## Electronic Supplementary Information

## Influence of the negative charge density of metalate nanosheets on their bottom-up synthesis

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(a) Different layered titanates



Trititanate A<sub>2</sub>Ti<sub>3</sub>O<sub>7</sub>

Tetratitanate A<sub>2</sub>Ti<sub>4</sub>O<sub>9</sub>



Pentatitanate  $A_2$ Ti<sub>5</sub>O<sub>11</sub>

Lepidocrocite-type titanate A<sub>0.7</sub>Ti<sub>1.825</sub>O<sub>4</sub>

(b) Different layered tantalates



Monotantalate ATaO<sub>3</sub>

Hexatantalate A<sub>4</sub>Ta<sub>6</sub>O<sub>17</sub>

(c) Different layered niobates



Triniobate  $ANb_3O_8$ Hexaniobate  $A_4Nb_6O_{17}$ Figure S1Crystal structures of different layered metalates

	M-O distance [Å]	Standard deviation of M-O [Å]	Average deviation of O <sup>a</sup> -M-O <sup>b</sup> from 90°	Average deviation of O <sup>a</sup> -M-O <sup>c</sup> from 180°
KNb <sub>3</sub> O <sub>8</sub>	2.03	0.21	10.1°	19.4°
$K_4Nb_6O_{17}$	2.03	0.19	8.2°	16.7°
$Rb_4Nb_6O_{17}$	2.01	0.16	7.9°	16.7°
RbTaO <sub>3</sub>	1.99	0.11	6.6°	15.6°
Na <sub>2</sub> Ti <sub>3</sub> O <sub>7</sub>	1.99	0.18	6.8°	14.3°
$Tl_2Ti_4O_9$	1.98	0.14	9.6°	18.3°
$Cs_2Ti_5O_{11}$	2.01	0.19	9.4°	17.5°
Cs-lepidocrocite	1.99	0.13	9.1°	16.4°

**Table S1** Distortion of  $MO_6$  (M = Nb, Ta, and Ti) octahedera in layered metalates





Figure S2 XPS peaks of Mn3p for manganese oxides with different Mn oxidation states



Figure S3 XRD patterns of the samples prepared by heating mixtures of TMAOH  $5H_2O$  and  $HCoO_2$  at (a) 80, (b) 110, and (c) 140 °C.



**Figure S4** AFM image of the colloidal particles prepared by mixing  $(Li,H)_xCoO_2$  and TMAOH in water at TMAOH/Co = 1.