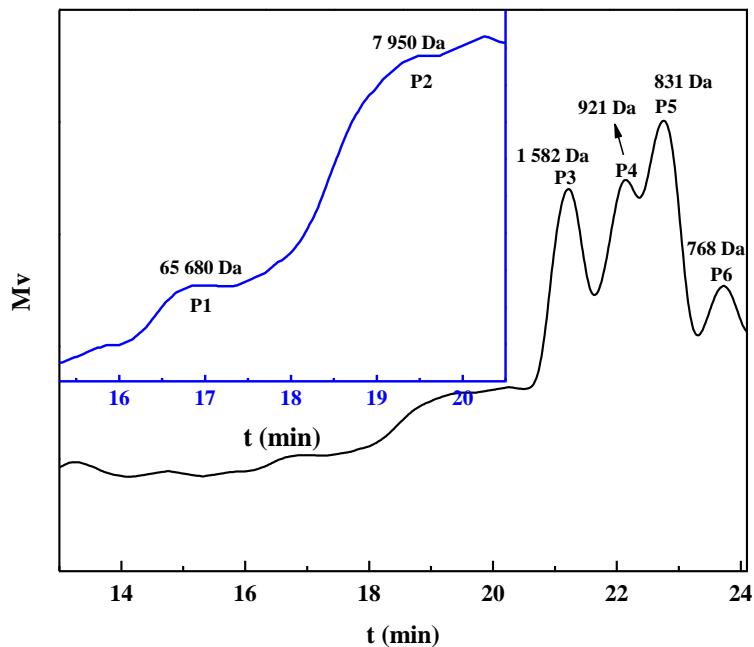


## Supplementary Information

### Preparation and characterization of homogeneous and enhanced casein protein-based composite films via incorporating cellulose microgel

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**Fig. S1** GPC elution diagrams of native casein.

**Table S1**

Summary of number- and weight-average molecular weight (Mn and Mw) and polydispersity index (PDI) of casein measured by GPC

| Casein | Mn [Da] | Mw [Da] | PDI  |
|--------|---------|---------|------|
| Peak 1 | 64101   | 65680   | 1.02 |
| Peak 2 | 6154    | 7950    | 1.29 |
| Peak 3 | 1558    | 1582    | 1.02 |
| Peak 4 | 877     | 921     | 1.05 |
| Peak 5 | 823     | 831     | 1.01 |
| Peak 6 | 740     | 768     | 1.03 |

**Table S2**

Summary of number- and weight-average molecular weight (Mn and Mw) and polydispersity index (PDI) of gelatin, Gel-NaOH/urea, C-gel-NaOH/urea, and GCC measured by GPC.

|                 | Mn [kDa]   | Mw [kDa]   | PDI   |
|-----------------|------------|------------|-------|
| Gelatin         | 0.80~17.74 | 0.88~63.19 | 3.561 |
| Gel-NaOH/urea   | 0.58~1.62  | 0.60~2.21  | 1.364 |
| C-gel-NaOH/urea | 0.73~3.69  | 0.78~10.62 | 2.879 |
| GCC             | 627.19     | 636.60     | 1.015 |

**Note:** Gel-NaOH/urea means gelatin treated in NaOH/urea aqueous media at the absence of cellulose and epichlorohydrin; C-gel-NaOH/urea means ECH-coupled gelatin in NaOH/urea aqueous media.

**Table S3**

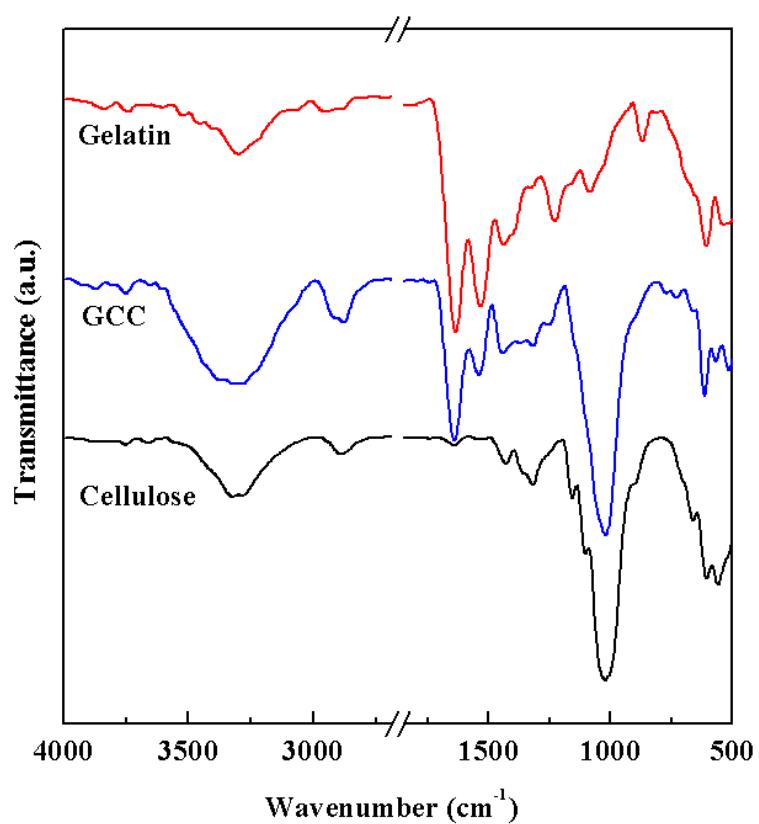
The N content ( $W_N$ , %), protein content ( $W_{Pro}$ , %) and yield (%) of GCC microgel.

|         | $W_{Gel}^a$ (%) | $W_{Cel}^b$ (%) | $n_{ECH}:n_{AGU}^c$ | $W_N$ (%) | $W_{Pro}$ (%) | Yield (%) |
|---------|-----------------|-----------------|---------------------|-----------|---------------|-----------|
| Gelatin | 100             | 0               | -                   | 14.46     | 90.38         | --        |
| GCC     | 20              | 80              | 2                   | 2.99      | 18.69         | 21.3      |

<sup>a</sup> $W_{Gel} = [\text{weight of gelatin}/(\text{weight of cellulose} + \text{weight of gelatin})] \times 100$ .

<sup>b</sup> $W_{Cel} = [\text{weight of cellulose}/(\text{weight of cellulose} + \text{weight of gelatin})] \times 100$ .

<sup>c</sup> $n_{ECH}:n_{AGU}$  = the molar ratio of epichlorohydrin to anhydroglucosamine unit of cellulose.



**Fig. S2** FTIR of gelatin, cellulose, and GCC microgel.