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ENERGETICS OF WATER MOLECULE ADSORPTION ON MODIFIED BENTONITE SURFACES

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Abstract: In this study, the energetics of adsorption of water molecules on modified (enriched and activated) Dehkanabad bentonite were studied in depth. Based on experimental data, adsorption isotherms and differential heats of adsorption (Q_d) were determined, and molar differential entropy (ΔS_d) values were calculated using the Gibbs–Helmholtz equation. The results of the study showed that water molecules adsorbed on the surface of modified bentonite in a stepwise manner, and the values of adsorption heat and entropy changed depending on the degree of surface filling. In the initial stages, high-energy adsorption centers were active and characterized by high heat (~ 71.7 kJ/mol), while in the later stages, the heat decreased and manifested itself in the form of plateaus. Entropy analysis revealed that the mobility and disorder of water molecules in the surface phase were sometimes higher than that of liquid water. Kinetic observations showed that the time to reach adsorption equilibrium depends on the degree of surface coverage and the redistribution of structural cations. The results of this work provide an important scientific basis for assessing the efficiency of modified bentonite-based sorbents in absorbing water vapor and determining the directions of their practical application.

Keywords: Bentonite, modification, adsorption, differential heat, isotherm, entropy, termokinetic.

Introduction. Bentonite clays are widely recognized as low-cost yet highly effective sorbent materials in various adsorption-based technologies. Their applications span the removal of a wide range of pollutants, including metal ions [1], phenolic compounds [2], organic molecules [3], polymers [4], pesticides [5], radionuclides [6], and dyes [7]. Despite their notable adsorption capacity, natural bentonite clays often require additional modification to optimize their physicochemical properties for specific industrial or environmental protection purposes.

Various modification techniques can be employed to enhance the surface reactivity and tailor the adsorption performance of bentonite clays. These modifications are designed to alter the structural and surface characteristics of the clay, thereby improving interactions between the clay surface and target adsorbates [8]. In the synthesis of porous materials, increasing the specific surface area is particularly important, as the adsorption capacity of a sorbent is directly related to the available surface area for interaction and inversely related to the average pore size [9].

A range of activation and modification strategies have been developed for natural bentonite clays. The choice of method depends on the physicochemical nature of the target pollutants and the phase (liquid or gaseous) in which the adsorption process takes place. Effective modification should ideally employ cost-efficient activating agents that not only enhance adsorption potential but also ensure economic feasibility by minimizing the production cost of the final sorbent material [10].

In recent years, significant advancements in the fields of clay crystallochemistry and mineralogy have considerably expanded our understanding and research capabilities concerning the surface chemistry of clay-based minerals [11, 12]. These studies have demonstrated that the adsorption behavior of layered silicates is intrinsically linked to their crystal structure and crystallochemical characteristics. Moreover, extensive research has identified a clear relationship between the structural organization of clay minerals and their surface chemical, physicochemical, and colloid-chemical properties [13, 14].

Overall, the modification of bentonite clays represents a promising strategy for developing efficient, selective, and economically viable adsorbents for various environmental and industrial applications—an approach strongly supported by a deeper understanding of their structural and surface chemistry.

Research Object and Methods. The differential heat of adsorption was measured using a DAK 1-1 calorimeter based on the Tian-Kalve model. The adsorption isotherm was determined using a volumetric method. The accuracy of adsorption isotherm measurements was within 0.1%, and the heat measurement accuracy was up to 1% [15].

Benzene was used as the adsorbate. Prior to adsorption experiments, the benzene was purified and dried under vacuum conditions. Its vapor pressure was equilibrated by removing dissolved gases until it matched the vapor pressure data for pure benzene as reported in standard reference tables. The conformity of these results with literature data [16] was confirmed.

Water adsorption on the modified bentonite was carried out at 303 K.

Research Results and Discussion. The adsorption characteristics of water molecules on bentonite chemically modified with 10% acid were systematically investigated to assess changes in surface properties and porosity. Specifically, enriched and activated Dehkanabad bentonites were used as adsorbents. The adsorption isotherm of water vapor was recorded at a constant temperature of 303 K, and the resulting data are presented in Figure 1. Analysis of the isotherm within the relative pressure range of $0.05 < P/P_s < 0.4$ revealed a linear relationship when described using the Brunauer-Emmett-Teller (BET) model based on monolayer adsorption in this interval.

From the linear portion of the BET plot, the monolayer adsorption capacity (a_m) was determined to be 0.01767 mmol/g, indicating a well-developed surface capable of accommodating a significant amount of water molecules in a single adsorbed layer. Additionally, the BET energy constant, which reflects the strength of the interaction between the adsorbate (water) and the adsorbent surface, was calculated as 0.015 mmol/g, suggesting a moderate to strong adsorption affinity.

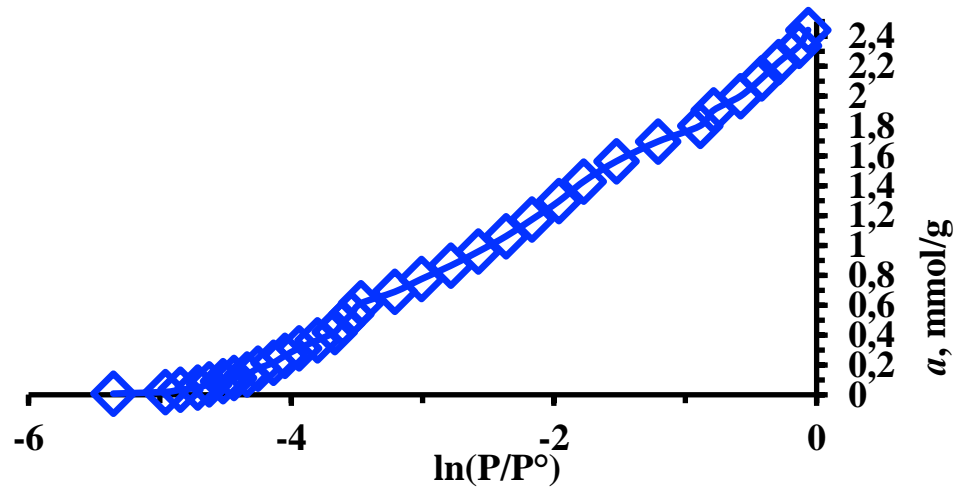


Figure 1. Adsorption isotherms of water molecules on bentonite modified with 10% acid at 303 K

To calculate the specific surface area of the modified bentonite, it was assumed that each water molecule in the monolayer occupies approximately 16 \AA^2 . Based on this assumption, the total specific surface area of the adsorbent was estimated to be $168 \text{ m}^2/\text{g}$. This value highlights a significant increase in surface development resulting from acid treatment, which is likely associated with the removal of impurities and partial dissolution of the clay structure, thereby enhancing porosity and surface accessibility.

These findings provide valuable insights into the physicochemical changes induced by acid activation and emphasize the potential of purified Dehkanabad bentonite for applications involving water vapor adsorption, such as humidity control, catalysis, and environmental remediation.

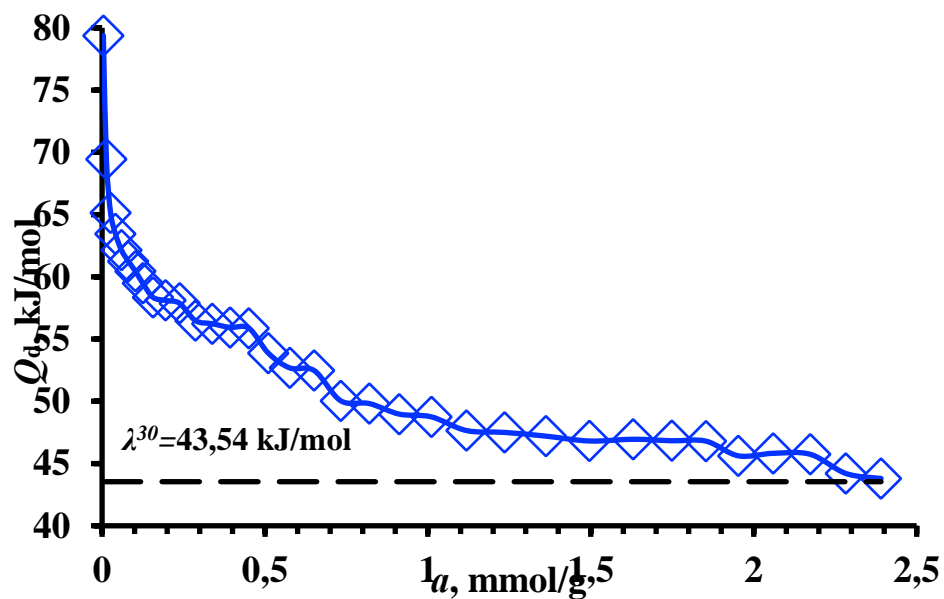


Figure 2. Differential heat of adsorption of water molecules on bentonite modified with 10% acid at 303 K. Dashed lines represent the heat of condensation of water at 303 K.

Due to the stepwise nature of the adsorption heat curve, the corresponding entropy profile exhibits a polyextremal character marked by several inflection points. It is noteworthy that, for adsorption coverages exceeding 500 mmol/g and until the formation of fully hydrated complexes—specifically, the hexahydrated species in the form of $(\text{H}_2\text{O})_6/\text{Na}^+$ —the entropy values remain lower than those of bulk liquid water. However, as the system progresses toward the formation of highly hydrated complexes containing approximately six to nine water molecules per Na^+ ion, the entropy values begin to surpass those of liquid water.

This tendency indicates that the configuration and dynamics of the adsorbed water molecules become increasingly disordered and thermodynamically favorable as the surface coverage progresses, particularly with the expansion of the hydration shell. The sharp increase in differential entropy with progressive adsorption supports the formation of more mobile and less structured water domains at the higher stages of adsorption.

Moreover, it was found that the integral molar entropy of adsorption exceeds that of bulk liquid water by approximately 5.7 J/mol·K. This positive entropy difference indicates that, on average, the mobility and configurational freedom of water molecules in the H_2O –bentonite system are slightly greater than those in the liquid phase. Such behavior may be associated with the heterogeneity of adsorption sites on the bentonite surface, including interlayer regions, edges, and defect-related areas, which offer varying degrees of confinement and interaction energy.

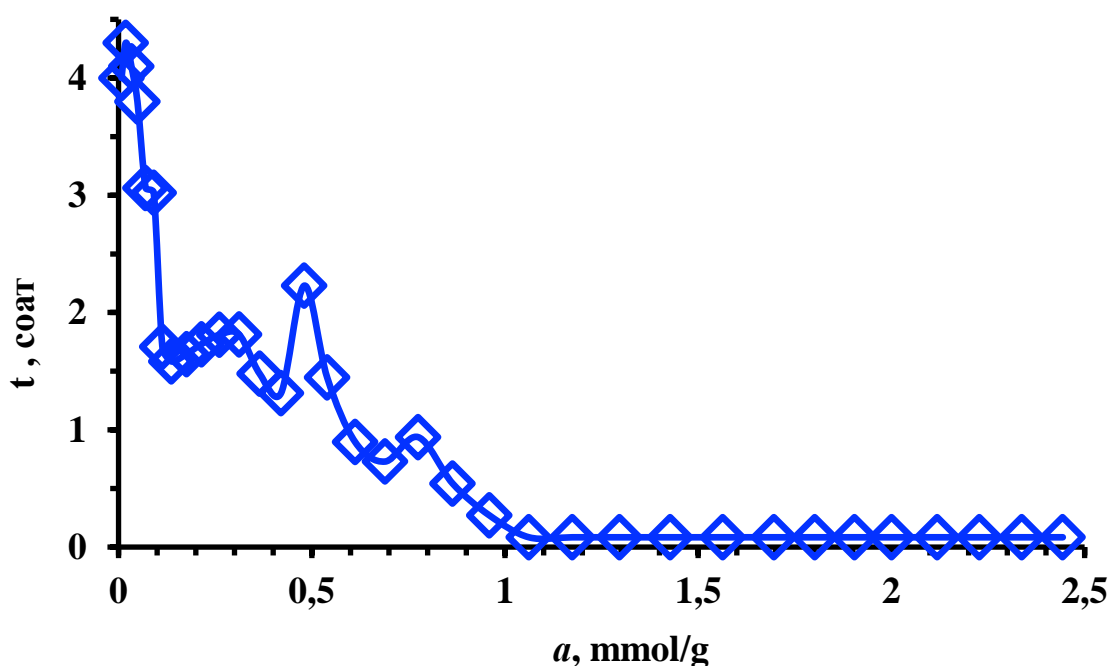


Figure 4. Equilibration time of water molecule adsorption on bentonite modified with 10% acid at 303 K

These findings provide a critical perspective on the thermodynamic landscape of water–clay interactions and indicate that, under certain hydration conditions, adsorbed

water can exhibit dynamic behavior—both in terms of entropy and molecular mobility—that is comparable to or even exceeds that of bulk water.

As illustrated in Figure 4, the relationship between adsorption equilibration time and surface coverage reveals that the time required to reach equilibrium significantly increases at lower surface coverages, particularly around approximately 110 mmol/g. Under such conditions, the system may require up to 5 hours to attain adsorption equilibrium. Conversely, at higher surface coverages, equilibrium is achieved more rapidly, typically within 1 to 1.5 hours.

The pronounced variation in adsorption kinetics as a function of surface coverage suggests the involvement of complex underlying mechanisms governing the adsorption process. Notably, the slower kinetics observed at low coverage levels cannot be solely attributed to the redistribution of water molecules across the adsorbent surface. Rather, this behavior is likely influenced by the redistribution and migration of exchangeable cations within the clay structure.

When water molecules interact with the adsorbent, cations initially occupying fixed positions in the interlayer space may relocate to energetically more favorable adsorption sites. These sites include basal and lateral surfaces as well as structural voids located on both sides of the layered silicate framework. This process of cation redistribution—from relatively stable lattice positions to a more dynamic hydration environment—introduces an additional kinetic barrier that becomes particularly pronounced at lower hydration states.

Therefore, the delay observed in establishing equilibrium at low surface coverages reflects not only the time required for adsorbate molecules to diffuse and reorganize on the surface but also the timescale of structural rearrangements involving cation mobility and coordination changes. These coupled phenomena underscore the importance of considering both physical adsorption and ion-exchange dynamics to fully understand the complex nature of water–clay interactions.

Conclusion. The adsorption process of water molecules on modified (enriched and activated) Dehqonobod bentonite was investigated through comprehensive thermodynamic and kinetic analyses. Based on the obtained differential heat (Q_d) and adsorption isotherm data, it was established that adsorption occurs in a stepwise manner, with distinct energetic states characterizing each stage. In the initial stages, high adsorption heat values (~ 71.7 kJ/mol) were recorded, indicating strong interactions between water molecules and highly energetic active sites. As surface coverage increased, the adsorption heat gradually decreased, approaching values typical of condensation heat, reflecting the progressive occupation of less active sites.

Furthermore, molar differential entropy values (ΔS_d), calculated using the Gibbs–Helmholtz equation, demonstrated that the mobility and degree of disorder of water molecules on the modified bentonite surface vary across different stages. At certain stages, entropy values exceeded those of liquid water, suggesting that water molecules on the surface exhibit even greater freedom of movement than in the bulk liquid state.

Kinetic analyses revealed that the time required to reach adsorption equilibrium is significantly dependent on the degree of surface coverage. At low coverage levels, equilibrium is attained more slowly, a process attributed not only to the diffusion of water molecules but also to the redistribution of structural cations along basal and lateral surfaces.

Overall, water molecule adsorption on modified bentonite surfaces is a complex, multi-stage process governed by the interplay of thermodynamic, kinetic, and structural factors. These findings provide a valuable scientific foundation for the industrial application of bentonite in moisture control, gas separation, and catalytic systems.

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