**Supporting Information**

**Resource utilization of organic spent adsorbent to prepare three-dimensional sulfate-functionalized layered double oxide for superior removal of azo dye**

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**Fig. S1** XRD patterns of 3D-LDH and 3D-LDO (a); FT-IR spectrum (b), SEM image (c), and Element mapping images (d) of 3D-LDO

(a) The characteristic peaks of 3D-LDH at 2*θ*=11.47°, 23.12°, 34.93°, 60.68°corresponding to the (003), (006), (009), (110) planes were well allocated to the typical phases of MgAl LDHs (Li et al. 2008). After calcination, a series of new peaks belonged to LDO (MgAl2O4) emerged for 3D-LDO accompanying with the disappearance of LDH peaks, suggesting the successful conversion from LDH to LDO (Lei et al. 2017); (b) The broad bands near 3453, 1640 and 457 cm-1 were associated with the hydroxyl group, the interlayer water molecules, and M-O band, respectively. And the peak at 1384 cm-1 was ascribed to -COO-group stretching vibration of citric acid (Tran et al. 2018); (c-d) The SEM image and Element mapping images confirmed the 3D flower-like structure of 3D-LDO composed of O, Mg, and Al elements.



**Fig. S2** Liner fitting curves for MO adsorption on 3S-LDO and 3D-LDO by Langmuir isotherm (a and c) and Freundlich isotherm (b and d)



**Fig. S3** Effect of contact time on the MO adsorption by 3D-LDO



**Fig. S4** Liner fitting curves for MO adsorption on 3S-LDO and 3D-LDO by pseudo-first-order (a and c) and pseudo-second-order (b and d) kinetic models



**Fig. S5** N2 adsorption–desorption isotherm of 3S-LDO (a) and 3D-LDO (b)



**Fig. S6** Zeta-potential of 3S-LDO as a function of pH (a); Effect of pH on the MO adsorption by 3S-LDO (b)

**Table S1** The release of Mg2+, Al3+, and SO42- of 3S-LDO (1 g/L) in deionized water

|  |  |  |  |
| --- | --- | --- | --- |
| Sample | The amounts of released ions (mg/g) | | |
| Mg2+ | Al3+ | SO42- |
| 3S-LDO | 68.90 | 0.57 | 4.41 |
| 3D-LDO | 24.47 | 1.96 | - |

**Table S2** Parameters for adsorption thermodynamic of MO on 3S-LDO and 3D-LDO

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Samples | △*H*0 (kJ/mol) | △*S*0 (J/(mol·K)) | △*G*0 (kJ/mol) | | | *R*2 |
| 25 °C | 35 oC | 45 oC |
| 3S-LDO | -13.79 | -42.158 | -3.91 | -1.66 | -1.59 | 0.9702 |
| 3D-LDO | -20.25 | -65.931 | -3.04 | -1.79 | -0.59 | 0.9864 |

**Table S3** Surface parameters of 3S-LDO and 3D-LDO

|  |  |  |  |
| --- | --- | --- | --- |
| Samples | Specific surface area (m2/g) | Total pore volume (cm3/g) | Average pore diameter (nm) |
| 3S-LDO | 19.64 | 0.11 | 22.75 |
| 3D-LDO | 206.02 | 0.70 | 13.63 |

# References

Lei C, Zhu X, Zhu B, Jiang C, Le Y, Yu J (2017) Superb adsorption capacity of hierarchical calcined Ni/Mg/Al layered double hydroxides for Congo red and Cr(VI) ions. J Hazard Mater 321:801-811. https://doi:10.1016/j.jhazmat.2016.09.070

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