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Growth Simulation of Carbon Nanotubes in liquid Helium using Molecular Dynamics

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Recently, it has been demonstrated that carbon nanomaterials can be synthesized also by arc discharge generated in a liquid medium such as water, liquid nitrogen or liquid helium. Shigematsu et al. have demonstrated that carbon nanomaterials can be produced by dc arc discharge in liquid helium. Then, we are working on the clarification of the creation mechanism by analyzing to experiment from both sides of the calculation. And, whether how much time the discharged carbon atom grew up to the size of which extent was examined. The motion of an individual carbon atom was pursued. Carbon velocity after the carbon atoms or the carbon clusters collide is calculated as a perfectly inelastic collision according to the law of conservation of momentum. The process of growing up from the carbon atoms to the carbon clusters that contained a lot of carbon atoms was shown. In addition, it was shown that the number of carbon atoms contained in the cluster for the logarithm at the time of peaks of the number of carbon clusters almost became a linear relationship. The result of this simulation suggests that the growth speed of the carbon clusters in liquid helium be predictable.
Key words: Molecular Dynamics, liquid Helium, Carbon Nanotubes

1. INTRODUCTION

Interest in nanomaterials has been rapidly growing for the past several years. In particular, carbon nanomaterials including fullerene molecules, carbon nanotubes, nanohorns and nanoonions are promising new materials for a variety of potential applications.
These carbon nanomaterials have been produced by various methods such as arc discharge in a buffer gas, laser ablation, chemical vapor deposition and annealing of nanodiamonds. Among them, the arc discharge method is the most common and is widely used by many researchers and manufacturers. Recently, it has been demonstrated that carbon nanomaterials can be synthesized also by arc discharge generated in a liquid medium such as water[1]-[5], liquid nitrogen[6]-[8] or liquid helium[8]. The liquid arc method, which does not required vacuum jacket, it more economical than the conventional gas arc method and may have some advantages in the mass production of carbon nanomaterials. Thus far, most types of liquid arc method have employed continuous arc discharge powered by a direct current power supply [1]-[4],[6][7]. In particular, arc discharge method in low temperature liquid is expected to discover the new type carbon nanomatelials and to elucidate formation mechanism of fullerene and nanotubes. Recently, Shigematsu et al.[7] have demonstrated that carbon nanomaterials can be produced by dc arc discharge in liquid helium. In low temperature liquid, it is possible for carbon nanomaterials to create slowly and for carbon cluster to show the creating process using by high speed microscopy. Then, we are working on the clarification of the creation mechanism by analyzing to experiment from both sides of the calculation. Numerical simulation using molecular dynamics forecasts how much time the discharged carbon atoms grow up to the size of which extent. In generation in liquid helium, it is said that uniting is started after the energy of the discharged carbon atom becomes equal to a surrounding environmental temperature. Therefore, this simulation was targeted situation that was after discharged carbon atoms were had become equal the temperature of helium. And it was assumed that time when the carbon had collided for the first time mutually was united. In this place, when a dc arc discharge experiment is done in liquid helium, creating speed of carbon cluster is simulated by Molecular Dynamics. And, whether how much time the discharged carbon atom grew up to the size of which extent was examined.

2. NUMERICAL METHOD

In this calculation, to examine the process that the carbon cluster generated, the motion of an individual carbon atom was pursued. The carbon atom begins moving according to the initial velocities of carbon atoms given from the uniform random number series so as to fit the Maxwell distribution at the given temperature of liquid Helium. When the carbon atom collides with other carbon atoms while moving, the carbon velocity after the collision is calculated according to the law of conservation of momentum. In this calculation, if the carbon atom or the carbon cluster collided, it was assumed that they were united. Therefore, the collision of the carbon atom or the carbon cluster is considered to be a perfectly inelastic collision, and the carbon velocity after the collision is calculated according to the law of conservation of momentum.
Velocity and mass of the carbon cluster after the

collision are calculated by the follows:

$$v_c = \frac{m_{c1}v_{c1} + m_{c2}v_{c2}}{m_{c1} + m_{c2}}$$

$$m_c = m_{c1} + m_{c2}$$

Where, v_{c1} and v_{c2} is a velocity vector of the carbon atom or the carbon cluster before collision, m_{c1} and m_{c2} is a mass of them before collision. And, v_c and m_c is a velocity vector and mass of the carbon cluster respectively after collision.

All the carbon atoms included in the uniting carbon clusters keep moving at the same velocity. The growth of the carbon was requested by pursuing the motion of the carbon that repeated such a collision and uniting. Though the interaction between atoms is given in a molecular dynamics method by the potential that consists of the function at the position of the atom usually[8], because the growth speed of the carbon cluster is targeted in this calculation, they are not considered.

3. CALCULATION CONDITIONS

In this study, we used three-dimensional computational domain which contain 500 carbon atoms. Computational domain and initial state of carbon atoms is shown in Fig.1. Initial state of carbon atoms were formed to state of noncontact using a random number series. The density of carbon in this system was decided from the experiment having been demonstrated by Shigematsu et al.[9]. And, the size of the computational domain was estimated to be 1.3 μ m from the density. This calculation was done in the condition of constant NVT of the faction of volume V, atomicity N, and T in the temperature. We used the periodic boundary conditions for all directions. The initial velocities of carbon atoms are given from the uniform random number series so as to fit the Maxwell distribution at the given temperature 4.2K that is temperature of liquid Helium. This calculation was advanced until 0.35ms by the time step 1ps.

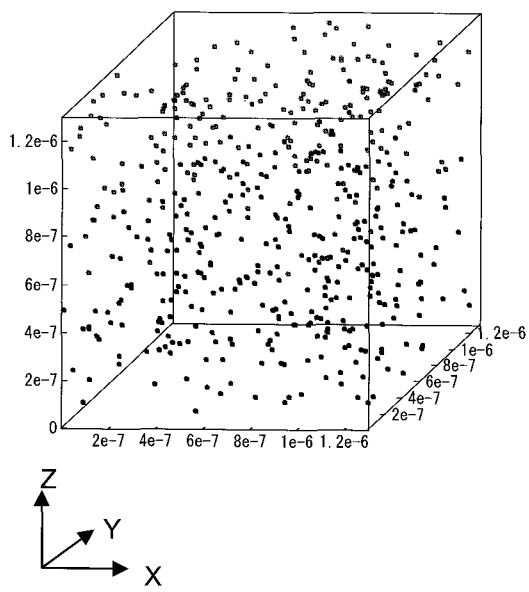

Fig.1 Computational domain (initial state)

4. RESULTS

4.1 Calculated instantaneous carbon configurations

Figure 2 and 3 show the calculated instantaneous carbon configurations at 0.1ms and 0.35ms, respectively. The carbon atom that exists alone in the initial state collides with the time passage with other carbon atoms, and forms the carbon cluster that consists of two carbon atoms. Afterwards, this carbon cluster collides repeatedly with other carbon atoms and other carbon clusters, and the number of carbons of clusters increases. These figures show that the formation of the carbon clusters advance with the time passage.

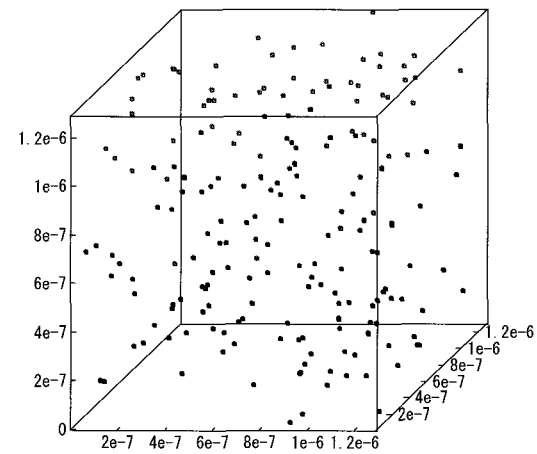

Fig.2 Calculated instantaneous carbon configurations (0.1ms)

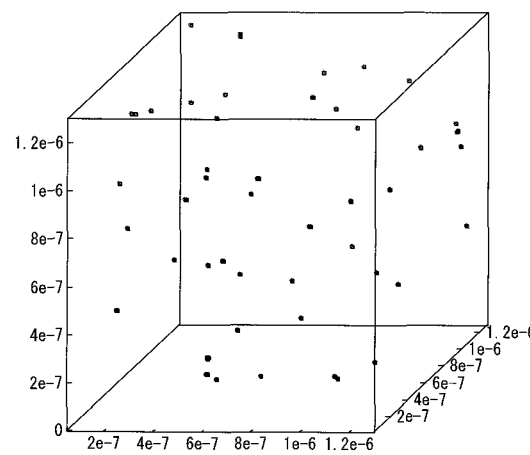

Fig.3 Calculated instantaneous carbon configurations (0.35ms)

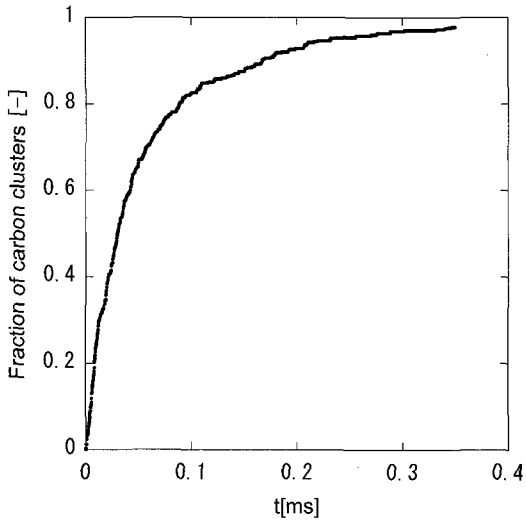

Fig.4 Fraction of carbon clusters

4.2 Fraction of carbon clusters

Figure 4 shows the fraction of carbon clusters with time. Because the carbon clusters is formed when the carbon atom collides with other carbon atoms, those carbon atoms are counted as a cohering carbon. The fraction of clusters is normalized by dividing the number of carbon atoms of all carbon clusters by the entire carbon atomicity. The fraction of carbon clusters increases rapidly at once after it begins to calculate though the fraction of carbon clusters in 0.0ms is 0 because all the carbon atoms exist alone in the initial state. A rapid increase at the fraction of carbon clusters continues to about 0.07ms, after passing 0.07ms, the ratio of an increase becomes small. In this condition of assuming it in liquid helium, the carbon atom exists alone collides at the time before 0.07ms, and the generation of the carbon clusters is promoted. Afterwards, though the number of carbon atoms that the carbon clusters contains increases when the collision is repeated, oppositely, the number of the carbon atom exists alone and carbon clusters decreases. Therefore, it is thought that the chance to collide decreases and it becomes this result.

4.3 Histogram of carbon clusters

Figure 5 shows a histogram made by the generated carbon clusters classified by the number of carbon atoms. A horizontal axis is the number of carbon atoms contained in the carbon cluster, and the axis in the vertical direction is the total amount of carbon clusters. The results of 0.05, 0.1, 0.2, 0.3 and 0.35ms are shown. A lot of carbon clusters that consist of the carbon atom of ten or less are generated in 0.05ms. And, it is shown that the number of carbon atoms increases with time, and the number of carbon clusters decreases oppositely. In especially 0.1-0.3ms, the number of carbon atoms of certain one carbon cluster increases rapidly. It is thought that this was caused by the collision of the carbon clusters that consists of the number of comparatively a

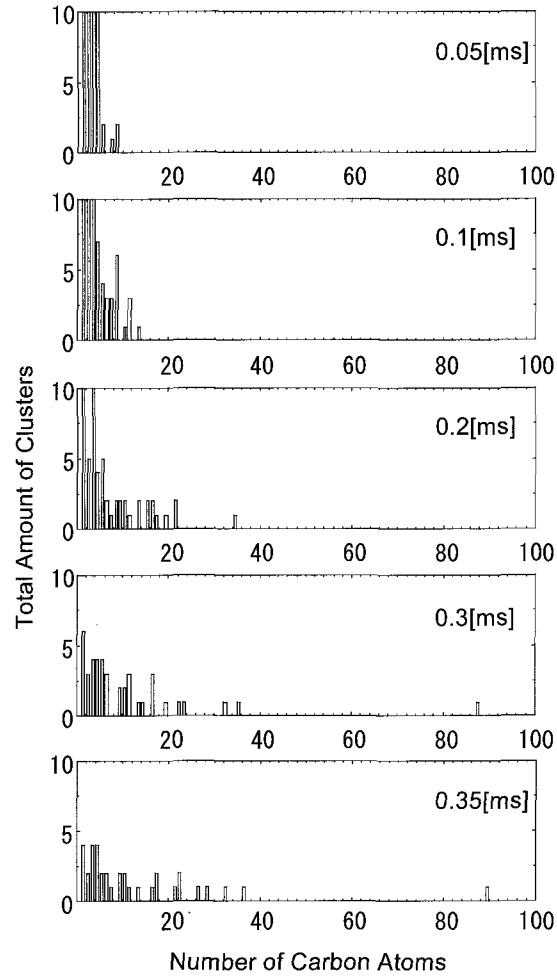

Fig.5 Histogram of carbon clusters

lot of carbon atoms.

4.4 Number of carbon clusters

Figure 6 shows the number of Carbon clusters that contains carbon atom from two to six with time. The axis in the vertical direction shows the number of carbon clusters. The number of carbon clusters that consist of two carbon atoms increases rapidly at once when the calculation is started. Afterwards, the number of carbon clusters that consist of three carbon atoms increases, and, in addition, the number of carbon clusters that consist of four carbon atoms increases afterwards. The number of carbon atoms of the carbon cluster increases by uniting it to other isolated carbon atom or carbon clusters. Therefore, the number of carbon clusters that contain a lot of carbon atoms as time shifting little by little increases. Oppositely, the numbers of carbon clusters that contain a little carbon atom decreases as the number of carbon clusters that contain a lot of carbon atoms increases. Therefore, the peak appears in graphs of the number of carbon clusters classified by the number of carbon atoms. It is thought that the reason with two peaks of the number of carbon clusters that consists of

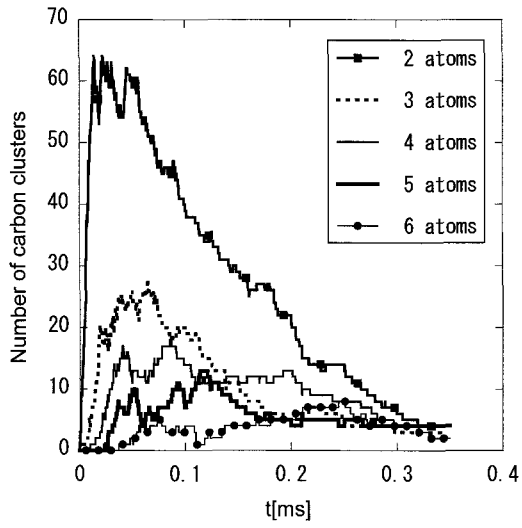

Fig.6 Number of carbon clusters

four carbons is that it is generated by uniting the carbon clusters that this consists of the carbon atoms of the number of (1 , 3) or (2 , 2). When the number of contained carbon atoms becomes five or more, it becomes difficult to distinguish time to show the peak because the number of peaks increases and the value of the peak becomes small. It is thought that the peak of the number of carbon clusters that consists of the carbon atoms of two or three number is not smooth shape, because of the union of these carbon clusters.

4.5 Growth speed of carbon clusters

The time of the peak in graphs of the number of carbon clusters that contained the carbon atom from 2 to 6 that had been shown in figure 6 was examined. Figure 7 shows the number of carbon atoms contained in the carbon cluster for that time. A horizontal axis is a common logarithm at time, and the axis in the vertical direction is a number of carbon atoms contained in the carbon cluster. In this graph, time that the number of each cluster increases most is indicated, and the relation between time and the growth of carbon clusters is shown. It can be confirmed that peaks of the number of content carbon atoms almost become the linear relationships for the common logarithm at time. It can be considered that this inclination of the straight line will show the growth speed of the carbon though the number of clusters of composition particles is the data of the early stage from two to six. As a result, the generation of the carbon clusters in this condition of assuming it in liquid helium can be estimated.

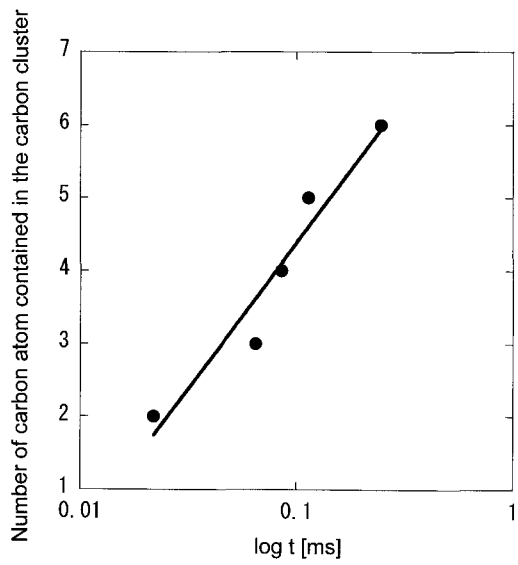

Fig.7 Growth speed of carbon clusters

5. CONCLUSIONS

In this study, the growth process of the carbon cluster in liquid helium was requested by the simulation using simple model Molecular Dynamics. And, whether how much time the discharged carbon atom grew up to the size of which extent was examined. The process of growing up from the carbon atoms to the carbon clusters that contained a lot of carbon atoms was shown. In addition, it was shown that the number of carbon atoms contained in the cluster for the logarithm at the time of peaks of the number of carbon clusters almost became a linear relationship. The result of this simulation suggests that the growth speed of the carbon clusters in liquid helium be predictable. In this research, the generation of the carbon clusters was simulated by using the simplified model. If the formation of the carbon clusters is analyzed more strictly, it is necessary to consider the interaction between atoms by the potential given by the function at the position of the atom.

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