Study on the Mechanism of Action of Adsorption of Radioactive Particles by Activated Carbon

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Abstract. Radioactive particles usually have a positive charge, while the surface of activated carbon has a negative charge, the activated carbon has an adsorption effect on the radioactive particles. The radioactive particles are adsorbed in the pores and surface of the activated carbon, and will be removed from the environment. In Materials Studio simulation, the activated carbon and molecules to be adsorbed are modeled as a series of particles, and the adsorption process is simulated by simulating the interactions between particles. Using Materials Studio, an activated carbon adsorption model was established to simulate the process of dry activated carbon adsorbing water molecules (H_2O) and methyl iodine (CH_3I). Adsorption isotherms were used to simulate and calculate the adsorption amount and rate, the influence of activated carbon adsorbing H_2O on the adsorption of CH_3I was analyzed. Based on methods such as molecular dynamics simulation and adsorption isotherms, the adsorption rate, adsorption amount, and adsorption position of radioactive particles adsorbed on activated carbon can be predicted.

Keywords: Materials Studio, Activated carbon, Radioactive particles, Simulation, Molecular dynamics

1 Introduction

Activated carbon is a widely used adsorbent for radioactive waste treatment. Its mechanism of action involves adsorption and chemical reaction. The high porosity and surface area of activated carbon endow it with strong adsorption capacity. Radioactive particles usually have a positive charge, while the surface of activated carbon has a negative charge, which allows the activated carbon to adsorb the radioactive particles [1-3]. Radioactive particles are adsorbed in the pores and surface of the activated carbon, thus removing the radioactive particles from the environment. The activated carbon can also remove radioactive substances through chemical reactions. For example, activated carbon can undergo chemical reaction with radioactive elements through oxidation-reduction reaction, converting them into more stable compounds. This chemical reaction can completely remove radioactive elements from environment[4-5].

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In the Materials Studio simulation, the activated carbon and the molecules to be adsorbed are modeled as a series of particles, and the adsorption process is simulated by simulating the interactions between the particles. The advantage of Materials Studio simulation is that it can consider the interaction between adsorbed molecules as well as the influence of the pore structure of the activated carbon. The simulation results can provide information on adsorption rate, adsorbing capacity, and adsorption location. The adsorption isotherm simulation is based on the adsorption isotherm equation to model the adsorption process. In this simulation, the adsorption properties of activated carbon are obtained through experimental measurements and the adsorption process is modeled using isotherm equations. The advantage of this simulation is that the parameters in the adsorption isotherm equation can be determined from the experimental data, improving the accuracy of the simulation results. The two simulation methods can be used in combination to predict the adsorption process more accurately [6-7].

2 Simulation Setup

Calculations were accomplished using Sorption module in MS, mainly using Fixed Pressure mode, to obtain adsorption energy, adsorbing capacity, and system configuration. The parameter settings of adsorption process model was simplified, and high accuracy has been adopted for calculation to obtain data on the impact of humidity on adsorption. The "Add Atom" command was used to add radical group atoms to the upper and lower surfaces. The parameter settings shown in Figure 1.

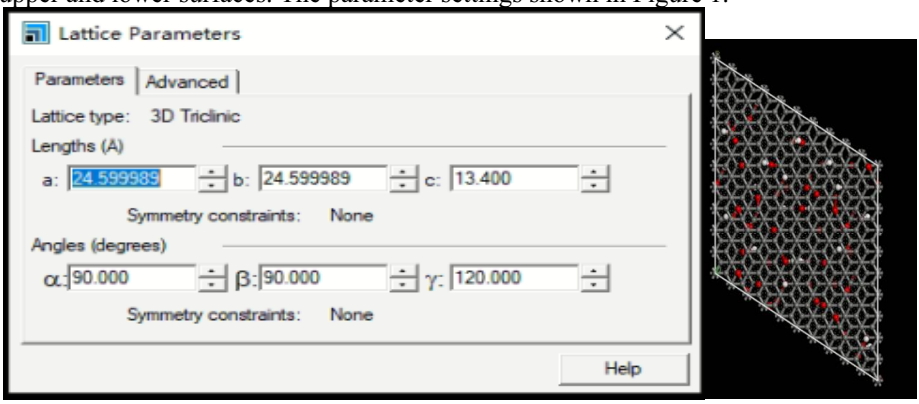


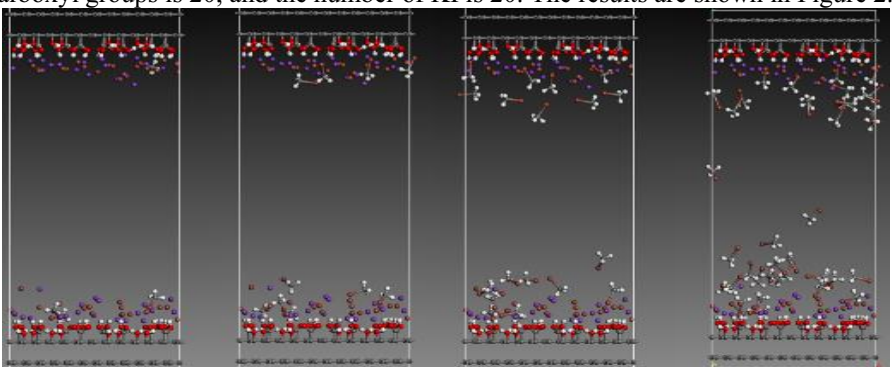
Fig. 1. MS Simulation Setup.

The activated carbon is set as a layer with a certain amount of potassium iodide (KI) and functional groups such as hydroxyl and carboxyl groups (over 100 carbon atoms), and a certain amount of H_2O and CH_3 are added to evaluate the binding and adsorption ability of activated carbon elements, KI, functional groups with H_2O and CH_3I. After completing the simulation of the binding effect of H_2O on activated carbon, a certain amount of CH_3I molecules are added to evaluate the effect of activated carbon that has already adsorbed H_2O on the adsorption of CH_3I.

3 Simulation Results

3.1 Adsorption of CH_3I by Dried Activated Carbon

The CH_3I molecules are added to the activated carbon layer, and the number is 0.02, 0.05, 0.1 and 0.2 times of the carbon atom. The Monte Carlo setting for quantitative adsorption capacity is that the number of simulation steps is 1000000, the temperature is 300 K; the number of surface C atoms is 288, 20 each of the number of hydroxyl carboxyl groups is 20, and the number of KI is 20. The results are shown in Figure 2.



Grey: Carbon; White-Grey-Brown: CH_3I; White-Red: H_2O; Purple-Brown: KI; Grey-Red-White: C-OH; Grey-White-Red: C-COOH

Fig. 2. Simulation of Adsorption of CH_3I on Dried Activated Carbon.

As the number of adsorbed CH_3I increases, it can be seen that the distribution of CH_3I is close in the middle, and the number of adsorption locations close to the C surface is limited and quickly filled. When the adsorption number of CH_3I exceeds 40, a part of the CH_3I is located far from the C surface, indicating that the number of CH_3I has exceeded the amount of CH_3I that can be adsorbed on the C surface. The CH_3I that cannot be adsorbed is in the free states and distributed in the center of the structure.

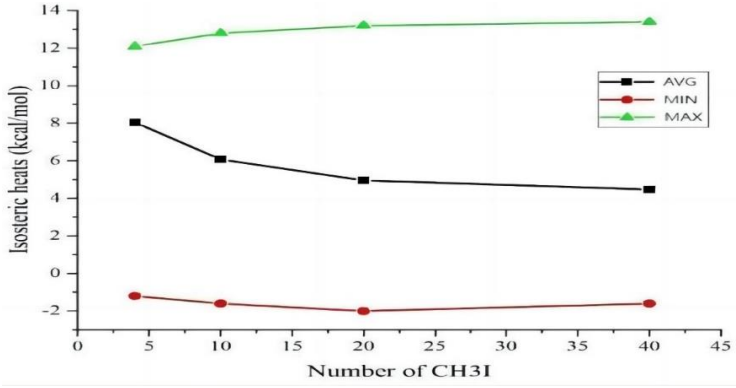


Fig. 3. Number of adsorbed CH_3I - adsorption energy curve.

The number of CH_3I adsorbed-adsorption energy curve shows in figure 3. CH_3I can be spontaneously adsorbed on the C surface. As the number of adsorbed CH_3I increasing, the average adsorption energy increases. This is due to the decrease of adsorption locations with strong relative adsorbing capacity for CH_3I on C surface.

3.2 Binding Effect of H_2O on Activated Carbon

Set up a layer of activated carbon lamella (100 carbon atoms) with a certain number of KI and functional groups such as hydroxyl and carboxyl groups. The number of H_2O is 0.02, 0.05, 0.1, 0.2, and 0.3 times that of carbon molecules. The Monte Carlo setting for quantitative adsorption capacity is that the number of simulation steps is 1000000, the temperature is 300K, the quantities of carbon, hydroxyl and carboxyl groups, and KI are 288, 20, and 20, respectively. The results are shown in Figure 4.

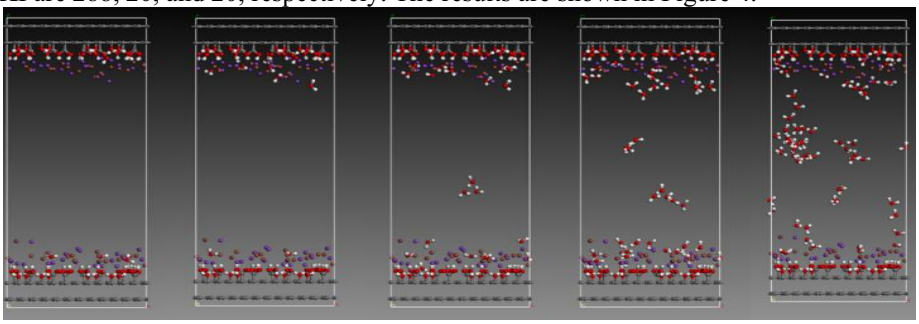


Fig. 4. Adsorption of Water on Dried Activated Carbon.

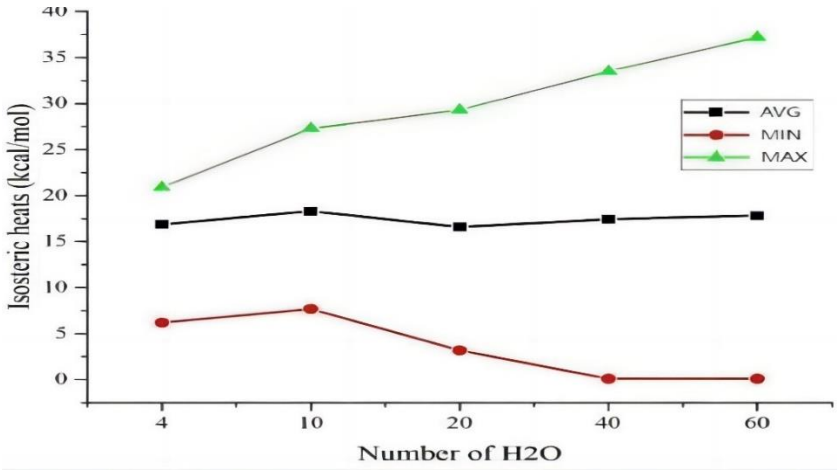


Fig. 5. Number of adsorbed H_2O - adsorption energy curve.

Similar to the results for CH_3I, as the number of adsorbed H_2O increases, the distribution of H_2O is close to the middle positions, it indicates that the number of adsorption sites close to the C surface is limited and quickly filled. The difference is that when the

number of H_2O adsorbed exceeds 20, a part of H_2O can be seen in the position far away from the C surface, it indicates that the adsorption capacity of C surface for H_2O is weaker than that for CH_3I. The reason for this difference may be that the polarity of H_2O is weaker than that of CH_3I, the CH_3I has a stronger charge interaction with surface radical groups on the C surface. The number of H_2O adsorbed-adsorption energy curve shows in figure 5. Similar to CH_3I, the average adsorption energy of H_2O on the C surface is positive, indicating that the adsorption process of H_2O on the C surface is an exothermic reaction, and H_2O can spontaneously adsorb on the C surface. It can also be noted that the value of H_2O adsorption energy is greater than that of CH_3I, it indicates that H_2O adsorbed on the surface of C is more stable than CH_3I.

3.3 Adsorption of CH_3I on Activated Carbon with Adsorbed H_2O

The effect of activated carbon adsorbed H_2O adsorbed the CH_3I is evaluated through adding a single number of CH_3I molecules into the dry activated carbon adsorption model for H_2O, the results are shown in Figure 6, and the Curve of the number of added H_2O and the number of adsorbed CH_3I is shown in figure 7.

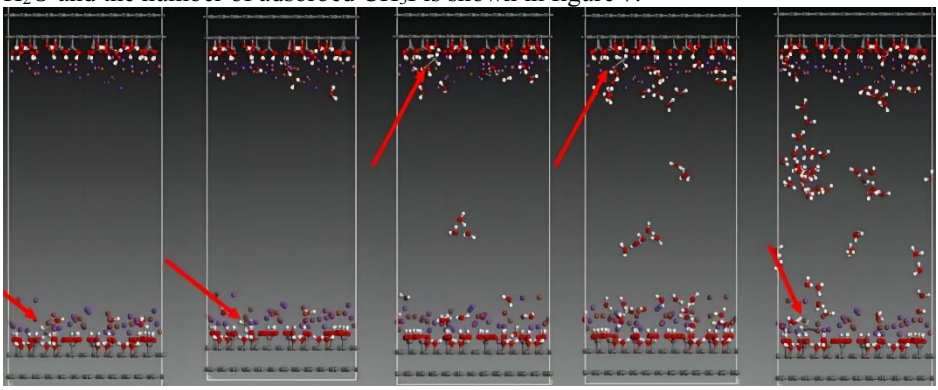


Fig. 6. Simulation of Adsorption of CH_3I on Activated Carbon with Adsorbed H_2O.

By simulating the adsorption of 10 CH_3I molecules on a C surface that has already adsorbed different amounts of water, it can be found that when the number of water is less than 10, CH_3I can be adsorbed well on the surface. When the number of water exceeds 20, some CH_3I begins to move away from the surface, it indicates that when the number of adsorbed water exceeds a certain value, the H_2O adsorbed on the surface cause a decrease in the CH_3I adsorption locations on the C surface.

4 Conclusions

As the number of adsorbed CH_3I increases, the average adsorption energy of C surface for CH_3I increases. This is due to the decrease of adsorption locations with strong relative adsorption capacity for CH_3I on C surface as the amount of CH_3I increases. The

polarity of H_2O is weaker than that of CH_3I, and compared to H_2O, the charge interaction between CH_3I and the surface groups of C is stronger. Therefore, there are many locations on the surface of C that can adsorb CH_3I, indicating that when the amount of adsorbed water exceeds a certain value, the H_2O adsorbed on the surface cause the CH_3I adsorption locations on the surface of C. It indicates that using activated carbon to adsorb radioactive particles requires measures to control the humidity of activated carbon and ensure that the adsorption capacity does not decrease.

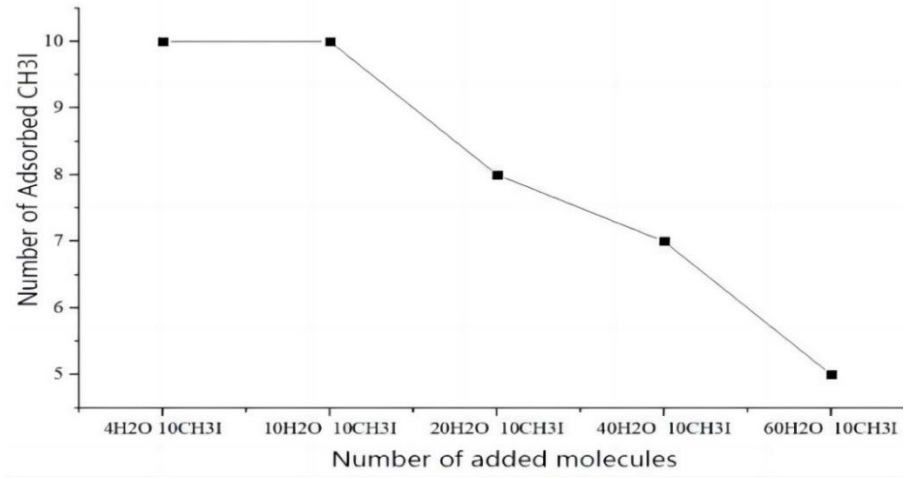


Fig. 7. Curve of the number of added H_2O and the number of adsorbed CH_3I.

References

1. Wang, L.L., Liu, F., Zhan,J., Su, X.D., Chen, X.J., Li, L., Zhou, M.L., Zou, H., Ding, C.C., Xie, H..(2023) Adsorptive Removal of Gaseous Methy Iodide by Triethylenediamine Modified Activated Carbon Fiber. Atomic Enery Science and Technology,57, 1800-1809. 10.7538/yzk.2022.youxian.0869.
2. Obruchikov, A.V., Merkushkin, A.O., Magomedbekov, E.P., (2021) Radioiodine removal from air streams with impregnated UVIS carbon fiber. Nuclear Engineering and Technology,53,1717-1722 10.1016/j.net.2020.10.011.
3. Qiao,B.Q., Liu, C.Y., Zheng, W., Li, Q., Lin, P., Wang, Z.h.,Chen,J.J., Zhou, D.S.,Liu,X.J., Li,Y.G.(2021) Dynamic adsorption properties of activated carbon for radioactive noble gas treatment in offshore floating nuclear power plant. Journal of Radioanalytical and Nuclear Chemistry,327, 207-215. 10.1007/s10967-020-07382-4.
4. Choi, B.S., Kim, S.B., Moon, J., Seo, B.K., (2021) Evaluation of decontamination factor of radioactive methyl iodide on activated carbons at high humid conditions. Nuclear Engineering and Technology. 53, 1519-1523. 10.1016/j.net.2020.10.020.
5. Raschip, I E, Yakimets I, Martin, C P, Paes S S, Vasile C, and Mitchell J R.(2009) Effect of water content on thermal and dynamic mechanical properties of xanthan powder: a comparison between standard and novel techniques. Powder Technology, 182, 436-443. DOI: 10.1016/j.powtec.2007.07.010.

6. Zhang, Q., Gao, Y., Xu, Z., Wang, S., Kobayashi, H., & Wang, J. (2020). The effects of oxygen functional groups on graphene oxide on the efficient adsorption of radioactive iodine. Materials, 13, 1-13. 10.3390/ma13245770.
7. Pelekani C, and Snoeyink V., (2000). Competitive adsorption between atrazine and Methylene blue on activated carbon: The importance of pore size distribution. Carbon: An International Journal Sponsored by the American Carbon Society, 38, 1423-1436.10.1016 /S0008-6223(99)00261-4.