Supporting Information

Aqueous Two-Phase Droplet-Templated Colloidosomes Composed of Self-Formed Particles via Spatial Confined Biomineralization

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Supplementary Tables

**Table S1.** The strength of hydrogels and colloidosomes.

<table>
<thead>
<tr>
<th>Types of materials</th>
<th>Hydrogels</th>
<th>Colloidosomes with $\tau = 60$ min</th>
<th>Colloidosomes with $\tau = 120$ min</th>
<th>Colloidosomes with $\tau = 240$ min</th>
<th>Colloidosomes with $\tau = 300$ min</th>
</tr>
</thead>
<tbody>
<tr>
<td>Strength (g)</td>
<td>82.8(± 1.5)</td>
<td>99.5(± 3.2)</td>
<td>135.8(± 3.1)</td>
<td>164.5(± 2.9)</td>
<td>190.2(± 2.5)</td>
</tr>
</tbody>
</table>

Supplementary Figures

**Figure S1.** Typical images of ATPS alginate droplets with diameter of 258 ± 9.1 μm (a), 308 ± 8.9 μm (b), 409 ± 15.9 μm (c) and oval shapes as $D_t = 200$ μm and $C_A = 0.3\%$ (d).
Figure S2. Optical microscopy images of colloidosomes. (a) $C_E = 2\%, \tau = 360\text{ min}$; (b) $C_E = 5\%, \tau = 120\text{ min}$.

Figure S3. The pore size of different compactness of colloidosomes with $\tau = 120\text{ min}$ (a), 240 min (b) and 300 min (c).

Figure S4. The SEM image of cross section of colloidosomes.
Figure S5. Encapsulation efficiency of BSA in colloidosomes with different templates size (a), $d_A = 183 \pm 3 \mu m$, $d_B = 236 \pm 6 \mu m$, $d_C = 349 \pm 12 \mu m$ and different biomineralization time (b), $\tau_A = 60$ min, $\tau_B = 120$ min, $\tau_C = 240$ min, $\tau_D = 300$ min.

Supplementary Movies

Movie S1. ATPS alginate droplet formation in microfluidic coaxial capillary device.

Movie S2. Stability of colloidosomes with and without alginate.