

1 Supplementary

2 **Freeze-crosslinking approach for preparing carboxymethyl cellulose**

3 **nanofiber/zirconium hydrogels as fluoride adsorbents**

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16 **Characterization**

17 The compressive strengths of the samples were measured on a texture analyser (TA.XTplus,
18 Stable Micro Systems Co. Ltd., UK). The swollen under wet conditions were compressed to a
19 strain of 0.8 with a cylindrical probe (20 mm diameter) at a speed of 1 mm s^{-1} . The Young's
20 moduli were calculated from the slopes of the stress–strain curves in the linear region (0.01–
21 0.05 strain). For TG tests, approximately 4 mg of sample was heated in an open aluminium pan
22 to 413 K (rate of 10 K/min). For BET tests, the samples were degassed at 333 K under reduced
23 pressure for 5 hours prior to each measurement. For SANS analysis, the scattering intensities
24 from the sample cell and the solvent were subtracted by using measured transmission and the
25 volume fraction of the solvent. Samples for the SANS experiments were prepared using D_2O as
26 solvent.

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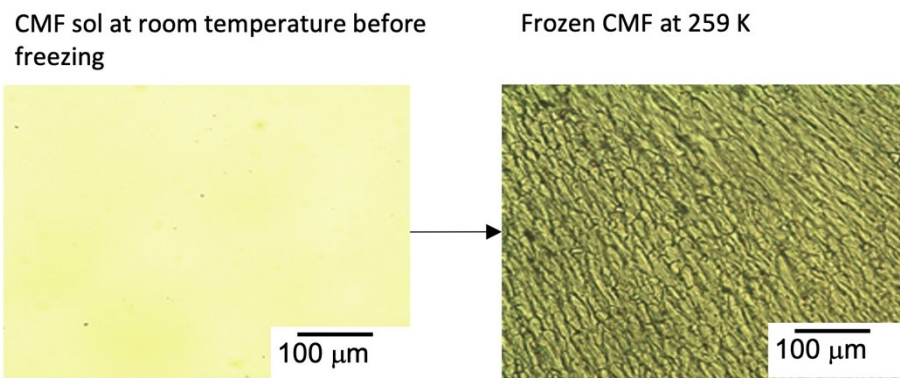
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38 **Figure S1.** Optical microscopy images of CMF before and after freezing.

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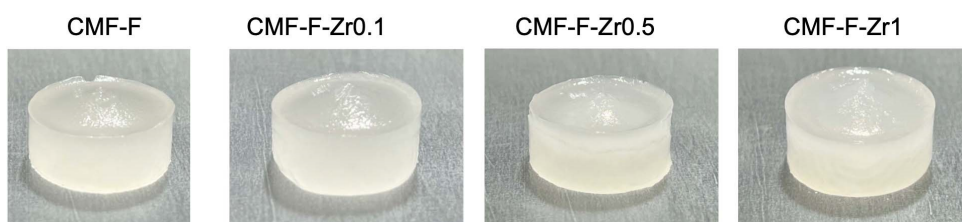
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51 **Figure S2.** Photographs of the CMF-F and CMF-F-Zx (x = 0.1, 0.5, or 1).

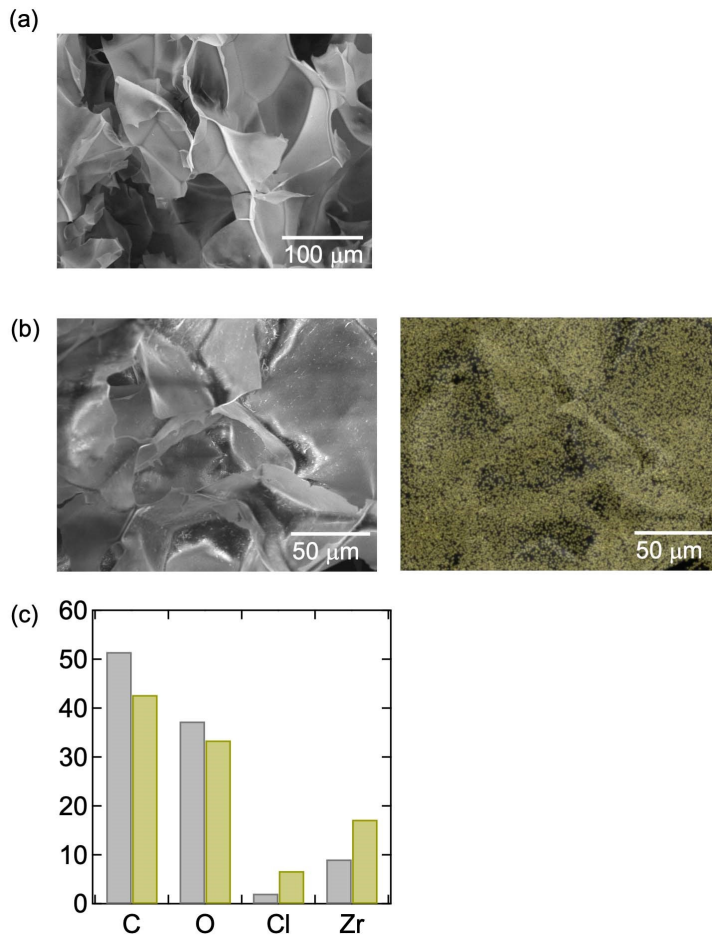
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59 **Figure S3.** (a) SEM images of freeze-dried CMF-F. (b) SEM-EDX images for CMF-F-Z3. The yellow color
60 represents Zr. (c) The relative ratios of C, O, Cl, Zr in CMF-F-Z0.5 (gray) and CMF-F-Z3 (yellow)
61 estimated from SEM-EDX results.

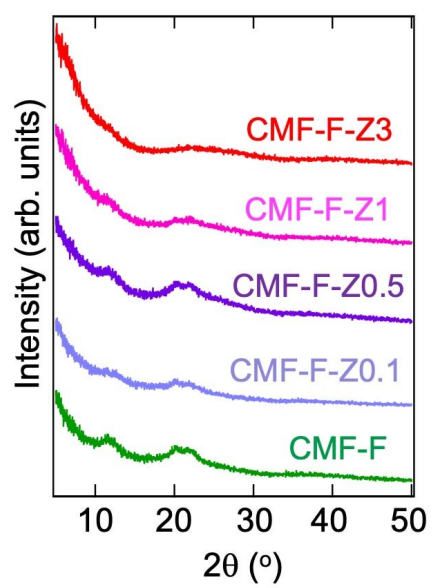
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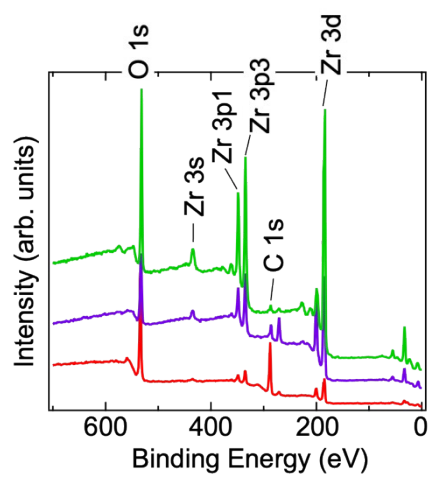


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68 **Figure S4.** PXRD patterns of freeze-dried CMF-F and CMF-F-Z_x (x = 0.1, 0.5, 1, or 3).

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74 **Figure S5.** XPS survey scans of CMF-F-Z3 (red), ZrCl₂O·8H₂O (purple), ZrO₂ (green).

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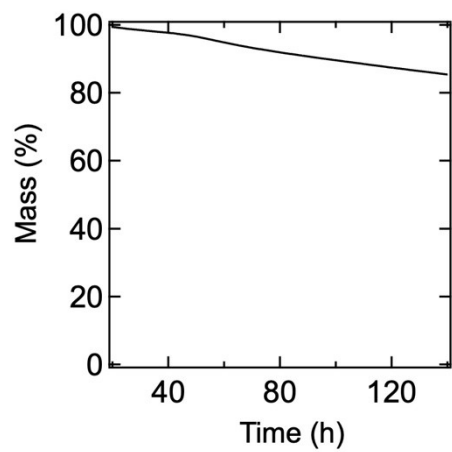


Figure S6. TG curve of freeze-dried CNF-F-Z3 hydrogel.

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93 **Table S1** Langmuir isotherm fitting parameters for the adsorption of fluoride on CMF-F-Z3.

94 The equation is as $Q_e \text{ (mg g}^{-1}\text{)} = (b \times Q_{max} \times C_e)/(1 + b \times C_e)$, where $b \text{ (L mg}^{-1}\text{)}$ is a

95 constant related to the free energy of adsorption, $C_e \text{ (mg L}^{-1}\text{)}$ is the equilibrium

96 concentration of fluoride in the solution, $Q_e \text{ (mg g}^{-1}\text{)}$ is the amount of fluoride

97 adsorbed per unit weight of CMF-F-Z3, and $Q_{max} \text{ (mg g}^{-1}\text{)}$ is the maximum adsorption

98 capacity.

Material	Langmuir isotherm model		
	$Q_{max} \text{ (mg g}^{-1}\text{)}$	$b \text{ (L mg}^{-1}\text{)}$	R
CNF-F-Z3	24.1	0.0462808	0.9957

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