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Removal of Cu^{2+} from aqueous solution by an optimal synthetic humic-like acid formed by an abiotic humification process

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Humic acid can effectively bind several hazardous metals and has been regarded as a promising adsorbent. Previous studies indicate that carboxylic groups dominate Cu^{2+} binding to humic acid. In this study, to determine the optimal synthesis conditions by which humic acid with higher COOH contents could be produced, synthetic humic-like acids (SHLAs) were synthesized under 11 different abiotic humification conditions, including different temperatures (25 ˚C - 45 ˚C), pH (6 - 8), precursor species (glycine, catechol and glucose), initial precursors' concentrations (0.25M:0.25M - 1M:1M glycine:catechol) and MnO_2catalyst addition amount (1.3% -2.5%). The results indicated that higher COOH contents were achieved in systems comprising low concentrations of glycine and catechol (0.25M: 0.25M) at low temperature (25 ˚C), pH=8 and with relatively high catalyst concentrations (2.5% MnO_2). On the basis of this initial study, SHLA was prepared using catechol (0.25M) and glycine (0.25M) with a MnO_2 catalyst (2.5%) at pH = 8 and 25 ˚C. The adsorption behaviour of Cu^{2+}onto SHLA from aqueous solution was investigated, including adsorption efficiency, kinetics, isotherms, and thermodynamics. The SHLA exhibited a range of adsorption efficiencies for Cu^{2+} depending on conditions, e.g. 27 – 99% at pH of 1 - 8, 76 -77 % at temperatures of 25 - 45 ˚C, 54 – 92 % for SHLA concentrations of 0.5 - 2 g/L and 31 – 98 % for initial Cu^{2+} concentrations of 150 - 10 mg/L. The adsorption kinetics were fitted to pseudo-first-order, pseudo-second-order, intraparticle diffusion and Elovich models. The pseudo-second-order model provided the best fit for experimental data (R^2 = 0.9995-1, $p \leq 0.0001$), indicating that chemisorption was most likely the rate-limiting step for adsorption. The equilibrium adsorption data showed good fits to both the Langmuir (R^2 = 0.9928 – 0.9982, $p \leq 0.0001$) and Freundlich (R^2 = 0.9497 – 0.9667, $p \leq 0.0001$) models. The maximum adsorption capacity (q_m) of SHLA increased from 46.44 mg/g to 58.78 mg/g with increasing temperature from 25 ˚C to 45 ˚C. Thermodynamic parameters (ΔG^0=2.50-3.69 kJ/mol; ΔS^0=0.06 kJ/(mol·K); ΔH^0=15.23 kJ/mol) and values of R_L(0.0142-0.3711) and n (3.264-3.527) showed that the adsorption of Cu^{2+} onto SHLA was favourable, spontaneous and endothermic in nature. Overall, SHLA shows great potential as both an amendment for Cu^{2+} immobilisation in soil and an adsorbent to remove Cu^{2+} from water and wastewater.

***Keywords*:** Synthetic humic-like acid; Copper ion, Adsorption, Kinetics; Isotherm; Thermodynamics