Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information for

Biomimetic Ion-crosslinked Layered Double Hydroxide/Alginate hybrid film

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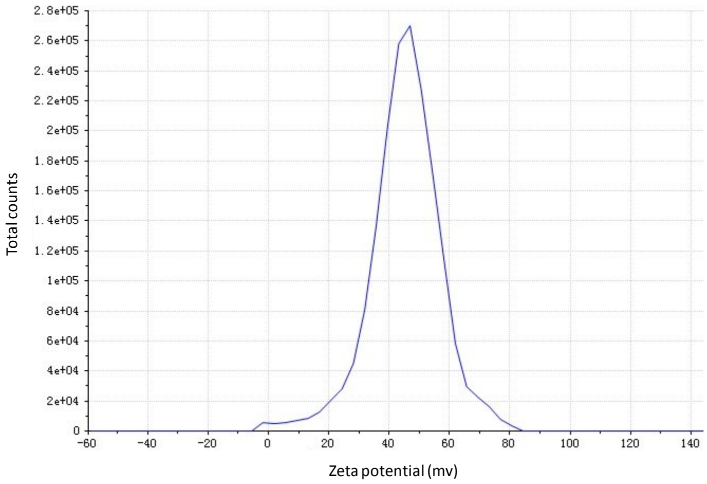

Figure S1. Zeta potential test for Ni-Al-NO_3 LDH nanoplatelets, showing a zeta potential of 45.3 mV.

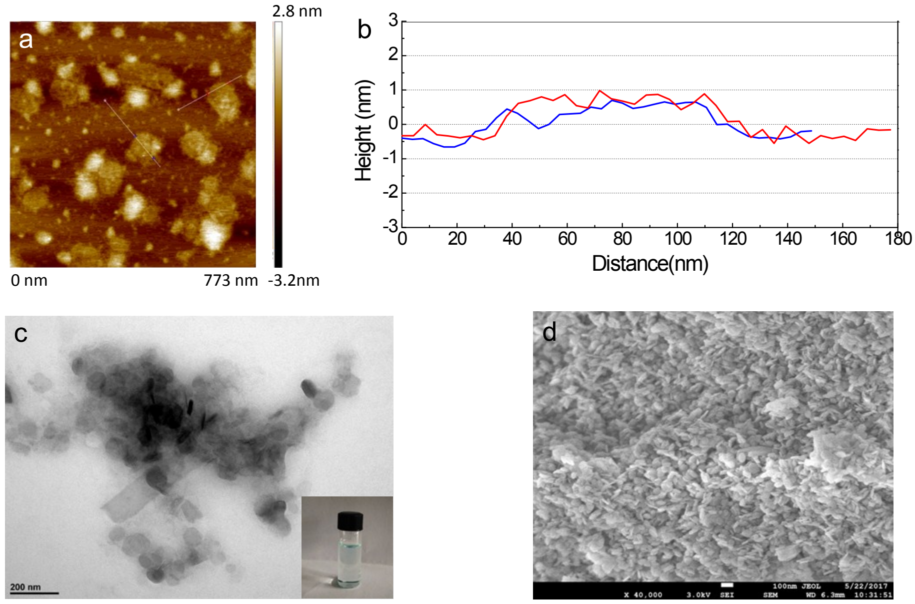

Figure S2. (a) AFM image of exfoliated LDH nanosheets. (b) Height profile along the marked line in (a). (c) TEM image of LDH nanosheets. The inset is the photograph of transparent colloidal suspension of exfoliated Ni-Al LDH nanosheets. (d) SEM image of LDH.

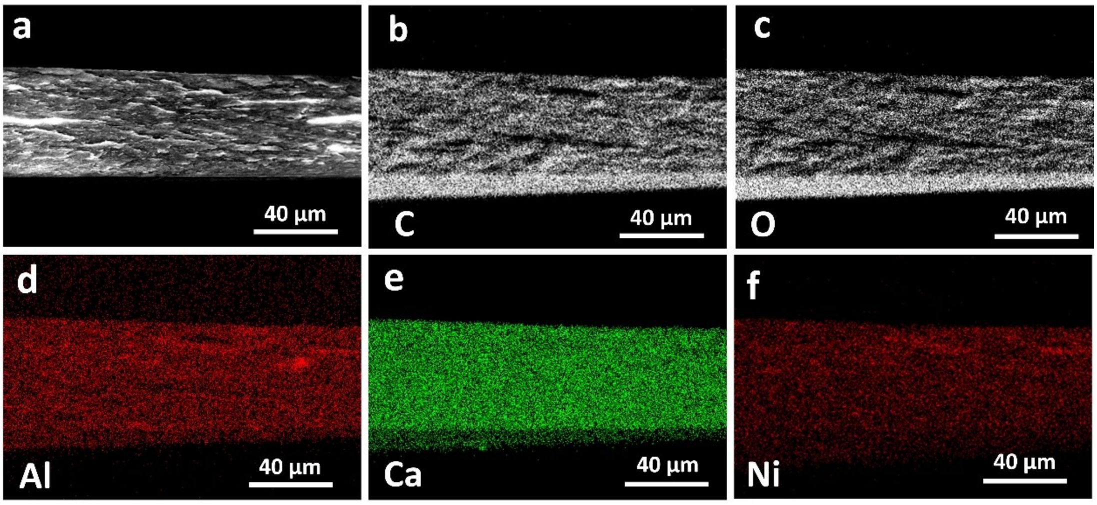

Figure S3. (a) SEM images of the cross section of 10% LDH/ALG-Ca^{2+} hybrid film. (b-f) Energy-dispersive X-ray (EDX) mapping of (b) carbon, (c) oxygen, (d) aluminum, (e) calcium and f) nickel. All elements are distributed homogeneously, indicating that LDH and ALG did not separate microscopically.

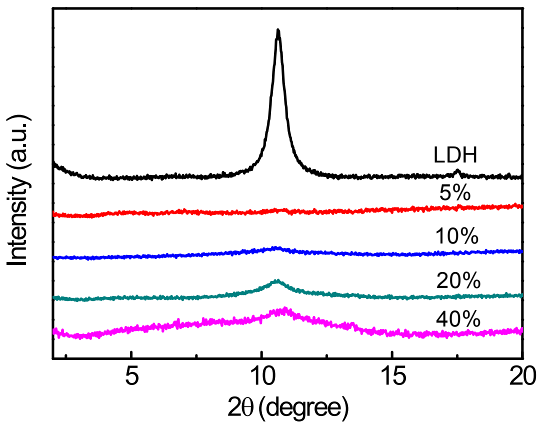

Figure S4 X-ray diffraction curves for LDH and LDH/ALG-Ca^{2+} hybrid films with different LDH contents and constant mixing mass ratio of $CaCl_2$ to ALG (0.5). For LDH, a sharp diffraction peak appear at $2\theta=10.3^{o}$, corresponding to an interlayer distance of 0.86 nm. When LDH content is lower than 20%, the diffraction peak of LDH/ALG-Ca^{2+} hybrid films disappear basically, indicating well exfoliation without aggregation of LDH. For the sample containing 20% or more LDH, the diffraction peak position is the same as that of LDH, indicating the presence of LDH aggregation stack.

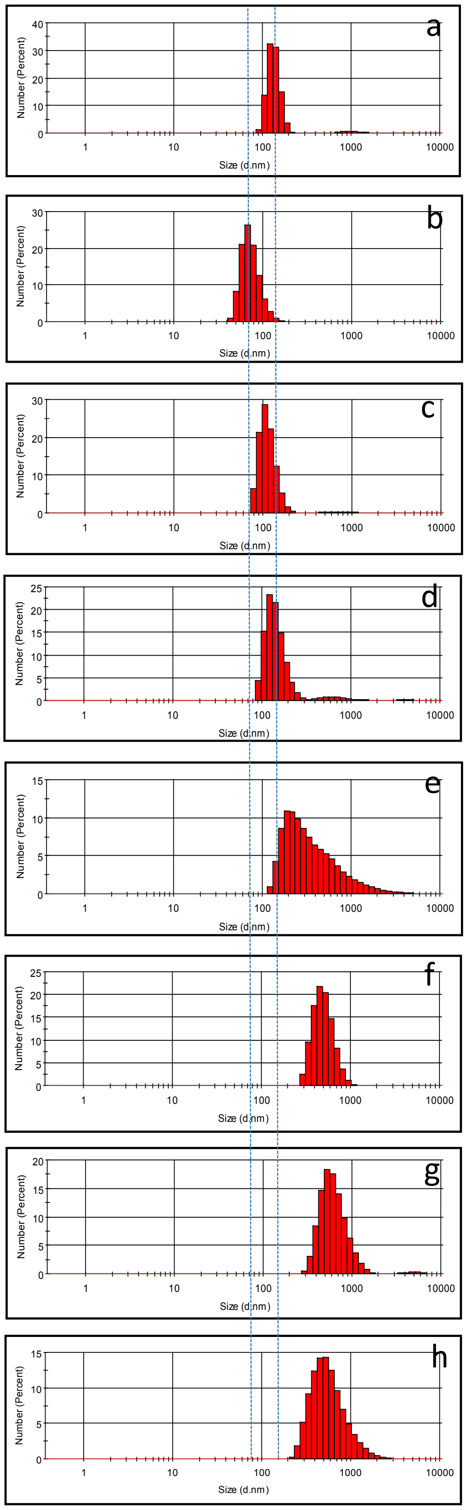

Figure S5 Dynamic light scattering for ALG, LDH and LDH/ALG/$CaCl_2$ mixed solutions with different LDH contents and constant mass ratio of $CaCl_2$ to ALG (0.5). (a) ALG, (b) LDH, (c) 5% LDH, (d) 10% LDH, (e) 20% LDH, (f) 30% LDH, (g) 40% LDH, (h) 50% LDH. Blue dash lines correspond to the hydrodynamic diameter of ALG solution and LDH dispersion. When LDH content is higher than 20%, the hydrodynamic diameter of LDH/ALG/$CaCl_2$ mixed solutions is obviously higher than those of ALG solution and LDH dispersion.

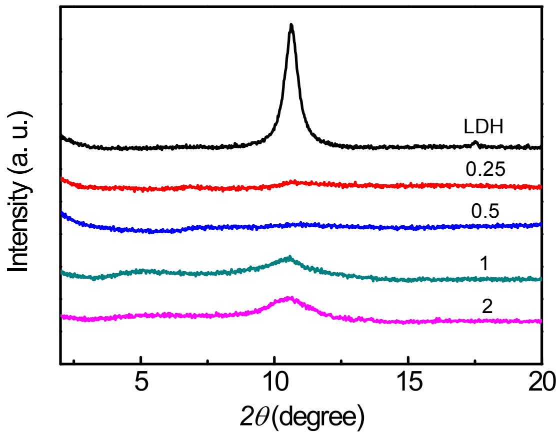

Figure S6 X-ray diffraction curves for LDH and LDH/ALG-Ca^{2+} hybrid films with different mixing mass ratio of $CaCl_2$ to ALG and constant LDH content (10%). For LDH, a sharp diffraction peak appear at $2\theta=10.3^{o}$, corresponding to an interlayer distance of 0.86 nm. When the mixing mass ratio of $CaCl_2$ to ALG is 0.25 or 0.5, the diffraction peak disappear basically, indicating well exfoliation without aggregation of LDH. For the sample with high mixing mass ratio of $CaCl_2$ to ALG (1 or 2), the diffraction peak position is the same as that of LDH, indicating the presence of LDH aggregation stack.

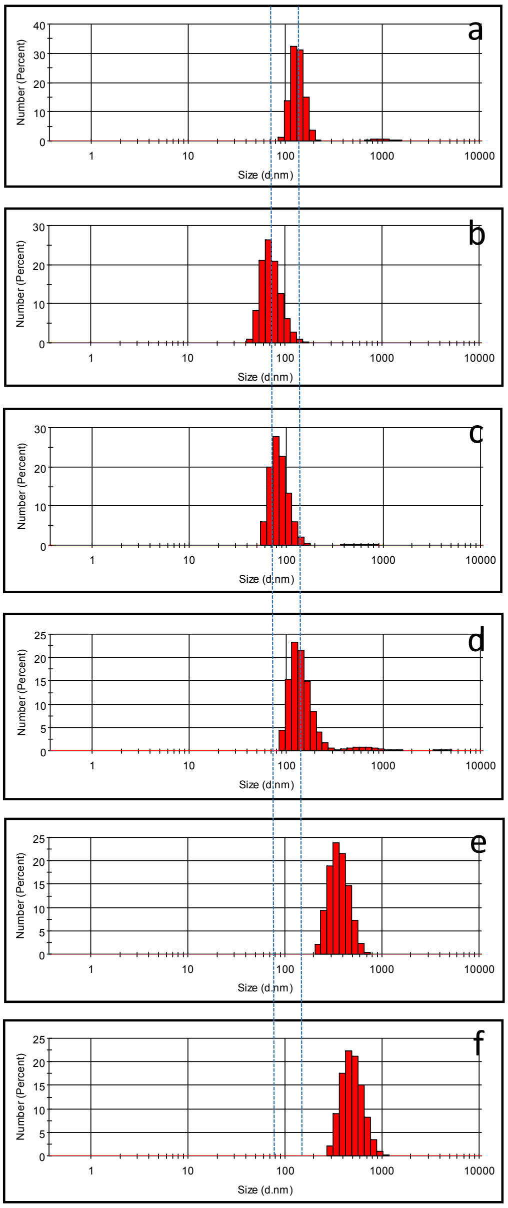

Figure S7 Dynamic light scattering for ALG, LDH and LDH/ALG/$CaCl_2$ mixed solutions with different mass ratio of $CaCl_2$ to ALG and constant LDH content (10%). (a) ALG, (b) LDH, (c) 0.25, (d) 0.5, (e) 1, (f) 2. Blue dash lines correspond to the hydrodynamic diameter of ALG solution and LDH dispersion. When the mass ratio of $CaCl_2$ to ALG is larger than 0.5%, the hydrodynamic diameter of LDH/ALG/$CaCl_2$ mixed solutions is obviously higher than those of ALG solution and LDH dispersion.

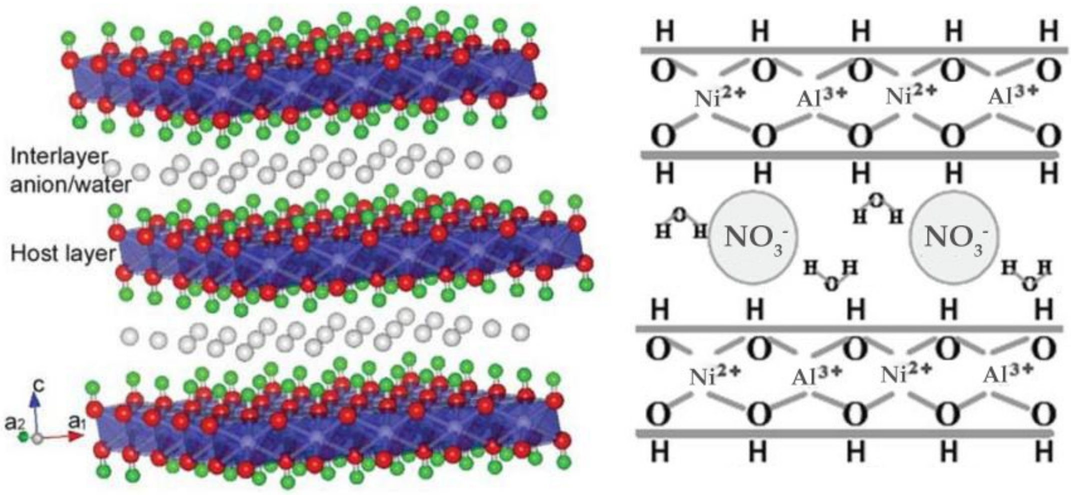

Figure S8 The structure representation of LDH stack. Single-layer Ni-Al-NO_3 LDH is a two-dimensional nanoplatelet, which contains plenty of hydroxyl groups on surface and charges positively. Multiple single-layer Ni-Al-NO_3 LDH platelets stack together, forming LDH stacks. Some interlayer water and NO_3^- exist between Ni-Al-NO_3 LDH nanoplatelets.

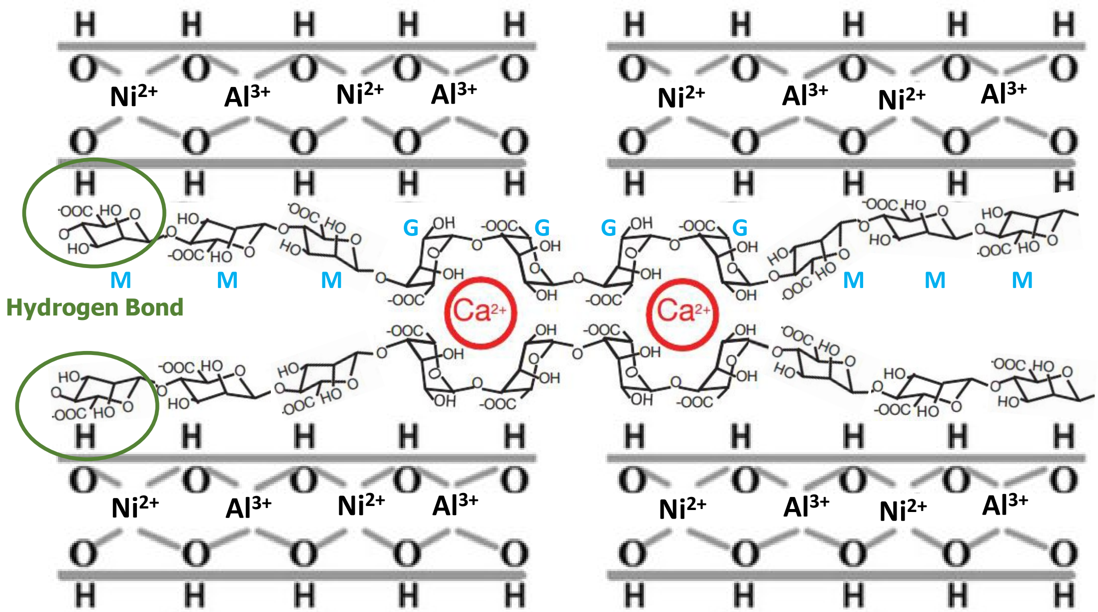

Figure S9 Proposed structural mode for LDH/ALG-Ca^{2+} hybrid film. ALG contains G units and M units. G units are cross-linked by Ca^{2+} and have relatively high rigidness. Carboxyl groups in M units form hydrogen bond with the hydroxyl groups on the surface of LDH.

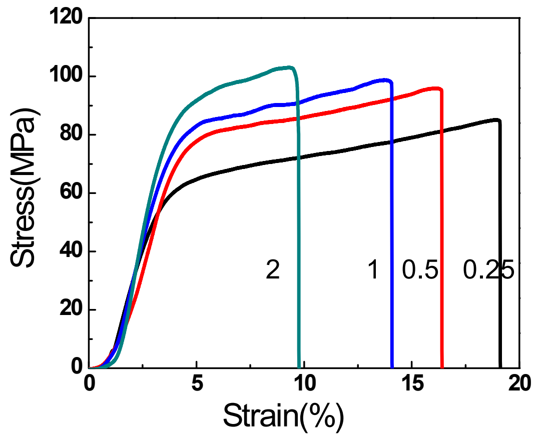

Figure S10 Tensile stress-strain curves of Ca^{2+}-crosslinked ALG with different mass ratio of $CaCl_2$ to ALG. With the mass ratio of $CaCl_2$ to ALG increasing from 0.25 to 2, the tensile strength of Ca^{2+}-crosslinked ALG increases continuously from 85 MPa to 103 MPa.

Table S1 The mixing volumes and concentrations of LDH, ALG and $CaCl_2$ solutions in the experimental process. The calculated LDH content and the mass ratio of $CaCl_2$ to ALG is listed in the last two columns.

LDH con. (wt%)	LDH vol. (ml)	ALG con. (wt%)	ALG vol. (ml)	$CaCl_2$ con. (wt%)	$CaCl_2$ vol. (ml)	$m_{LDH}/(m_{LDH}+m_{ALG})$	m_{CaCl2}/m_{ALG}
0.1	3	0.1	57	0.1	28.5	**5%**	0.5
0.1	6	0.1	54	0.1	27	**10%**	0.5
0.1	12	0.1	48	0.1	24	**20%**	0.5
0.1	18	0.1	42	0.1	21	**30%**	0.5
0.1	24	0.1	36	0.1	18	**40%**	0.5
0.1	30	0.1	30	0.1	15	**50%**	0.5
0.1	6	0.1	54	0.1	13.5	10%	**0.25**
0.1	6	0.1	54	0.1	27	10%	**0.5**
0.1	6	0.1	54	0.1	54	10%	**1**
0.1	6	0.1	54	0.1	108	10%	**2**