**Tables:**

**Table 1s**. Experimental levels of independent variables for Mt nanoparticles, Mt-Hyamine and Mt-Hyamin-SDS modified montmorillonites.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Independent variables | Levels | | | | |
| -2 (-α) | -1 | 0 | 1 | +2 (+α) |
| pH (A) | 3 | 5 | 7 | 9 | 11 |
| Temperature (°C, B) | 25 | 32.5 | 40 | 47.5 | 55 |
| Adsorbent dosage (g/L, C) | 0.5 | 1 | 1.5 | 2 | 2.5 |
| Initial dye concentration (mg/L, D) | 10 | 32.5 | 55 | 77.5 | 100 |

**Table 2s.** Independent variables in CCD matrix and the response values for Mt-Hyamine, Mt-Hyamine-SDS and Mt nanoparticles.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Run** | **pH** | **Temperature (°C)** | | **Adsorbent dosage (g/L)** | **Initial dye concentration (mg/L)** | **Removal efficiency (%)** (**a)** | **Removal efficiency (%)** (**b)** | **Removal efficiency (%) (c)** |
| 1 | 5 | 47.5 | | 1 | 32.5 | 96.34 | 93 | 98.35 |
| 2 | 11 | 40 | | 1.5 | 55 | 99.95 | 99.7 | 99.99 |
| 3 | 3 | 40 | | 1.5 | 55 | 93.51 | 91 | 98.71 |
| 4 | 5 | 32.5 | | 2 | 32.5 | 98.12 | 99.4 | 99.27 |
| 5 | 7 | 40 | | 0.5 | 55 | 90.45 | 92 | 97.24 |
| 6 | 5 | 32.5 | | 1 | 77.5 | 91.32 | 92.7 | 98.12 |
| 7 | 7 | 55 | | 1.5 | 55 | 99.88 | 98.5 | 98.84 |
| 8 | 7 | 40 | | 1.5 | 55 | 96.72 | 96.2 | 97.98 |
| 9 | 7 | | 40 | 1.5 | 55 | 96 | 97 | 98.32 |
| 10 | 7 | 40 | | 1.5 | 10 | 97.8 | 99.9 | 99.98 |
| 11 | 9 | 47.5 | | 2 | 32.5 | 99.88 | 99.7 | 99.95 |
| 12 | 7 | 40 | | 1.5 | 55 | 97 | 96.2 | 98.24 |
| 13 | 5 | 47.5 | | 1 | 77.5 | 94.07 | 92 | 98.12 |
| 14 | 7 | 40 | | 1.5 | 55 | 96 | 97 | 98.28 |
| 15 | 7 | 25 | | 1.5 | 55 | 97.54 | 95 | 98.26 |
| 16 | 7 | 40 | | 1.5 | 55 | 95.8 | 96.5 | 97.78 |
| 17 | 9 | 47.5 | | 2 | 77.5 | 99.24 | 99.6 | 99.84 |
| 18 | 9 | 32.5 | | 2 | 77.5 | 97.89 | 98.7 | 99.61 |
| 19 | 9 | 32.5 | | 1 | 32.5 | 96.35 | 98.7 | 99.23 |
| 20 | 5 | 47.5 | | 2 | 32.5 | 98.36 | 99.7 | 98.94 |
| 21 | 9 | 47.7 | | 1 | 32.5 | 99.18 | 98.9 | 99.25 |
| 22 | 7 | 40 | | 2.5 | 55 | 99.4 | 99.9 | 99.98 |
| 23 | 9 | 32.5 | | 2 | 32.5 | 99.88 | 99.3 | 99.71 |
| 24 | 9 | 32.5 | | 1 | 77.5 | 94 | 97.4 | 99.31 |
| 25 | 7 | 40 | | 1.5 | 100 | 97.53 | 99 | 99.76 |
| 26 | 5 | 32.5 | | 1 | 32.5 | 93.44 | 94.6 | 98.28 |
| 27 | 5 | 32.5 | | 2 | 77.5 | 95.36 | 98 | 99.12 |
| 28 | 5 | 47.5 | | 2 | 77.5 | 97.6 | 97.8 | 99.12 |
| 29 | 7 | 40 | | 1.5 | 55 | 97 | 95.5 | 97.68 |
| 30 | 9 | 47.5 | | 1 | 77.5 | 97.92 | 96.9 | 99.54 |

**Table 3s**. Kinetic parameters of CV adsorption on Mt-Hyamine.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Models** | **parameters** | ***C*0 (mg/L)** | | | |
| **4** | **6** | **8** | **10** |
| **Pseudo first**  **order** | *k1* (min-1) | 0.1697 | 0.1683 | 0.2589 | 0.2202 |
| R2 | 0.9323 | 0.9161 | 0.9559 | 0.9792 |
| **Pseudo second order** | *k2* (mg g−1 min−1) | 0.0792 | 0.0543 | 0.0610 | 0.0417 |
| R2 | 0.9898 | 0.9944 | 0.9954 | 0.9816 |
| **Elovich** | *α* (mg/g.min) | 53.98 | 63.59 | 574.7 | 115.86 |
| *β* (g/mg) | 2.5641 | 2.613 | 3.185 | 2.697 |
| R2 | 0.7961 | 0.8884 | 0.7745 | 0.7227 |
| **MPFO** | *kM* (min)-1 | 0.0167 | 0.0203 | 0.0216 | 0.0258 |
| R2 | 0.7030 | 0.8351 | 0.8090 | 0.8387 |
| **Intra particle diffusion** | *ki* | 0.1494 | 0.2322 | 0.2772 | 0.3338 |
| *I* | 2.0620 | 3.0892 | 4.6133 | 5.7687 |
| R2 | 0.5235 | 0.5666 | 0.4662 | 0.4640 |
| **Langmuir** | *kL* (min)-1 | 0.00048 | 0.0018 | 0.0023 | 0.0506 |
| *a* | 0.9984 | 0.9993 | 0.9950 | 0.8541 |
| R2 | 0.9895 | 0.9936 | 0.9951 | 0.9843 |
| **Fractal-Langmuir** | *f* | 0.9927 | 0.9997 | 0.9917 | 0.9994 |
| *n* | 1.021 | 0.8215 | 0.8509 | 1.033 |
| *kFL* (min)-α | 0.0021 | 0.0001 | 0.0042 | 0.0002 |
| R2 | 0.9980 | 0.9978 | 0.9972 | 0.9978 |

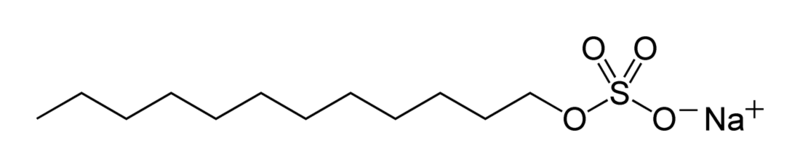
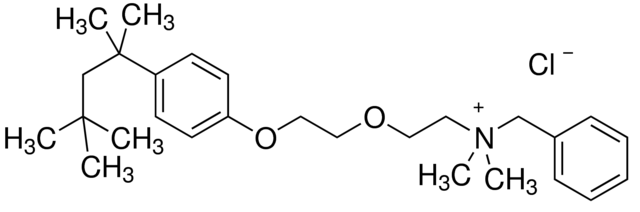
**Table 4s**. Kinetic parameters of CV adsorption on Mt-Hyamine-SDS.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Models** | **parameters** | ***C*0 (mg/L)** | | | |
| **4** | **6** | **8** | **10** |
| **Pseudo first order** | *k1* (min-1) | 0.0484 | 0.0366 | 0.0353 | 0.0775 |
| R2 | 0.9934 | 0.9972 | 0.9813 | 0.9674 |
| **Pseudo second order** | *k2* (mg g−1 min−1) | 0.0239 | 0.1167 | 0.0087 | 0.0149 |
| R2 | 0.9760 | 0.9583 | 0.98400 | 0.9908 |
| **Elovich** | *α* (mg/g.min) | 0.9018 | 0.7696 | 1.316 | 5.02 |
| *β* (g/mg) | 1.3672 | 0.7928 | 0.6579 | 0.6193 |
| R2 | 0.9314 | 0.9417 | 0.9704 | 0.9379 |
| **MPFO** | *kM* (min)-1 | 0.0194 | 0.0195 | 0.0149 | 0.0227 |
| R2 | 0.9764 | 0.9956 | 0.9562 | 0.9708 |
| **Intra particle diffusion** | *ki* | 0.1995 | 0.4217 | 0.3349 | 0.4802 |
| *I* | 1.1873 | 1.9019 | 1.2789 | 3.7027 |
| R2 | 0.7405 | 0.8243 | 0.7830 | 0.7135 |
| **Langmuir** | *kL* (min)-1 | 0.00007 | 0.00005 | 0.00005 | 0.0144 |
| *a* | 0.9991 | 0.9991 | 0.9991 | 0.8866 |
| R2 | 0.9762 | 0.9586 | 0.9841 | 0.9972 |
| **Fractal-Langmuir** | *f* | 0.9993 | 0.9991 | 0.9991 | 0.9990 |
| *n* | 1.16 | 1.27 | 1.64 | 1.42 |
| *kFL* (min)-α | 0.00006 | 0.00002 | 0.00007 | 0.00002 |
| R2 | 0.9983 | 0.9985 | 0.9989 | 0.9969 |

**Table 5s**. Thermodynamic parameters of CV adsorption on Mt-Hyamine and Mt-Hyamine-SDS.

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
|  | adsorbent |  |  | Temperature (°C) | *∆G*°(kJ/mol) | *∆H*°(kJ/mol) | *∆S*° (kJ/mol K) |
|  |  |  |  | 25 | -42.406 |  |  |
|  | Mt-Hyamine |  |  | 40 | -44.775 | +4.179 | +0.156 |
|  |  |  |  | 47 | -45.872 |  |  |
|  |  |  |  | 55 | -47.120 |  |  |
|  |  |  |  | 25 | -36.267 |  |  |
|  | Mt-Hyamine-SDS |  |  | 40 | -38.455 | +7.213 | +0.145 |
|  |  |  |  | 47 | -39.476 |  |  |
|  |  |  |  | 55 | -40.642 |  |  |

**Figures:**



b)

a)

**Figure 1s**. The chemical structure of a) Hyamine surfactant and b) SDS surfactant

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**Figure 2s.** N2 adsorption-desorption isotherms of a) Mt, b) Mt-Hyamine and c) Mt-Hyamine-SDS.

**C:\Users\X452\AppData\Local\Temp\Rar$DIa0.351\5.tifC:\Users\X452\AppData\Local\Temp\Rar$DIa0.531\6.tif**

**C:\Users\X452\AppData\Local\Temp\Rar$DIa0.530\7.tif**

**Figure 3s**. The EDX micrograph of (a) Mt nanoparticles, (b) Mt-Hyamine and (c) Mt-Hyamine-SDS

**C:\Users\X452\AppData\Local\Temp\Rar$DIa0.872\2a.tifC:\Users\X452\AppData\Local\Temp\Rar$DIa0.573\2b.tif**

**C:\Users\X452\AppData\Local\Temp\Rar$DIa0.408\2c.tif**

**Figure 4s**. The actual data versus the predicted data for dye removal of a) Mt, b) Mt-Hyamine and c) Mt-Hyamine-SDS.

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C:\Users\X452\AppData\Local\Temp\Rar$DIa0.036\3b.tif

**C:\Users\X452\AppData\Local\Temp\Rar$DIa0.996\3c.tif**

**Figure 5s**. The effect of pH and time on dye removal efficiency (*R* %) for a) Mt, b) Mt-Hyamine and c) Mt-Hyamine-SDS.

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**Figure 6s**. The effect of the main parameters pH (A), temperature (B), adsorbent dosage (C) and initial dye concentration (D) on dye removal efficiency (*R* %) for a) Mt, b) Mt-Hyamine and c) Mt-Hyamine-SDS.

C:\Users\X452\AppData\Local\Temp\Rar$DIa0.985\5a.tifC:\Users\X452\AppData\Local\Temp\Rar$DIa0.439\5b.tif

**C:\Users\X452\AppData\Local\Temp\Rar$DIa0.469\5c.tifC:\Users\X452\Desktop\5d.tif**

**C:\Users\X452\Desktop\5e.tifC:\Users\X452\Desktop\5f.tif**

**C:\Users\X452\Desktop\5g.tifC:\Users\X452\Desktop\5h.tif**

**C:\Users\X452\Desktop\5i.tif**

**Figure 7s**. Response surface and counter plots for Mt nanoparticles (a, b, c), Mt-Hyamine (d, e, f) and Mt-Hyamine-SDS (g, h, i), (dye removal efficiency: R %)

**Figure 8s**. The plot of ln*K*C versus 1/*T* for CV adsorption on Mt-Hyamine and Mt-Hyamine-SDS*.*