# Ultrasonication-Tailored Graphene Oxide of Varying Sizes in Multiple-Equilibrium-Route-Enhanced Adsorption for Aqueous Removal of Acridine Orange

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**Table S1.** Fitting parameters of the pseudo-first-order and pseudo-second-order models for the adsorption kinetics of CG and EG

| Temp /K | Samples | Experimental capacity (mg/g) | PFO | | | PSO | | |
|---|---|---|---|---|---|---|---|---|
| | | | $Q_e$ (mg/g) | $K_1$ (1/min) | $R^2$ | $Q_e$ (mg) | $K_2$ (g /mg·min) | $R^2$ |
| 283.15 | GO | 1024.44 | 959.73 | 0.45 | 0.9604 | 994.38 | 0.00114 | 0.9682 |
| | GOU10 | 907.04 | 851.35 | 0.34 | 0.9832 | 891.72 | 0.000815 | 0.9933 |
| | GOU30 | 948.89 | 920.58 | 1 | 0.9848 | 926.00 | 0.01 | 0.9823 |
| | GOU60 | 1062.96 | 1024.07 | 0.42 | 0.9932 | 1009.78 | 0.01 | 0.9834 |
| 298.15 | GO | 1111.85 | 1126.70 | 0.42 | 0.9932 | 1104.00 | 0.00263 | 0.9868 |
| | GOU10 | 1331.85 | 1252.01 | 0.36 | 0.9782 | 1200.00 | 0.00206 | 0.9654 |
| | GOU30 | 1278.89 | 1287.00 | 0.43 | 0.9988 | 1263.00 | 0.00274 | 0.9911 |
| | GOU60 | 1228.52 | 1202.96 | 0.57 | 0.9945 | 1192.00 | 0.00691 | 0.9921 |
| 313.15 | GO | 951.85 | 913.97 | 0.42 | 0.9851 | 941.38 | 0.00127 | 0.9891 |
| | GOU10 | 807.41 | 792.62 | 0.62 | 0.9749 | 811.91 | 0.00243 | 0.9795 |
| | GOU30 | 1193.33 | 1208.04 | 0.58 | 0.9931 | 1212.89 | 0.00411 | 0.9908 |
| | GOU60 | 1107.04 | 1129.58 | 1.60 | 0.9839 | 1130.17 | 0.09849 | 0.9838 |
| 283.15 | INRGO | 2145.19 | / | / | / | / | / | / |
| | INRGOU10 | 2054.81 | / | / | / | / | / | / |
| | INRGOU30 | 3158.15 | / | / | / | / | / | / |
| | INRGOU60 | 2320.00 | / | / | / | / | / | / |
| 298.15 | INRGO | 2152.22 | / | / | / | / | / | / |
| | INRGOU10 | 2577.78 | / | / | / | / | / | / |
| | INRGOU30 | 2284.81 | / | / | / | / | / | / |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | INRGOU60 | 3016.30 | / | / | / | / | / | / |
| 313.15 | INRGO | 1912.96 | / | / | / | / | / | / |
| | INRGOU10 | 1920.86 | / | / | / | / | / | / |
| | INRGOU30 | 2440.00 | / | / | / | / | / | / |
| | INRGOU60 | 1858.52 | / | / | / | / | / | / |

**Table S2.** Linear fitting results regarding the capacity of different-size GO as a function of sonication time.

| Parameters / Samples | Intercept | | Slope | | $R^2$ |
|---|---|---|---|---|---|
| | Value | Standard Error | Value | Standard Error | |
| 283.15 CG | 953.31524 | 56.01518 | +1.30069 | 1.6518 | 0.2367 |
| 298.15 CG | 1218.50786 | 83.12502 | +0.77079 | 2.45122 | 0.0471 |
| 313.15 CG | 910.07833 | 117.50939 | +4.19317 | 3.46516 | 0.422 |
| 283.15 EG | 2270.26548 | 434.3065 | +5.97088 | 12.807 | 0.09803 |
| 298.15 EG | 2215.84476 | 204.86201 | +11.67731 | 6.04105 | 0.6513 |
| 313.15 EG | 2025.80762 | 247.04644 | +0.2911 | 7.285 | 0.0007 |

**Table S3.** Fitting parameters of Langmuir, Freundlich, and Tempkin adsorption isotherms of CG.

| Temp /K | Samples | Experimental capacity maximum (mg/g) | Freundlich | | | Langmuir | | | Tempkin | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | n | $K_L$ (L/mg) | $R^2$ | $Q_m$ (mg/g) | $K_L$(L/mg) | $R^2$ | $B_T$ (KJ/ml) | $A_T$ (L/mg) | $R^2$ |
| 283.15 | GO | 1806.67 | 4.68 | 699.96 | 0.95 | 1850.63 | 0.04 | 0.77 | 173.94 | 17.75 | 0.83 |
| | GOU10 | 984.44 | 6.25 | 419.20 | 0.90 | 781.85 | 2.29 | 0.81 | 90.06 | 146.77 | 0.93 |
| | GOU30 | 1066.67 | 7.07 | 580.12 | 0.74 | 1060.52 | 21.3 | 0.92 | 120.78 | 115.09 | 0.83 |
| | GOU60 | 926.67 | 9.87 | 648.88 | 0.68 | 981.28 | 14.92 | 0.94 | 83.46 | 2895.16 | 0.77 |
| 298.15 | GO | 2025.19 | 8.95 | 1039.39 | 0.88 | 1768.83 | 9.92 | 0.87 | 143.74 | 2754.65 | 0.95 |
| | GOU10 | 1518.52 | 8.49 | 806.89 | 0.87 | 1378.95 | 12.21 | 0.86 | 127.04 | 829.91 | 0.95 |
| | GOU30 | 1727.41 | 8.53 | 685.66 | 0.76 | 1095.59 | 10.74 | 0.90 | 102.24 | 1089.06 | 0.83 |
| | GOU60 | 1848.15 | 8.91 | 669.59 | 0.74 | 1047.14 | 13.71 | 0.90 | 93.74 | 1734.60 | 0.82 |
| 313.15 | GO | 897.04 | 8.96 | 559.64 | 0.79 | 899.37 | 2.25 | 0.95 | 76.38 | 2113.72 | 0.89 |
| | GOU10 | 1644.44 | 9.30 | 652.32 | 0.86 | 975.16 | 64.51 | 0.93 | 77.58 | 9015.33 | 0.92 |
| | GOU30 | 913.33 | 9.21 | 543.69 | 0.79 | 830.68 | 17.28 | 0.93 | 71.99 | 2640.40 | 0.86 |
| | GOU60 | 1028.15 | 9.08 | 426.68 | 0.80 | 902.60 | 2.03 | 0.95 | 109.63 | 42.93 | 0.87 |

**Table S4.** Fitting parameters of Freundlich, Langmuir, and Tempkin adsorption isotherms of EG.

| Temp /K | Samples | Maximum experimental capacity (mg/g) | Freundlich | | | Langmuir | | | Tempkin | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | n | $K_L$ (l/mg) | $R^2$ | $Q_m$ (mg/g) | $K_L$ (l/mg) | $R^2$ | $B_T$ (KJ/ml) | $A_T$ (l/mg) | $R^2$ |
| 283.15 | INRGO | 2191.11 | 5.18 | 769.32 | 0.97 | 1741.89 | 0.754 | 0.77 | 175.56 | 260.85 | 0.92 |
| | INRGOU10 | 2808.15 | 5.12 | 984.43 | 0.93 | 2893.37 | 0.066 | 0.72 | 227.96 | 264.69 | 0.90 |
| | INRGOU30 | 2745.93 | 4.32 | 796.60 | 0.97 | 2617.07 | 0.086 | 0.77 | 243.11 | 81.19 | 0.90 |
| | INRGOU60 | 2316.30 | 5.09 | 839.15 | 0.97 | 2033.12 | 0.383 | 0.87 | 203.42 | 170.94 | 0.92 |
| 298.15 | INRGO | 2870.37 | 4.11 | 818.39 | 0.98 | 2732.53 | 0.114 | 0.86 | 203.42 | 170.94 | 0.95 |
| | INRGOU10 | 2586.67 | 4.19 | 749.85 | 0.97 | 2546.25 | 0.088 | 0.83 | 267.91 | 65.47 | 0.92 |
| | INRGOU30 | 3123.70 | 4.84 | 1022.25 | 0.96 | 3065.11 | 0.078 | 0.77 | 215.37 | 165.17 | 0.89 |
| | INRGOU60 | 2940.00 | 3.36 | 641.30 | 0.99 | 2858.39 | 0.084 | 0.94 | 243.22 | 291.03 | 0.91 |
| 313.15 | INRGO | 2200.74 | 4.42 | 656.82 | 0.97 | 2063.88 | 0.105 | 0.82 | 169.04 | 266.21 | 0.88 |
| | INRGOU10 | 2481.48 | 3.73 | 575.23 | 0.98 | 2316.59 | 0.071 | 0.87 | 213.98 | 51.62 | 0.88 |
| | INRGOU30 | 2374.07 | 3.79 | 595.34 | 0.95 | 2422.29 | 0.059 | 0.82 | 328.84 | 15.73 | 0.81 |
| | INRGOU60 | 4322.96 | 1.61 | 174.92 | 0.98 | 8495.96 | 0.006 | 0.95 | 328.84 | 15.73 | 0.58 |

**Table S5.** Deconvolution of C1s XPS spectra for individual CG and EG.

| Samples | C-C (284.8 eV) | C-O (286.5 eV) | C=O (288.5 eV) | $\pi \rightarrow \pi^*$ (290.9 ev) |
|---|---|---|---|---|
| GO | 46.8% | 45.95% | 6.84% | 0.41% |
| GOU10 | 56.03% | 35.72% | 8.14% | 0.11% |
| GOU30 | 47.56% | 43.02% | 8.87% | 0.55% |
| GOU60 | 55.93% | 36.09% | 7.92% | 0.06% |
| INRGO | 53.82% | 34.52% | 10.41% | 1.25% |
| INRGOU10 | 59.28% | 27.81% | 11.88% | 1.02% |
| INRGOU30 | 69.19% | 19.98% | 10.71% | 0.12% |
| INRGOU60 | 54.92% | 33.36% | 10.91% | 0.81% |

**Table S6.** Isotherm adsorption behavior of the CG and EG at varying temperatures.

| Samples | 283.15 K | 298.15 K | 313.15 K |
|---|---|---|---|
| GO | Freundlich | Tempkin | Langmuir |
| GOU10 | Tempkin | Tempkin | Langmuir |
| GOU30 | Langmuir | Langmuir | Langmuir |
| GOU60 | Langmuir | Langmuir | Langmuir |
| INGGO | Freundlich | Freundlich | Freundlich |
| INRGOU10 | Freundlich | Freundlich | Freundlich |
| INRGOU30 | Freundlich | Freundlich | Freundlich |
| INRGOU60 | Freundlich | Freundlich | Freundlich |

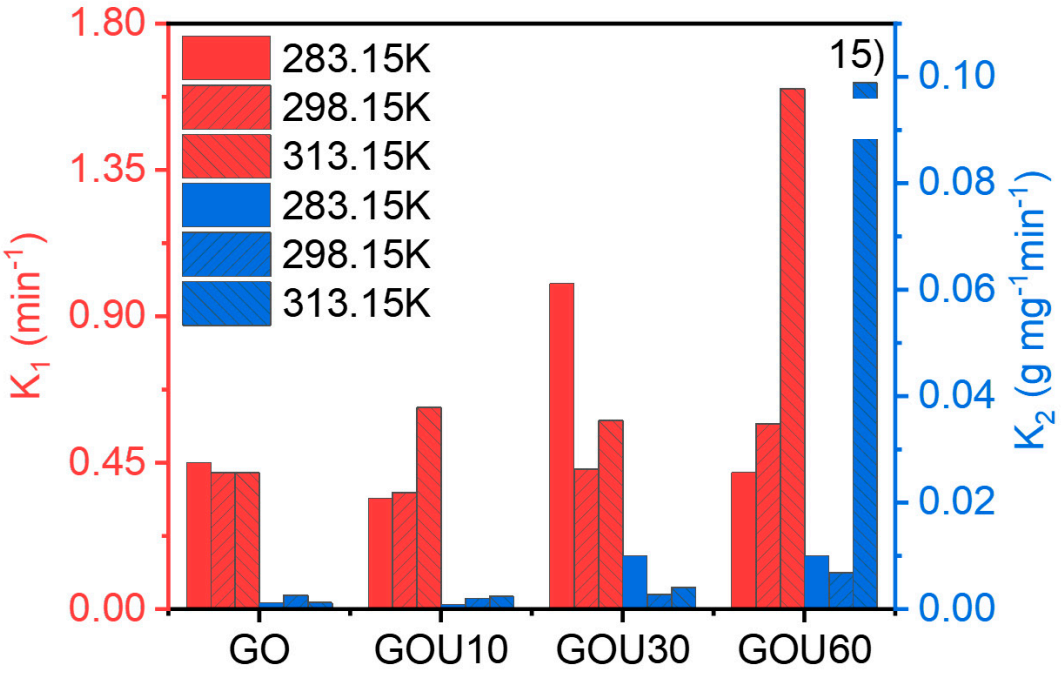


**Figure S1.** Comparison of pseudo-first-order and pseudo-second-order adsorption rate constants between control and experimental groups

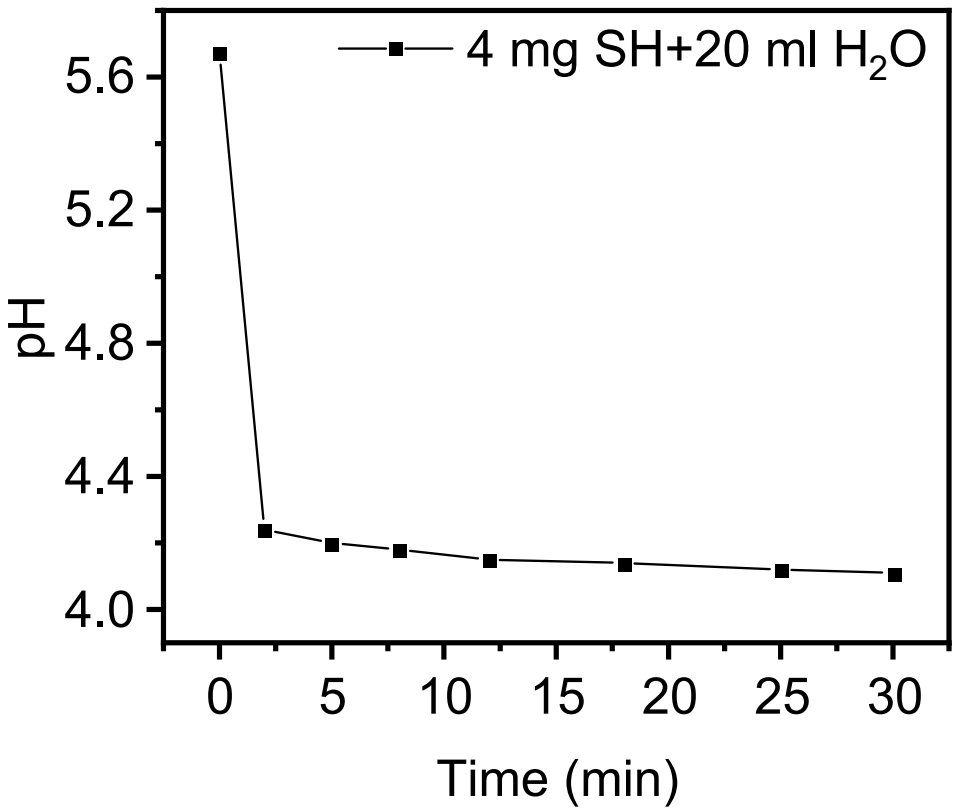


**Figure S2.** Effect of SH on the pH of aqueous solutions as a function of time

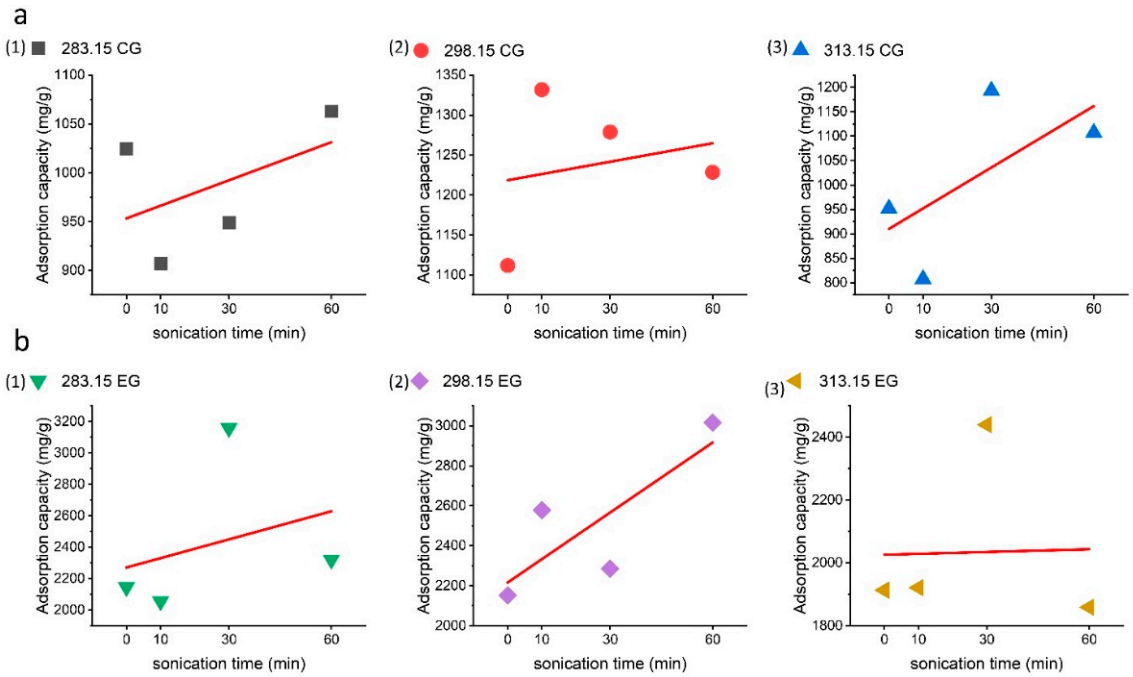


**Figure S3.** Statistics of the adsorption capacities of (**a**) CG and (**b**) EG at varying temperatures as a function of ultrasonication time. The kinetics adsorption capacity data collected from the experiments with the initial AO concentration at 100 mg/l were analyzed.