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学 位 論 文 要 旨
Dissertation Abstract

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学位論文題目：
Title of Dissertation　ADSORPTION MECHANISM AND SELECTIVITY OF HEAVY METALS AND PESTICIDES ON ZEOLITES AND MONTMORILLONITE
重金属及び殺虫剤のゼオライト及びモンモリロナイトへの吸着選択性と吸着機構）

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The adsorption mechanism and selectivity of heavy metals and an organophosphate pesticide on zeolites and montmorillonite was studied with the objective of assessing the capacity of zeolites and montmorillonite to remove heavy metals (Cu, Pb and Cd) and pesticides (Diazinon). The selectivity sequence of the removal and the factors affecting the removal capacity were studied in detail. The study further analyzed the mechanism of pollutant removal by the aforementioned adsorbents. Various types of zeolites, with variations in their cation exchange capacities (CEC), pore structures, Si/Al ratio and surface area were used in the study. Zeolites A4, faujasite X, faujasite Y, mordenite, Na-P1, clinoptilolite and montmorillonite were used for the heavy metal adsorption experiments. For the diazinon adsorption experiments, iron-modified montmorillonite was used.

The adsorption of Cu onto zeolites and montmorillonite as a reference was conducted at an initial Cu concentration range of 0-0.60 mM in the presence of 100 mM NH_4NO_3 at an initial pH of 5. The samples for the experiments were initially saturated with Na in order to obtain samples that had uniform exchangeable cations. Langmuir and Freundlich models were used in analyzing the equilibrium data and a selectivity sequence derived from the Langmuir calculation was A4 > faujasite X > modernite > Na-P1 ≈ montmorillonite ≈ faujasite Y > clinoptilolite. Zeolites A4 and faujasite X had high adsorptive capacities of 1429 mmol kg^{-1} and 909 mmol kg^{-1}, respectively. Zeolite A4 has the highest CEC among all the samples (6150 mmol kg^{-1}), and the adsorption capacity of Cu was largely influenced by the CEC of the samples. The adsorption mechanism was based on the exchange of Cu from solution with mostly Na which was the main exchangeable cation available.

Following the Cu adsorption experiment, Pb adsorption experiments were conducted. The adsorption of Pb on zeolites A4, faujasite X, faujasite Y and mordenite was studied at various initial pH with the purpose of assessing the pH dependence of Pb adsorption. The adsorption was conducted using 0-0.6 mM $PbNO_3$ in the presence of 100 mM NH_4NO_3 and pH adjustment done using HNO_3. The coexisting NH_4NO_3 served as a representative of other cations available in nature. The study was conducted at initial solution pH ranging from 3-5. Adsorption results were analyzed using Langmuir isotherm analysis. Adsorption was noted to be dependent on pH with increasing adsorption as pH increased from 3-5 for zeolites A4, faujasite X and faujasite Y. The adsorption of Pb on mordenite on the other hand did not show any dependence on pH since it was almost constant within the studied pH range. The adsorptive capacities were 2500, 2000, 588 and 179 mmol kg^{-1} for A4, faujasite X, faujasite Y and mordenite, respectively.

In the subsequent experiments, the adsorption of Cd onto zeolites A4, faujasite X, faujasite Y and mordenite was conducted at an initial Cd concentration range of 0-0.60 mM in the presence of 100 mM NH_4NO_3 at initial pH of 5. The samples for the

experiments were initially saturated with Na in order to obtain samples that had uniform exchangeable cations. Similar to the experiments mentioned above, the selectivity sequence was A4 > faujasite X > faujasite Y > modernite. Cation exchange was also the main factor that influenced adsorption.

Lastly, the adsorption of Diazinon, and organophosphate pesticide was conducted using montmorillonite as an adsorbent. Montmorillonite modified with iron was used. Two different types of iron-modified montmorillonite, each having different contents of iron and synthesized with different pH and levels of Fe hydrolysis were used. One was denoted “Fe-modified” and the other denoted as “FeOH- modified”. The color of the samples changed from greyish green to light-reddish brown after the modification. X-ray diffraction and physical observations were used for characterization of the samples. The d-spacing of the samples was greater than 15Å, indicating the formation of iron hydroxides in the interlayer space of montmorillonite. The amount of adsorption was calculated from the difference between the initial and final concentration of diazinon. The adsorption data were analyzed using the Langmuir adsorption isotherms. The amounts of diazinon adsorbed were 58.8 and 54.1 mmol kg^{-1} for Fe-modified and FeOH-modified respectively. The steep rise in their adsorption isotherms indicated the possibility of adsorption for low levels of diazinon in polluted water.

The results of this study can be used in designing or planning for the clean-up of polluted water using adsorption techniques. An important attribute of these findings was that the samples studied were shown to have the capacity of removing even a very low concentration of the studied pollutants, a property which is hardly achievable by most adsorbents. The results of this study are also important is assisting decision makers in choosing the most efficient, cost effective and environmentally friendly method of removing Cu, Pb, Cd and diazinon from polluted environments. These results can as well apply to other heavy metals and some organophosphate pesticides.

Key words: Heavy metals; Pesticides; Cu adsorption; Pb adsorption, Cd adsorption, Selectivity; Cation exchange Capacity, Langmuir isotherm analysis; Freundlich isotherm analysis.