## **Supporting Information**

## Hierarchical Alginate Biopolymer Papers Produced via Lanthanide Ions Coordination

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Fig. S1 Effect of (a)  $Nd^{3+}$  ion concentrations (0.01, 0.02, 0.05, 0.1, and 0.2 M) and (b) temperature on gelation time.



Fig. S2 FTIR spectral analysis of SA biopolymers and SA-Nd(III)-1 paper.



**Fig. S3** XPS spectra of (a) core-level C1s and (b) core-level O1s of the layered SA-Nd(III)-1 paper.



**Figure S4.** Comparison of the cross-sectional morphology and structure of the (a) SA-Gd(III), (b) SA-Ce(III), (c) SA-Yb(III), (d) SA-Ca(II), (e) SA-Fe(III), and (f) SA-Al(III) papers (SA concentrations was 2 wt %).



**Fig. S5** (a-b) Typical strain-stress curves and toughness of layered SA-Nd(III)-1 papers with different water contents. (c-d) Typical strain-stress curves and toughness of layered SA-Nd(III) papers which formed under different Nd<sup>3+</sup> ions concentrations. (e-f) Typical strain-stress curves and toughness of layered SA-Ce(III), SA-Gd(III) and SA-Yb(III) papers (SA, 2 wt%).



**Fig. S6** TGA curves of SA-Nd(III)-1, SA-Nd(III)-2, SA-Nd(III)-3 layered papers. The curves were obtained under atmosphere of nitrogen with a temperature rising rate of  $10^{\circ}$ C ·min<sup>-1</sup>

Samples	C (%)	N (%)	Nd (%)	Na (%)
<sup>a</sup> SA-Nd(III)-2	51.0	44.87	3.92	0.21
<sup>b</sup> SA-Nd(III)-2	51.23	44.64	3.90	0.23

**Table S1.** Atomic % of the SA-Nd(III)-2 paper before (<sup>a</sup>) and after (<sup>b</sup>) dipping in sodium chloride solution.

Table S2. The tensile strength and young's modulus of Alginate-based materials.

Samples	tensile	Young's	Dof	
	strength/MPa	modulus/GPa	Kel.	
SA/GO	69.32	3.8	[33]	
Al/GO	113	4.18	[34]	
SA-rGO	122	4.46	[38]	
GO/SA	2.33	-	[39]	
Alginate film	14	0.2	[40]	
Alginate-Based	7	0.03	[41]	
Nanofibrous	7	0.05		
Alginate-based	2.9	0.07	[42]	
film	,	0.07	['2]	
Alginate/pectin	75 7	_	[43]	
films	15.1			
SA-Nd(III)	124.2	5.25	This work	

Solvent uptake	SA-Nd(III) papers		
rate / %	25°Cª	90°C <sup>b</sup>	
Water	12.82°	18.34	
Ethanol	6.19	7.67	
THF	4.42	5.58	
DMF	3.07	4.15	
DMSO	4.63	5.88	
EDTA	d	d	

Table S3. Solvent uptake rate of the SA-Nd(III) hydrogel in various solvent.

<sup>a</sup> The sample was soaked in solvent at 25 °C for 24 h. <sup>b</sup> The sample was soaked in solvent at 25 °C for three days, 90 °C for 4 h in a sealed vessel. <sup>c</sup> The calculation formula of the solvent uptake rate: (W<sub>wet</sub> - W<sub>dry</sub>)/W<sub>dry</sub>×100%. <sup>d</sup> The sample was dissolved in this solvent.