Molecular simulation on adsorption of helium by activated carbon in 4-10 K

X T Xi [1,2], J Wang [1,2], L B Chen [1,2,*], Y Zhou [1,2] and J J Wang [1,2]

[1] CAS Key Laboratory of Cryogenics, Technical Institute of Physics and Chemistry, Beijing 100190, China
[2] University of Chinese Academy of Science, Beijing 100049, China

C2Po2B-08[10] ID:671

E-mail address: chenliubiao@mail.ipc.ac.cn

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1.Introduction

In recent years, the 4K-class regenerative cryocoolers have been successfully used in many felids owing their mechanical simplicity and high reliability. However, in the liquid helium temperature region, the specific heat of the regenerator material is much smaller than that of the helium gas, which increases the loss of the regenerator. Considering that activated carbon can adsorb a large amount of gas molecules at low temperature, using activated carbon with adsorbed helium as regenerator material to reduce the loss is proposed by some scholars. Due to the complexity of low temperature experimental systems, molecular simulation is a better way to analyze the influence of structure, temperature and pressure on the adsorption amount and isosteric heat from the molecular level. In this paper, we using the grand canonical Monte Carbo (GCMC) method to simulate adsorption of helium on amorphous carbons in 4-10 K.

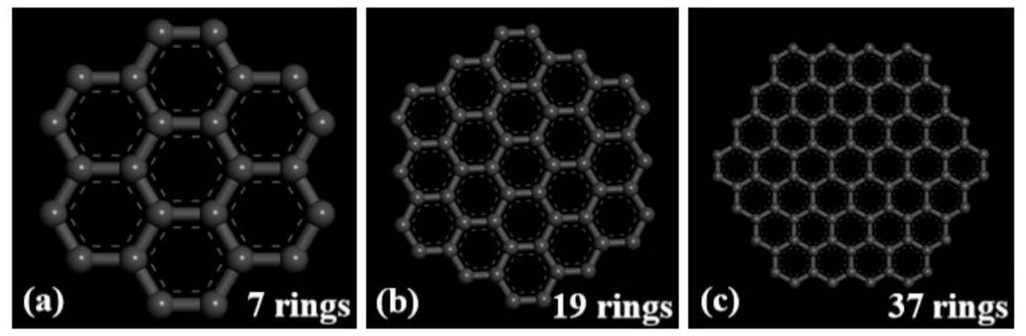
2.Study design

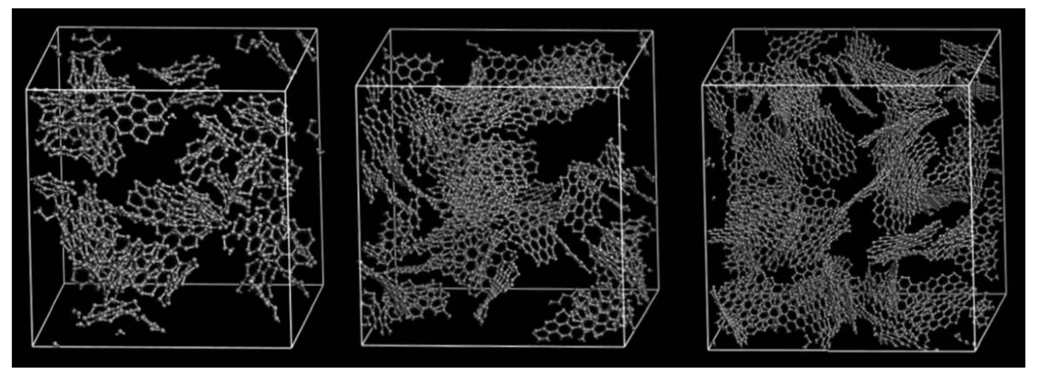
- Using three different size of graphite slices to construct amorphous carbons and analyzing their structural parameters.
- For the adsorption of helium on activated carbon, the effect of temperature, pressure, graphite slice's size and density of amorphous carbon on the adsorption amount and isosteric heat were analyzed.
- Effect of mixed filling method on amorphous carbon structure.

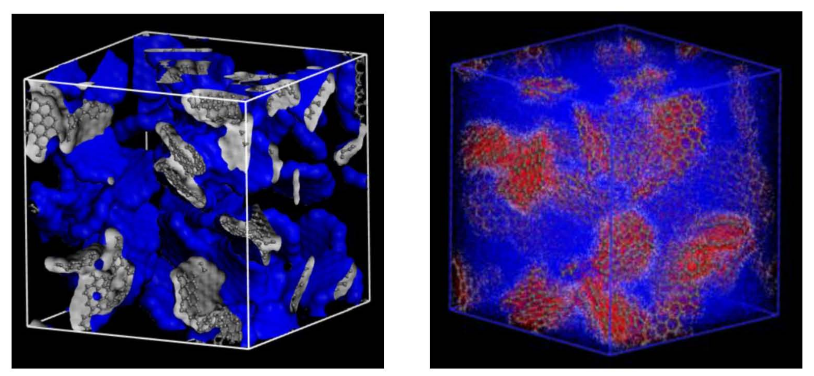
3.Method

(1) Amorphous carbon modeling

Three different size of graphite slices were used to construct different density amorphous carbon structures. Then the Atom Volume & Surface tool in Material Studio was used to obtain the specific surface area and pore volume of amorphous carbon.





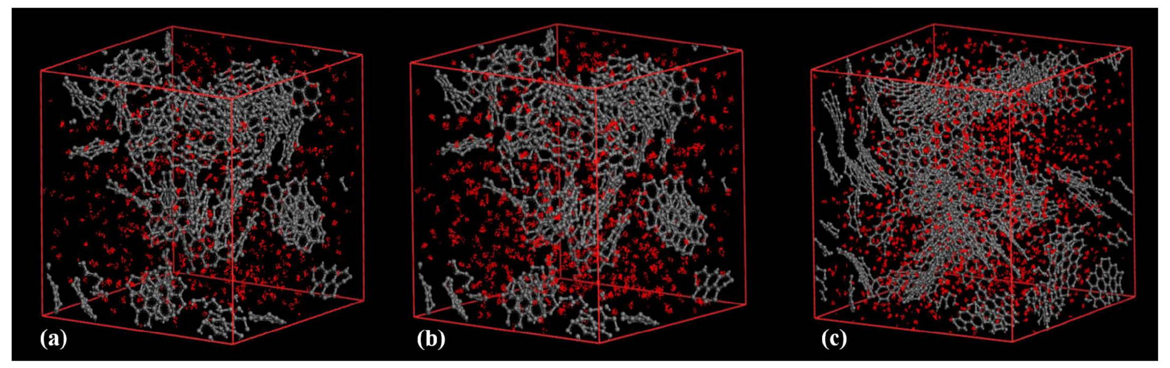


The blue part represents the free volume, the red part is the volume occupied by solid carbon.

(2) Adsorption simulation

The grand canonical Monte Carlo (GCMC):
In the grand canonical system (the volume, temperature and chemical potential are specified), a large number of movement, insertion and deletion operations of the particles are taken to balance the system. Hence, it's a powerful method for gas-solid physical adsorption process.

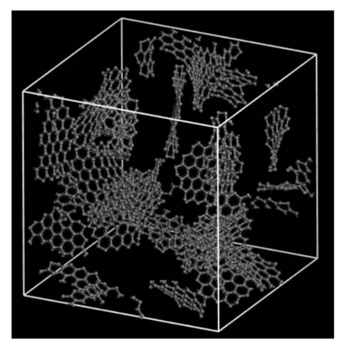
Material Studio 7.0's sorption model was used to simulate the adsorption isotherms and isosteric heats at 4 K, 6 K and 10 K.



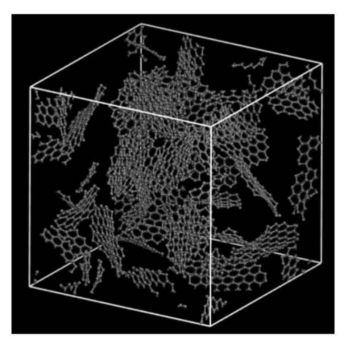
(a) 7 rings,6 K ,500 kPa (b) 7 rings,6 K ,2000 kPa (c) 19 rings,6 K,2000 kPa

(3) Mixed filling amorphous carbon

7 rings, 19 rings and 37 rings graphite slices were used to build amorphous carbon according to a certain amount of proportion (partial models).



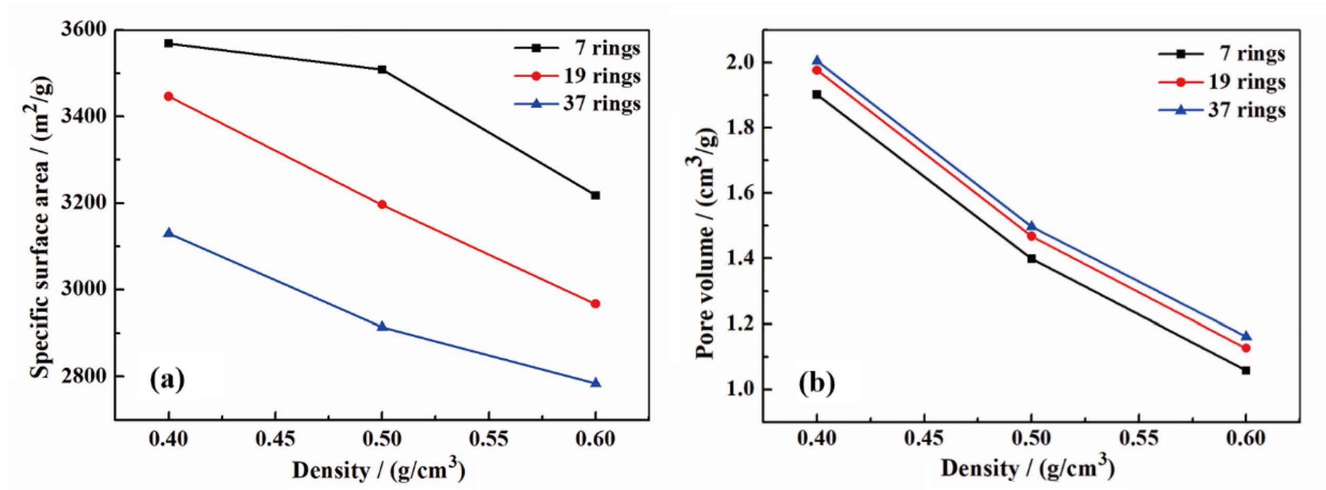
7 rings : 19 rings : 37 rings =1:1:1



7 rings : 19 rings : 37 rings =1:1:2

4.Result

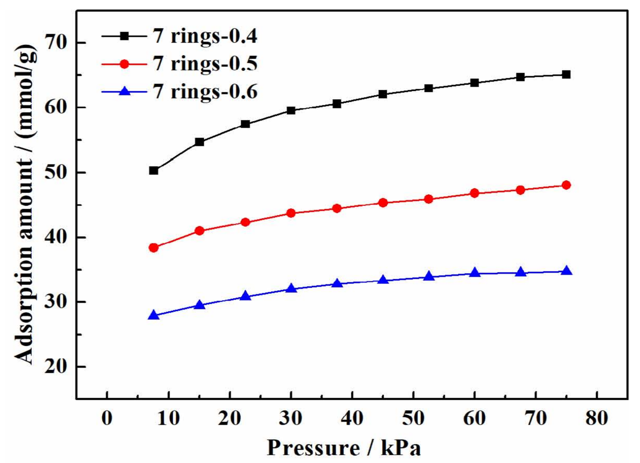
(1) Structural parameters of amorphous carbon (using single type of graphite slice)



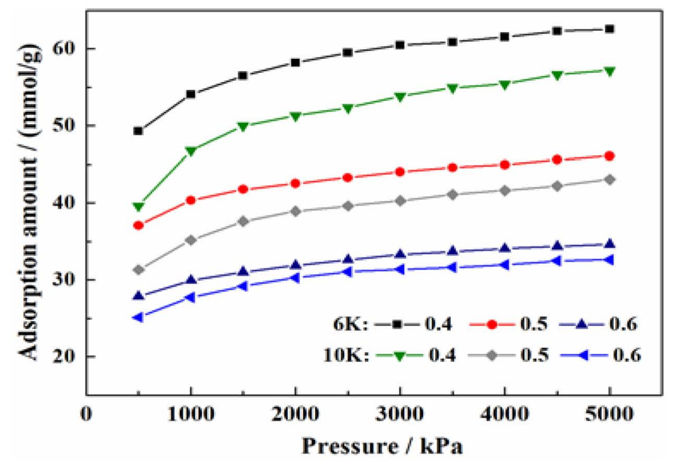
Structural parameters of amorphous carbon

- As the density increases, the specific surface area and pore volume decrease.
- When the density is constant, the smaller the size of the graphite slice, the larger the specific surface area and the smaller the pore volume.

(2) Adsorption simulation results

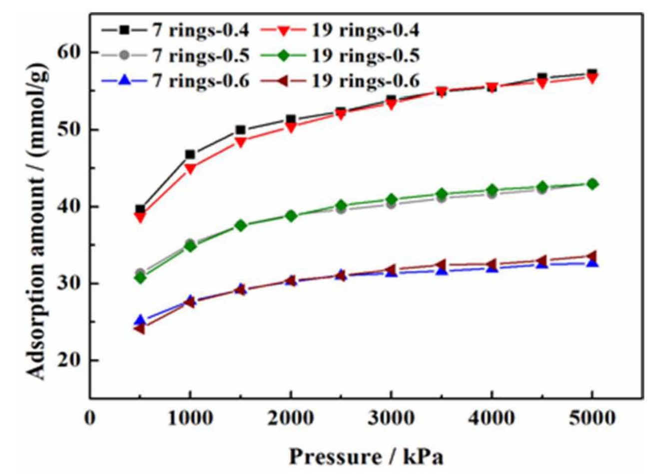


Adsorption amount of helium on amorphous carbon at 4 K



Adsorption amount of helium on amorphous carbon at 6 K and 10 K

When the size of graphite slice is unchanged, as the temperature and density decreases, the adsorption amount increases, and as the pressure increases, the adsorption amount increases and tends to a maximum value.

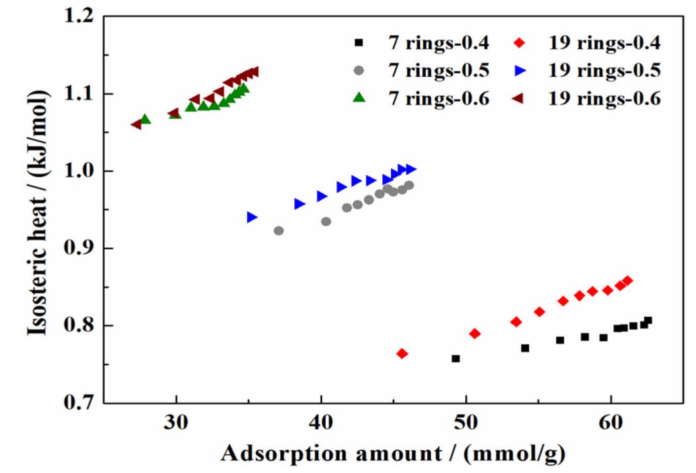


Adsorption amount of helium on different filled pattern amorphous carbon

The same density amorphous carbon filled with 7 rings or19 rings graphite slice have almost the same adsorption amount of helium in the same pressure and temperature.

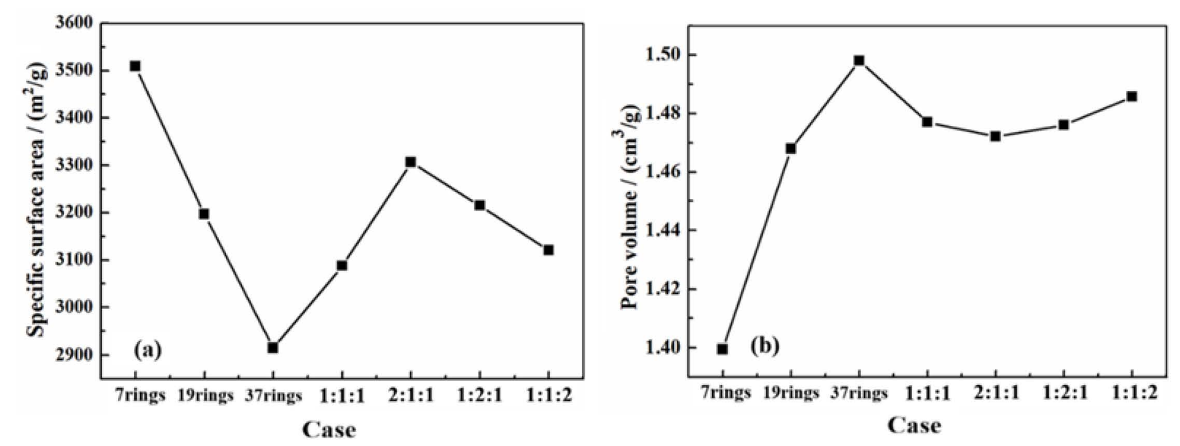
Specific surface area$_{(7rings)}$ > Specific surface area$_{(19rings)}$
Pore volume$_{(7rings)}$ < Pore volume$_{(19rings)}$

The isosteric heat increases as the adsorption amount, density and graphite slice's size increases.



The isosteric heat of different structures amorphous carbons at 6 K

(3) Effect of mixed filling method on amorphous carbon



a1: a2: a3 =7 rings : 19 rings : 37 rings (amount)

The amorphous carbon structure obtained by the mixed filling method can have both a higher specific surface area and a higher porosity.

5.Conclusion

(1) The specific surface area and pore volume of amorphous carbon increase as its density decreases.
(2) The amount of adsorption increases as the temperature decreases, and increases to a maximum as the pressure increases. At the same density, the amount of helium adsorbed by amorphous carbon constructed from 7 rings or 19 rings graphite slices is very similar.
(3) The larger the adsorption amount and the size of the filled graphite slice, the larger the isosteric heat.
(4) For an amorphous carbon structure composed of a single type of graphite slice, the smaller the size of the graphite slice, the larger the specific surface area and the smaller the pore volume.
(5) For an amorphous carbon structure composed of various graphite slices (mixed proportionally), its structural parameters are mainly affected by the graphite slice with the largest filling amount and it also has the structural characteristics of other size graphite slices. This mixed-filling method is advantageous for obtaining amorphous carbon with high specific surface area and pore volume.