Sample	Mg (wt %)	Al (wt %)	Mg/Al molar ratio	Mn (wt %)
M-LDO	31.39	19.68	1.79	1.36

## Table S1 Elemental analysis of as-prepared M-LDO sample in this work.



Fig. S1 Energy Dispersive X-ray (EDX) spectrum of M-LDH.

Note that a mass of C element was because of the use of conductive adhesive for SEM characterization.



Fig. S2 (a, b) low and (c) high magnification TEM images of M-LDH nanocomposites and the corresponding SAED patterns (d).

Sample	Lattice parameter							
	d <sub>003</sub> (Å)	d <sub>110</sub> (Å)	a (Å)	c (Å)				
LDH	7.4614	1.5153	3.0306	22.3842				
M-LDH	7.6644	1.5234	3.0468	22.9932				

Table S2 Lattice parameters of as-obtained materials in this work.

Note:  $d_{003}$  and  $d_{110}$  represent the interlayer distance and the average metal-mental distance in the brucite-like layer, respectively. And  $a = 2 d_{110}$ ,  $c = 3 d_{003}$ .



Fig. S3 Comparison of (a)  $N_2$ -sorption isotherms and (b) pore-size distribution of the samples asprepared in this work.

Sample	Removal of MO (%)	BET surface area (m <sup>2</sup> g <sup>-</sup>	Average	pore	size
		1)	(nm)		
LDH	41.73	8.4616	9.0738		
LDO	51.56	13.7174	7.7690		
M-LDH	53.18	27.0836	11.6723		
M-LDO	97.93	31.5640	11.9708		

**Table S3** Removal of MO, BET surface area and average pore size of the as-prepared samples in this study.



**Fig. S4** (a) The survey XPS spectrum and high-resolution Mn 2p (a1) of M-LDH; (b) The survey XPS spectrum and high-resolution Mn 2p (b1) of M-LDO.



**Fig. S5** Effect of adsorption temperature (303 K, 313 K, 323 K). (conditions: dosage of adsorbent: 10 mg, initial concentration of adsorbate:10 mg L<sup>-1</sup>).



Fig. S6 The contrast of experimental data and predicted isotherm models investigated.



**Fig. S7** SEM images of used adsorbents: (a) after the first time adsorption of MO and (b) calcined at 773 K for 1 h (R0); (c, d) the same process after the fifth thermal regeneration (R5).