples using the universal testing machine. Rectangular slabs of dried particular matter were placed across a slit and then indented from top with a metal blade with a speed of 2 mm/min until fracture occured. (b) Images of the slit and the metal blade.
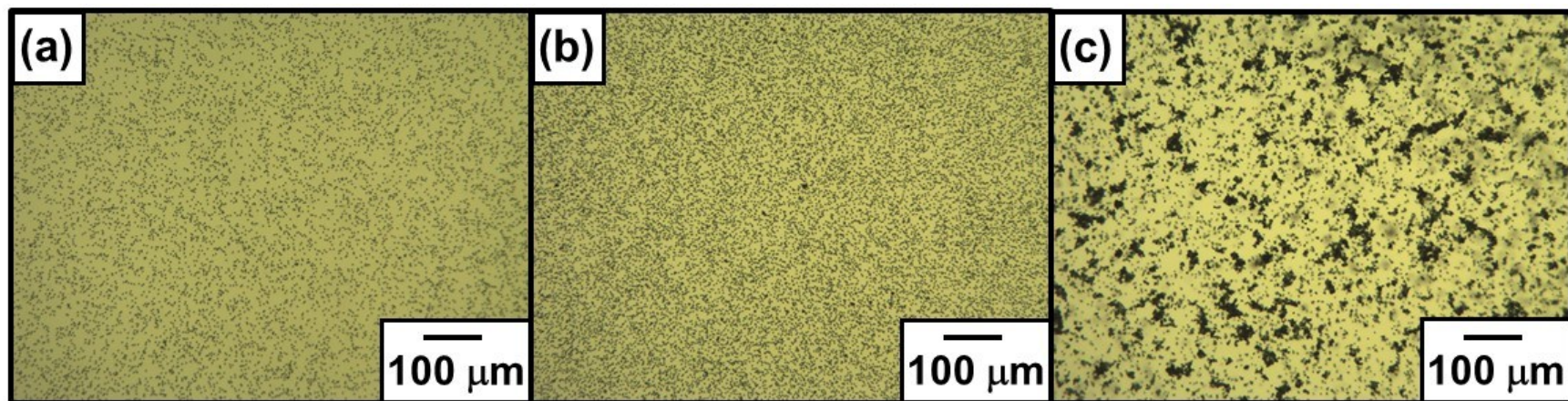
Figure S2. Optical microscopy images of aqueous dispersion of PDEA-PS latex particles: (a) pH 3.0 (b) 6.3 and (c)10.0. pH was adjusted using HCl or NH_3 aqueous solution.
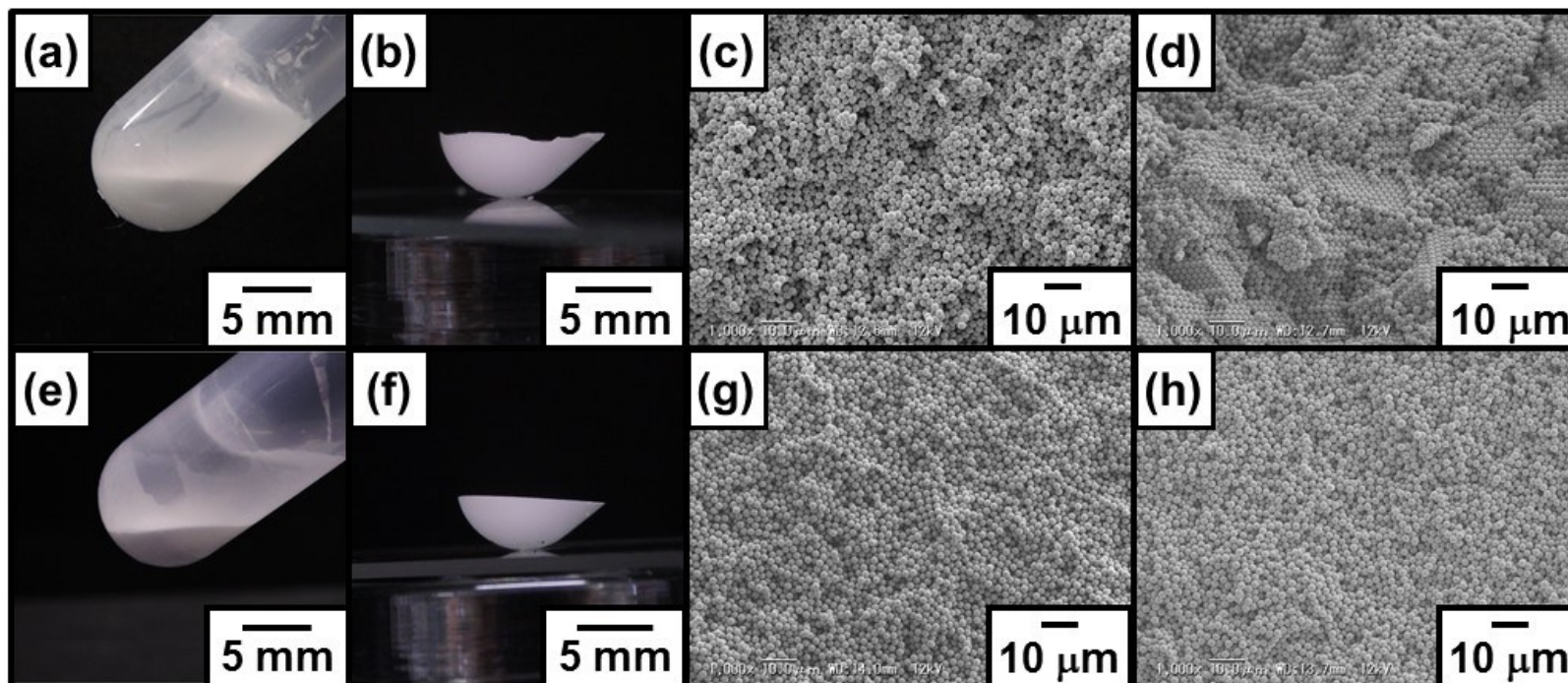
Figure S3. (a, b) Digital photographs of PDEA-PS particles sedimented in aqueous media with pH 10.0, and (b, f) digital photographs and (c, d, g, h) SEM images of dried PDEA-PS particulate materials: (a-d) without and (e-h) with centrifugal force (1355 *g*). Cross section images of the dried particulate materials: (c, g) Upper parts and (d, h) lower parts. The volume of the sediment decreased to approximately 77 % based on that without the application of centrifugal force. We confirmed that the PDEA-PS particles formed randomly-packed structure after the application of centrifugal force in dried particulate materials, which is the reason why the volume of the sediment did not decrease to 44%, where hexagonally close-packed arrangement is attained: 9.8 mm / 22.1 mm×100%.
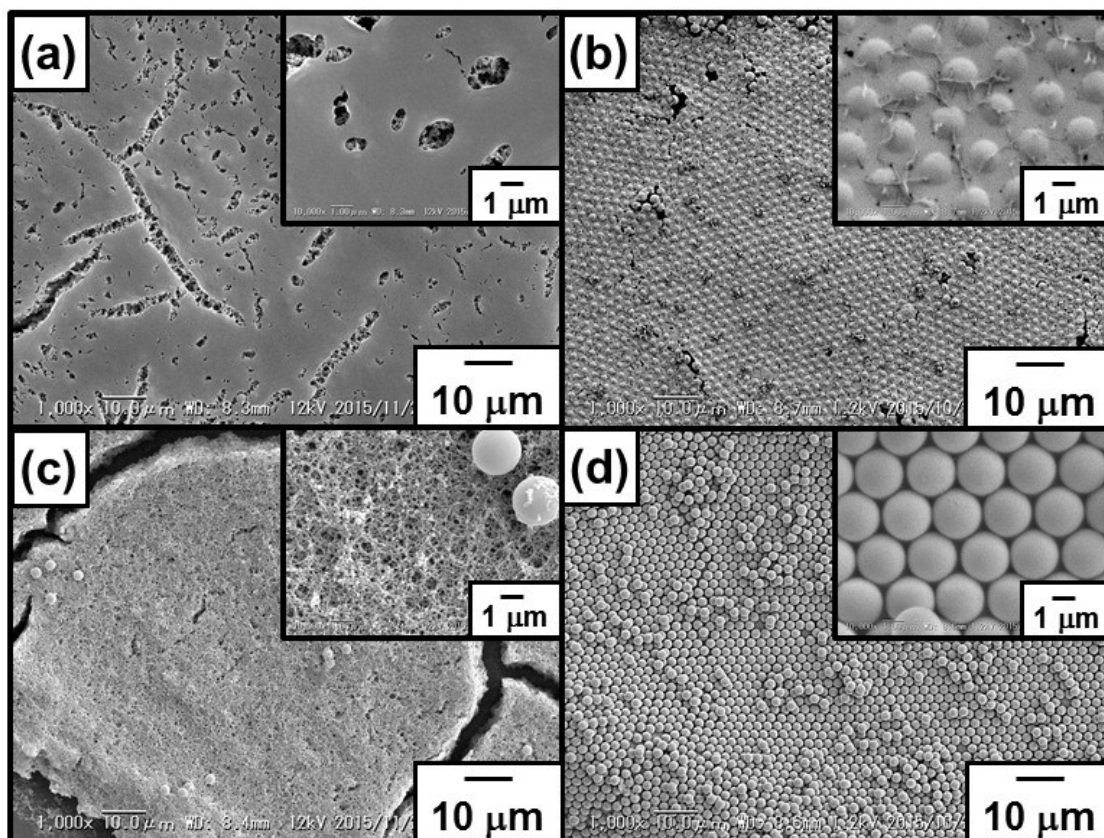
Figure S4. SEM images of PDEA-PS particles at the air-water surface (a, c) pH 3.0 and (b, d) pH 10.0. (a, b) air-exposed and (c, d) water-exposed side of the films.
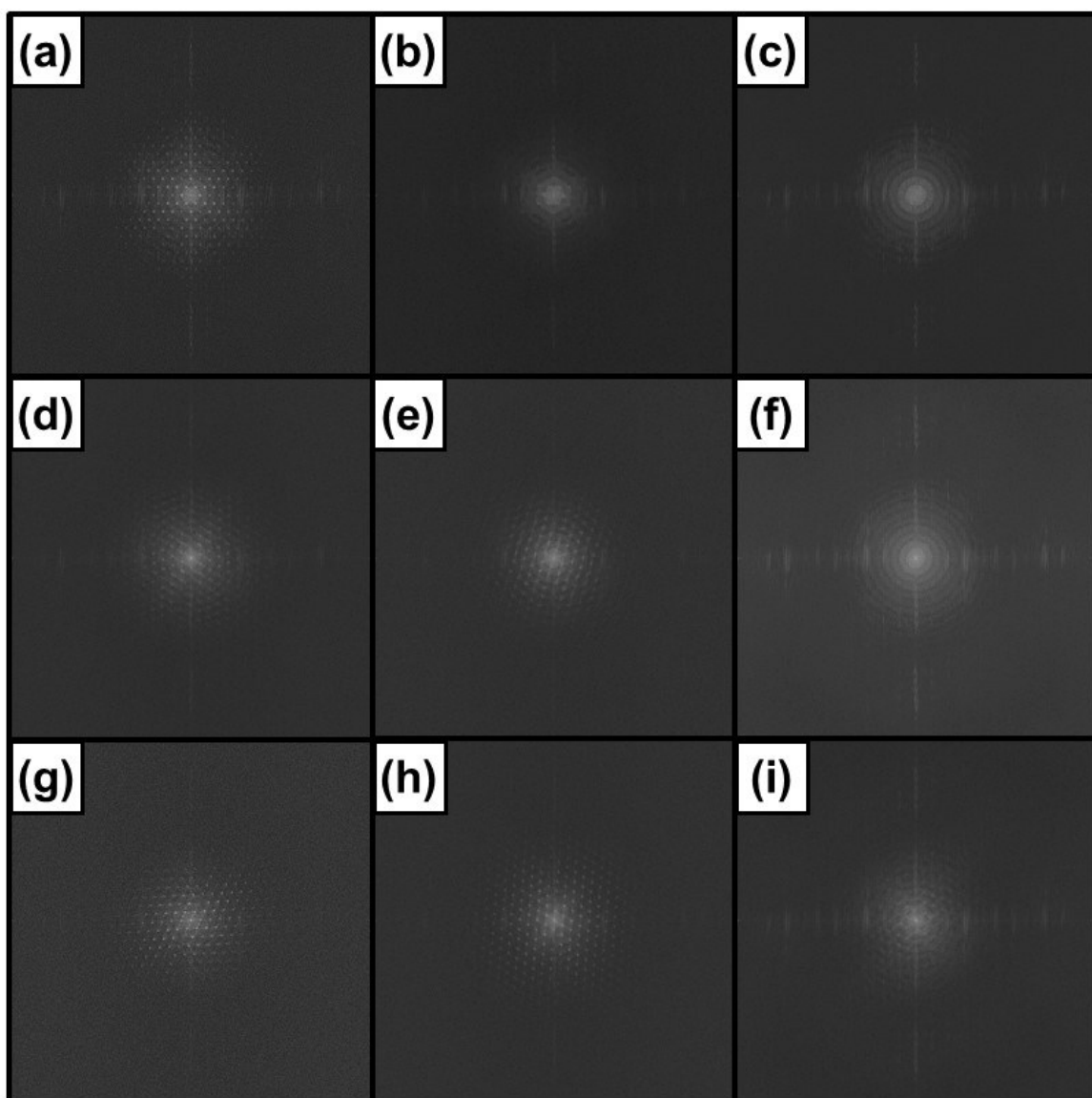
Figure S5   Fast Fourier transform (FFT) analysis of the SEM images of the dried particulate materials shown in Figures 5d-f and insets of Figures 7d-i: (a-c) surface and (d-i) cross-section of the materials dried at (a,d,g) pH 3.0, (b,e,h) pH 6.3 and (c,f,i) pH 10.0. (d-f) Upper middle parts and (g-i) lower parts of the dried particulate materials.