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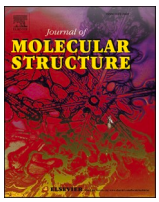

Optimization and mechanistic insight of phenol removal using an effective green kaolin adsorbent through experimental and computational approaches

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ABSTRACT

The discharge of phenols into the aquatic environment has detrimentally affected human health and the aquatic ecosystem. Hence, removing phenols from polluted water bodies has been a global concern. In this study, a natural and low-cost adsorbent, kaolin, was used to remove phenol from aqueous solutions. Experimental and computational approaches were applied to optimize the removal efficiency and provide mechanistic insight into the phenol removal process. Based on the response surface methodology findings, the optimum conditions were pH 1.6, 40 min, 50 mg/L, and 2 g with 92.19 % maximum percentage removal. The regeneration test shows that kaolin retained over 76.76 % of adsorption capacities. SEM and FESEM reveals the surface morphology and effectiveness of kaolin as phenol removal. The best fit for the Isotherm adsorption for the phenol on kaolin is Freundlich isotherm, which indicated multilayer adsorption. The hydroxyl group from phenol (-OH) and the siloxane group of kaolin (Si-O-Si) shifted, suggesting the presence of a hydrogen bond between kaolin and phenol during adsorption, as determined by one- and two-dimensional IR spectroscopy. Density functional theory calculations were used to support the experimental results by providing information on the Mulliken charge and electronic transition of the complex to improve understanding of the adsorption of phenol on kaolin. Based on the quantum theory of atoms in molecules analysis (QTAIM) and the reduced density gradient non-covalent interaction (RDG-NCI) technique, the chemical reaction occurring during the removal of phenol by kaolin was determined to occur through hydrogen bonding and classified as an intermediate type ($\nabla 2\rho(r) > 0$ and $H < 0$). Further characterizations employing molecular electrostatic potential, global reactivity, and local reactivity descriptors were conducted to investigate the mechanism of the phenol adsorption on kaolin. The findings demonstrated that phenol functions as an electrophile while kaolin acts as a nucleophile during the formation of hydrogen bonds, where the interaction occurred between the O_2 atom of kaolin (electron donor) and the H13 atom of phenol (electron acceptor).

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