Supporting Information for

Controllable photoluminescence properties of an anion dye intercalated

layered double hydroxide by adjusting the confined environment

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Figure S1. FT-IR spectra of (a) NO₃-LDH, (b) ANS, (c) ANS/PES-LDH, (d) ANS/HES-LDH, (e) ANS/HPS-LDH, (f) ANS/DES-LDH and (g) ANS/DDS-LDH.

Figure S2. Photographs of (a) ANS/PES-LDH, (b) ANS/HES-LDH, (c) ANS/HPS-LDH, (d) ANS/DES-LDH and (e) ANS/DDS-LDH under UV irradiation of 365 nm.

Figure S3. Photographs of the ANS(x%)/HPS-LDH films (a ~ h) with x = 10%, 15%, 20%, 25%, 30%, 50%, 75%, 100% under UV irradiation of 365 nm.

Figure S4. Fluorescence decay curves and residual plots of double-exponential fitting for the ANS(x%)/HPS-LDH film samples with varied *x*: (a) x = 10%, (b) x = 15%, (c) x = 20%, (d) x = 25%, (e) x = 30%, (f) x = 50%, (g) x = 75% and (h) x = 100%.

Figure S5. TG-DTA curves for (A) ZnAl-LDH, (B) ANS and (C) ANS(20%)/HPS-LDH.

Figure S6. The fluorescence emission spectra of the ANS(20%)/HPS-LDH film (stored at ambient temperature) recorded in 4 month.

Table S1: Chemical compositions and crystalloid parameters for the ANS/surfactant-LDH composites.

Table S2: Chemical compositions and interlayer distance (d_{003}) of the ANS(x%)/HPS-LDH composites.

S2

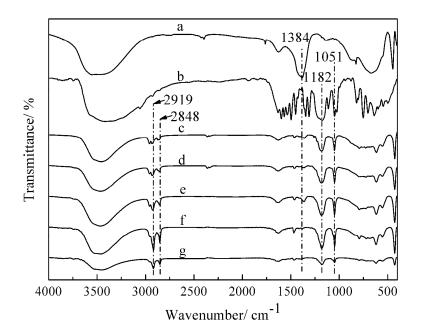


Figure S1. FT-IR spectra of (a) NO₃-LDH, (b) ANS, (c) ANS/PES-LDH, (d) ANS/HES-LDH, (e) ANS/HPS-LDH, (f) ANS/DES-LDH and (g) ANS/DDS-LDH.

The FT-IR spectra of NO₃-LDH, ANS, ANS/PES-LDH, ANS/HES-LDH, ANS/HPS-LDH, ZANS/DES-LDH and ANS/DDS-LDH are shown in Fig.S1, respectively. For the sake of clarity, only the main absorption bands were listed. The spectrum of NO₃-LDH (Fig.S1, curve a) shows a strong absorption band at 1384 cm⁻¹ and a weak one at 830 cm⁻¹ that can be attributed to the v₃ and v₂ stretching vibration of NO₃⁻ group respectively. The characteristic absorption band of carbonate at 1360 cm⁻¹ was not observed, confirming the absence of carbonate in the precursor. In the spectrum of pristine ANS (Fig.S1, curve b), the strong absorption bands at 1182 and 1051 cm⁻¹ are due to the O=S=O stretching vibration of the sulfonate group.¹ The spectra of ANS/surfactant-LDH composites (Fig. 2c, d, e, f, g) display characteristic bands of $-SO_3^-$ group at 1182 and 1051 cm⁻¹, in accordance with pristine ANS. The two strong absorption bands at 2848 and 2919 cm⁻¹ are attributed to the antisymmetric and symmetric stretching of $-CH_2$ groups of the alkyl chain.² However, the weak absorption bands at 1384 cm⁻¹ remain exist in the ANS/surfactant-LDH composites, confirming that NO₃⁻ was not exchanged by

ANS and HPS molecules completely. The results confirm the co-intercalation of NO_3^- , ANS and surfactant in the galleries of LDH.

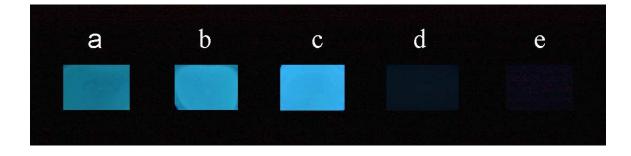


Figure S2. Photographs of (a) ANS/PES-LDH, (b) ANS/HES-LDH, (c) ANS/HPS-LDH, (d) ANS/DES-LDH and (e) ANS/DDS-LDH under UV irradiation of 365 nm.

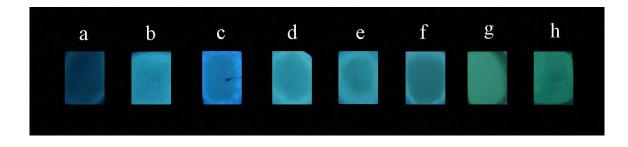


Figure S3. Photographs of the ANS(*x*%)/HPS-LDH films (a ~ h) with *x* = 10%, 15%, 20%, 25%, 30%, 50%, 75%, 100% under UV irradiation of 365 nm.

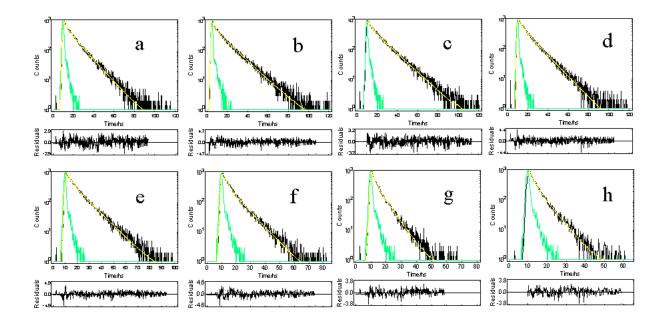


Figure S4. Fluorescence decay curves and residual plots of double-exponential fitting for the ANS(x%)/HPS-LDH film samples with varied *x*: (a) x = 10%, (b) x = 15%, (c) x = 20%, (d) x = 25%, (e) x = 30%, (f) x = 50%, (g) x = 75% and (h) x = 100%.

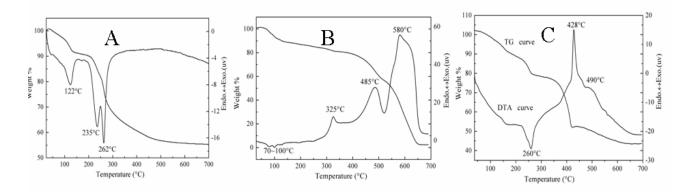


Figure S5. TG-DTA curves for (A) ZnAl-LDH, (B) ANS and (C) ANS(20%)/HPS-LDH.

TG-DTA curves of the pristine ANS, ZnAl-LDH and ANS(20%)/HPS-LDH are displayed in Figure S5, respectively. A weight loss below 125 °C was observed for the ZnAl-LDH (Figure S5 A), due to the loss of crystalline water.³ Upon increasing temperature, two weight-loss stages in 180 ~ 290 °C and 290 ~ 500 °C were observed, which can be attributed to the dehydroxylation and decomposition of nitrate ions⁴. For the thermal decomposition of pristine ANS (Figure S5 B), the weight loss around 70 ~ 130 °C with an endothermic peak in the DTA curve corresponds to the loss of crystalline water. Subsequently a

rapid weight loss in the temperature range 325 ~ 580 °C occurred accompanied with exothermic peaks in the DTA curve, corresponding to the combustion of the organic molecule. In the case of ANS(20%)/HPS-LDH sample (Figure S5 C), a rapid and major weight loss due to the dehydroxylation and decomposition of interlayer HPS and ANS anions in 180 ~ 560 °C occurs, and the first exothermic peak related to the decomposition of ANS anions shifts from 325 °C (Figure S5 A) to 428 °C (Figure S5 C). Consequently, it can be concluded that the thermal stability of ANS was enhanced to some extent after cointercalation into LDH matrix with HPS owing to the presence of host-guest interactions.

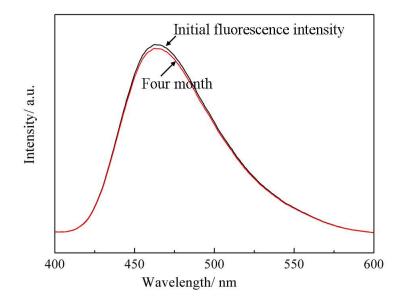


Figure S6. The fluorescence emission spectra of the ANS(20%)/HPS-LDH film (stored at ambient temperature) recorded in 4 month.

Table 1. Chemical compositions and crystalloid parameters for the ANS/surfactant-LDH composites.

Chemical composition	2θ(°)	$d_{003}(\text{\AA})$	FWHM for (003)
$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.072} (PES)_{0.286}(NO_3)_{0.002} \cdot 0.97H_2O$	5.07	17.4	0.32
$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.081}(HES)_{0.277}(NO_3)_{0.002}\cdot 0.92H_2O$	4.80	18.4	0.27
$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.085}(HPS)_{0.272}(NO_3)_{0.003}\cdot 0.98H_2O$	4.48	19.7	0.35
$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.040}\ (DES)_{0.319}(NO_3)_{0.001}\cdot 1.03H_2O$	4.07	21.8	0.34

$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.025} (DDS)_{0.334}(NO_3)_{0.001} \cdot 0.99H_2O$	3.66	24.1	0.28
$Zn_{0.637}Al_{0.363}(OH)_2(ANS)_{0.211} (NO_3)_{0.152} \cdot 0.97 H_2O$	4.40	19.6	0.40

Nominal <i>x</i>	Final <i>x</i>	Chemical composition	
0	0	$Zn_{0.640}Al_{0.360}(OH)_2(NO_3)_{0.36}\cdot 0.46H_2O$	8.92
10	7.8	$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.028}(HPS)_{0.330}(NO_3)_{0.002}\cdot 0.75H_2O$	19.6
15	13.4	$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.048}(HPS)_{0.309}(NO_3)_{0.003}\cdot 0.82H_2O$	19.6
20	25.8	$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.091}(HPS)_{0.265}(NO_3)_{0.004}\cdot 0.99H_2O$	19.7
25	28.6	$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.102}(HPS)_{0.255}(NO_3)_{0.003}\cdot 0.91H_2O$	19.7
30	36.3	$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.129}(HPS)_{0.226}(NO_3)_{0.005}\cdot 0.89H_2O$	19.7
50	49.1	$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.175}(HPS)_{0.181}(NO_3)_{0.004}\cdot 0.94H_2O$	19.8
75	56.4	$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.199}(HPS)_{0.154}(NO_3)_{0.007}\cdot 0.79H_2O$	19.8
100	100	$Zn_{0.640}Al_{0.360}(OH)_2(ANS)_{0.211}(NO_3)_{0.149}\cdot 0.96H_2O$	20.1

Table 2 Chemical compositions and interlayer distance (d_{003}) of the ANS(x%)/HPS-LDH composites.

Reference:

1 (a) Camerel, F.; Barberá, J.; Otsuki, J.; Tokimoto, T.; Shimazaki, Y.; Chen, L.Y.; Liu, S. H.; Lin, M.

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2 Rajamathi, J. T.; Arulraj, A.; Ravishankar, N.; Arulraj, J.; Rajamathi, M. Langmuir, 2008, 24, 11164–11168.

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