**Supporting Information**

**Hierarchical layered double hydroxide for the removal of charged dyes: The role of an anionic surfactant**

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**Section 1. The analysis of variance (ANOVA)**

The analysis of variance (ANOVA) was employed to determine the statistical significance of all modes as shown in Tables S3 and 4. Apparently, both response surface and contour plot values exhibited low "Prob > F" values (< 0.0500), indicating that the model term was significant for P removal (Hasan *et al.*, 2011). And the value of the coefficient *R*2 were all higher than 0.80 demonstrated the consistence between the calculated and observed results (Olmez, 2009). Therefore, it is reasonable to state that the models were reliable to predict the optimal conditions for MB and MO removal.



**Fig. S1.** Zeta potential analysis of HLDH.

**Table S1.** The features of SDS and dyes.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  | Abbreviation | Chemical formula | Molecular weight (g/mol) | Charge |
| Sodium dodecyl sulfate | SDS | C12H25SO4Na | 288.38 | -1 |
| Methylene blue | MB | C16H18ClN3S | 319.85 | -1 |
| Methyl Orange | MO | C14H14N3SO3Na | 327.33 | +1 |

**Table S2.** Central composite design and response values of MB.

|  |  |  |  |
| --- | --- | --- | --- |
| Run | Temperature (℃) | Initial MB concentration (g/L) | MB removal capacity (mg/g)*Y* |
| *X*1 | *X*2 | Actual | predicted |
| 1 | 25.00 | 50.00 | 37.23 | 35.26 |
| 2 | 49.14 | 125.00 | 60.23 | 57.37 |
| 3 | 35.00 | 125.00 | 45.25 | 49.43 |
| 4 | 45.00 | 50.00 | 37.64 | 36.49 |
| 5 | 20.86 | 125.00 | 57.52 | 55.82 |
| 6 | 35.00 | 125.00 | 49.08 | 49.43 |
| 7 | 35.00 | 125.00 | 53.42 | 49.43 |
| 8 | 35.00 | 231.07 | 75.68 | 67.97 |
| 9 | 25.00 | 200.00 | 60.62 | 66.33 |
| 10 | 35.00 | 125.00 | 53.14 | 49.43 |
| 11 | 35.00 | 18.93 | 21.08 | 24.23 |
| 12 | 35.00 | 125.00 | 46.24 | 49.43 |
| 13 | 45.00 | 200.00 | 60.76 | 67.29 |

**Table S3.** Central composite design and response values of MO.

|  |  |  |  |
| --- | --- | --- | --- |
| Run | Temperature (℃) | Initial MO concentration (g/L) | MO removal capacity (mg/g)*Y* |
| *X*1 | *X*2 | Actual | predicted |
| 1 | 25.00 | 120.00 | 60.2 | 71.75 |
| 2 | 45.00 | 120.00 | 101.13 | 121.91 |
| 3 | 25.00 | 600.00 | 334.81 | 334.53 |
| 4 | 45.00 | 600.00 | 395.65 | 404.60 |
| 5 | 20.86 | 360.00 | 252.14 | 248.42 |
| 6 | 49.14 | 360.00 | 350.21 | 333.43 |
| 7 | 35.00 | 20.59 | 1.23 | -17.38 |
| 8 | 35.00 | 699.41 | 370.21 | 368.32 |
| 9 | 35.00 | 360.00 | 280.63 | 295.63 |
| 10 | 35.00 | 360.00 | 266.25 | 295.63 |
| 11 | 35.00 | 360.00 | 300.62 | 295.63 |
| 12 | 35.00 | 360.00 | 322.32 | 295.63 |
| 13 | 35.00 | 360.00 | 308.35 | 295.63 |

**Table S4.** ANOVA for the quadratic model of MB removal.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Source | Sum of Squares | df | Mean square | F value | P valueProb > F |  |
| Model | 2037.41 | 5 | 407.48 | 13.06 | 0.0019 | significant |
| A–*X*1 | 2.40 | 1 | 2.40 | 0.077 | 0.7895 |  |
| B–*X*2 | 1913.52 | 1 | 1913.52 | 61.35 | 0.0001 |  |
| AB | 0.018 | 1 | 0.018 | 5.843E-004 | 0.9814 |  |
| A2 | 89.32 | 1 | 89.32 | 2.86 | 0.1344 |  |
| B2 | 19.27 | 1 | 19.27 | 0.62 | 0.4577 |  |
| Residual | 218.34 | 7 | 31.19 |  |  |  |
| Lack of Fit | 160.88 | 3 | 53.63 | 3.73 | 0.1178 | not significant |
| Pure Error | 57.46 | 4 | 14.36 |  |  |  |
| Cor Total | 2255.75 | 12 |  |  |  |  |

*R*2 = 0.9032; *R*2adj. = 0.8341; *R*2pre. = 0.4530; Adequate precision = 11.529.

**Table S5.** ANOVA for the quadratic model of MO removal.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Source | Sum of Squares | df | Mean square | F value | P valueProb > F |  |
| Model | 1.814E+005 | 5 | 36284.41 | 77.48 | < 0.0001 | significant |
| A–*X*1 | 7227.74 | 1 | 7227.74 | 15.43 | 0.0057 |  |
| B–*X*2 | 1.488E+005 | 1 | 1.488E+005 | 317.70 | < 0.0001 |  |
| AB | 99.10 | 1 | 99.10 | 0.21 | 0.6594 |  |
| A2 | 38.56 | 1 | 38.56 | 0.082 | 0.7824 |  |
| B2 | 25111.98 | 1 | 25111.98 | 53.63 | 0.0002 |  |
| Residual | 3277.96 | 7 | 468.28 |  |  |  |
| Lack of Fit | 1290.72 | 3 | 430.24 | 0.87 | 0.5282 | not significant |
| Pure Error | 1987.24 | 4 | 496.81 |  |  |  |
| Cor Total | 1.847E+005 | 12 |  |  |  |  |

*R*2 = 0.9823; *R*2adj. = 0.9696; *R*2pre. = 0.9335; Adequate precision = 28.704.

**Table S6.** Comparison of removal capacity of HLDH with different materials for MB and MO

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Species | Adsorbents | T (oC) | Dosage (g/L) | *q*m (mg/g) | Ref. |
| MB | CuS-NP-AC | 25 | - | 10.6 | (Mazaheri *et al.*, 2016) |
| PPy | 25 | 2.00 | 27.8 | (Chen *et al.*, 2016) |
| USM- chitin | 25 | 5.00 | 26.7 | (Dotto *et al.*, 2015) |
| Seaweed-ZnO-PANi | 32 | 0.50 | 20.6 | (Pandimurugan & Thambidurai, 2016) |
| HLDH | 25 | 5.00 | 58.7 | This study |
| MO | ZnAl-LDH | - | - | 248.3 | (Li *et al.*, 2014) |
| KGM/GO | 25 | - | 51.6 | (Gan *et al.*, 2015) |
| TRG | 25 | 0.02 | 89.3 | (Iqbal & Abdala, 2013) |
| Mg/Al LDH | 25 | 0.40 | 148.3 | (Ai *et al.*, 2011) |
| HLDH | 25 | 5.00 | 416.7 | This study |

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