## **Supporting Information**

# One-step hydrothermal growth of porous nickel manganese layered double hydroxide nanosheets film towards efficient visible-light modulation

Xingzhe Feng, Xinyi Wan, Ting Yang, Jiahui Huang, Jinmin Wang, Dongyun $\mathrm{Ma}^*$ 

School of Materials and Chemistry, University of Shanghai for Science and Technology, Shanghai 200093, China

Corresponding authors: D. Ma

Email addresses: dyma@usst.edu.cn

# **Supplementary Figures**



**Fig. S1.** Schematic diagram for the hydrothermal preparation process of NiMn-LDH film and subsequent annealing process.



Fig. S2. TG curves of the NiMn-LDH powders.



Fig. S3. The XPS survey spectrum of the as-grown NiMn-LDH film.



**Fig. S4.** Time-current curves of the NiMn-LDH film measured at -1.2 V for 45 s and 1 V for 35 s.



Fig. S5. Time-current curves of the  $Ni_6MnO_8$  film measured at -1.2 V for 25 s and 1 V for 25 s.



**Fig. S6.** Cycle performance of the NiMn-LDH film recorded at a wavelength of 550 nm.



Fig. S7. Cycle performance of the  $Ni_6MnO_8$  film recorded at a wavelength of 550 nm.



Fig. S8. CV curves of the NiMn-LDH film at different scan rates.



Fig. S9. CV curves of the  $Ni_6MnO_8$  film at different scan rates.



Fig. S10. EIS curves of (a) the NiMn-LDH film and (b) Ni<sub>6</sub>MnO<sub>8</sub> film.

Electrochemical impedance spectroscopy (EIS) was conducted to estimate the conductivity of the as-prepared film electrodes. Fig. S10 shows the resulting Nyquist plots of NiMn-LDH and Ni<sub>6</sub>MnO<sub>8</sub> film electrodes. The EIS curves of both electrodes consist of a straight line and a semicircle. The typical semicircle in the high-frequency region is attributed to the charge transfer resistance ( $R_{ct}$ ) caused by a faradic reaction that occurred at the active material surface.<sup>1,2</sup> Accordingly, the calculated  $R_{ct}$  for the NiMn-LDH and Ni<sub>6</sub>MnO<sub>8</sub> film electrodes are 19.8  $\Omega$  and 49.1  $\Omega$ , respectively.

### **Supplementary Tables**

Samples	Average crystallite	Dislocation density	Strain ( <b>ɛ</b> ×10 <sup>-2</sup> )				
	size (D) nm	$(\delta \times 10^{15}) \text{ m}^{-2}$	nm <sup>-2</sup>				
NiMn-LDH	22.6	0.0021	0.42				
Ni <sub>6</sub> MnO <sub>8</sub>	9.9	0.0037	0.76				

Table S1. Microstructural parameters of different samples

"Various microstructural parameters are calculated from the XRD data and  $2\theta$  values of the diffraction peaks. The average crystallite sizes of the two samples are calculated by using Sherrer's equation as follows:

$$(D) = \frac{0.9 \mathbb{Z}\lambda}{\beta \cos\theta}$$

where  $\lambda$  denotes the wavelength of Cu K $\alpha$  line (1.54 Å),  $\beta$  corresponds to full-width at half maximum, and  $\theta$  is the Bragg's angle.<sup>6</sup> The lattice strain ( $\epsilon$ ) and dislocation density ( $\delta$ ) are also calculated by using the following equation:<sup>7,8</sup>

$$(\varepsilon) = \frac{\beta \cot \theta}{4}$$
$$(\delta) = \frac{1}{D^2}$$

The calculated above parameters for NiMn-LDH and  $\mathrm{Ni}_6\mathrm{MnO}_8$  are summarized in Table S1."

Film	$\Delta T$ (%)	$t_{\rm c}({\rm s})$	$t_{b}(s)$	$CE (\mathrm{cm}^2 \cdot \mathrm{C}^{-1})$	Ref.
ZnO@Ni/Co-LDH	56.0% (550 nm)	0.7	2.7		[1]
NiAl-LDH	69.0% (400 nm)	45	45	30	[2]
PEDOT:PSS/LDH	32.0% (650 nm),	0.27	0.18	159	[3]
NiMn-LDH	68.5% (550 nm)	14.2	26.1	56.2	This work

**Table S2.** Electrochromic properties of the different LDH films.

#### References

- [1] M. Yu, R. Liu, J. Liu, S. Li and Y. Ma, Small, 2017, 13, 1702616.
- [2] M. M. Baig, M. T. Mehran, R. Khan, K. Mahmood, S. R. Naqvi, A. H. Khoja and I. H. Gul, *Surf Coat Tech.*, 2021, **421**, 127455.
- [3] X. Liu, J. Wang, D. Tang, Z. Tong, H. Ji and H. Qu, J. Electroanal. Chem., 2012, 687, 58-63.
- [4] D. Mondal and G. Villemure, ACS Appl. Nano Mater., 2020, 3, 6552-6562.
- [5] A. Zhou, X. Liu, Y. Dou, S. Guan, J. Han and M. Wei, J. Mater. Chem. C, 2016, 4, 8284-8290.
- [6] S. Supriya, S. Das, S. Senapati and R. Naik, J. Am. Ceram. Soc., 2023, 106, 5955-5964.
- [7] P. Priyadarshini, S. Das, D. Alagarasan, R. Ganesan, S. Varadharajaperumal and R. Naik, *Scientific Reports.*, 2021, 11, 21518.
- [8] A. Parida, S. Senapati, S. Samal, S. Bisoyi and R. Naik, ACS Appl. Nano Mater., 2023, 6, 11230-11241.