Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B. This journal is © The Royal Society of Chemistry 2021

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Fig. S1 TEM images of the surface layers between top and internal surfaces of (a) 0Cit/HA, (b)
1Cit/HA, (c) 2Cit/HA and (d) 3Cit/HA where the average surface layer thicknesses were 0, 1.7, 2.2
and 4.1 nm, respectively. (e) Illustration of the possible surface layer structures of the Cit/HA
nanoparticles.

# 49 Figure S2



#### 56 Figure S3

57





60 **Fig. S3** (a) Transmittance and haze value changes of the cal-Cit/HA-F films with the coordinated 61 amount of Cit, and the photographs of (b) cal-**0**Cit/HA-F and (c) cal-**3**Cit/HA-F.

# 64 Figure S4



- 68 Fig. S4 GD-OES elemental depth profiles of (a) 0Cit/HA-F, (b) 1Cit/HA-F, (c) 2Cit/HA-F and (d)
- **3**Cit/HA-F.

# 72 Figure S5



- **Fig. S5** Microscopic photographs of (a, e) cal-**0**Cit/HA-F, (b, f) cal-**1**Cit/HA-F, (c, g) cal-**2**Cit/HA-
- F and (d, h) cal-3Cit/HA-F before and after the tape peeling test and (i) their reduction rate of the
   film areas.

# 81 Figure S6

82



83 84

- 85 Fig. S6 Optical microscope images of the cell adhesion on 0Cit/HA-F at the culture time of 3 h
- 86 ((a) lower and (b) higher magnification).

#### 89 Scheme S1



93 Scheme S1 Illustration of the possible nanospace formation in the Cit/HA-F films based on the
 94 results in the N<sub>2</sub> adsorption and desorption isotherms.

#### 97 Scheme S2

98



99

100

- 101 Scheme S2. Illustration of the changes in (a-d) arrangement states and (a'-d') interaction forces
- 102 between the nanoparticles during the dry process of **3**Cit/HA-F. Here, only the nanoparticles in the
- 103 bottom layer are discussed for easily understanding.

#### *Figure* **S7**



Fig. S7. SEM and AFM surface topographic images of (a, b) cal-0Cit/HA-F and (c, d) cal3Cit/HA-F, and the RMS values of cal-0Cit/HA-F and cal-3Cit/HA-F were 7.69 nm and 4.59 nm,

 $112\;$  respectively. (e) Nanoparticle size distribution in the cal-3Cit/HA-F.