Effects of Substrate Distance on Radiation Heat Transfer in Thermal CVD Reactor

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Received March 6, 2007; Accepted April 23, 2007

Abstract: The mass production of carbon nanotubes (CNTs) must be premised for actual life. Recently, many studies have been conducted to closely examine the thermal and flow fields inside of the reactor for mass production of CNTs. In this paper, the radiation heat transfer effect is numerically studied for the multiple substrates with respect to the substrate distance. The computational results show that when the substrate distance is close, the temperature and flow film is observed due to the radiation heat transfer effect of each substrate. As the substrate distance becomes larger the radiation heat transfer effect decrease. When the three substrates are ready to grow the CNTs, the substrate distance is required to be less than 40 mm. Also, the back side of the first substrate, the back and front sides of the second substrate and the front side of the third substrate can be used to grow the CNTs at the three substrates.

Keywords: thermal CVD reactor, multiple substrates, substrate distance, CFD, radiation heat transfer

Introduction

Carbon nanotubes (CNTs) have been studied extensively to discover the characteristics that are responsible for their excellent properties [1,2]. Over the past few years, a variety of applications for CNTs, such as in field emission display (FED), secondary batteries and fuel cells, and gas storage, have received extended studies [3-8].

Research into the physical characteristics and applications of CNTs began with the growth of CNTs; now, mass production of CNTs must be achieved for real-life applications. Above all, the most essential factor for the mass production of CNTs is an understanding of their growth mechanism and the ability to control their growth, length, diameter, and nucleation [9].

In order to give a clear picture of the growth mechanism of CNTs, the results found experimentally must be effectively coupled with the theory based on these experiments. Later, based on the growth mechanism, CNTs can be capable of a great quality of decomposition. Thus, it is necessary to closely examine the thermal and flow fields of the gas mixture inside of the reactor in order to establish a systematic theory. Numerical analysis is used as one of these methods.

The growth rate of CNTs depends on temperature, pressure, geometry of the reactor and the flow rate of gas mixture. The optimal growth conditions of CNTs depend on the type of substrate, growth time, quality of the catalyst and the type of gas used in the mixture.

Kuwana and coworkers performed computational fluid dynamics (CFD) calculations to simulate the heat and mass transfer processes that take place during the growth of CNTs in a chemical vapor deposition (CVD) reactor. They suggested a new geometry for the reactor that varied the gas injection port - side wall injection - to remove irregular flow field caused by the substrate located inside the reactor [10]. Lu and coworkers simulated the effects that the temperature, pressure, concentration of gas, and inlet velocity of gas mixture have on the composition and growth of particles in a tubular CVD reactor [11].
coworkers showed that the growth rate of SiC may either be carbon-transport-limited or silicon-controlled, depending on the input carbon-to-silicon ratio in a hot wall reactor [12]. Recently, Kim and coworkers showed that the radiation effects must be considered for a numerical analysis within a thermal CVD reactor analysis. They also simulated the flow and thermal fields inside of the reactor with respect to growth temperature and substrate angle [13].

In this paper, we numerically analyzed a thermal CVD reactor [14] as one of the CVD methods. The thermal CVD method operates at a relatively high temperature because the energy sole source is heat. A thermal CVD reactor has several benefits for the mass production of CNTs because it is very easy to control and easy to set up multiple substrates. The thermal and flow fields inside of a thermal CVD reactor are a different aspect for the multiple substrates compared with single substrate. Thus, it is necessary to clarify the relationship between the radiation heat transfer effect and the multiple substrate distances. In the present study, we focus on a thermal and flow fields around the multiple substrates inside a thermal CVD reactor. A series of numerical analyses have been performed using the simulation of turbulence in arbitrary regions computational dynamics (STAR-CD) [15].

**Governing Equations**

For a steady and incompressible flow fluid, the conservation of mass, momentum and energy are provided using three-dimensional cylindrical coordinates as follows:

**Continuity:**
\[ \frac{1}{r} \frac{\partial}{\partial r} (r \nu_r) + \frac{\partial}{\partial \theta} (r \nu_\theta) + \frac{\partial}{\partial z} (\nu_z) = 0 \]  
(1)

**r-Momentum:**
\[ (V \cdot \nabla) \nu_r - \frac{1}{r} \nu_\theta = -\frac{1}{\rho} \frac{\partial p}{\partial r} + g_r + \nu \left( \nabla^2 \nu_r - \frac{\nu_r}{r^2} - \frac{2}{r} \frac{\partial \nu_r}{\partial \theta} \right) \]  
(2)

**θ-Momentum:**
\[ (V \cdot \nabla) \nu_\theta + \frac{1}{r} \nu_r \nu_\theta = -\frac{1}{\rho r} \frac{\partial p}{\partial \theta} + g_\theta + \nu \left( \nabla^2 \nu_\theta - \frac{2 \nu_\theta}{r^2} - \frac{2}{r} \frac{\partial \nu_\theta}{\partial \theta} \right) \]  
(3)

**z-Momentum:**
\[ (V \cdot \nabla) \nu_z - \frac{1}{\rho} \frac{\partial p}{\partial z} + g_z + \nu \nabla^2 \nu_z \]  
(4)

**Energy:**
\[ \rho c_p (V \cdot \nabla) T = k \nabla^2 T + \nabla \cdot q' \]  
(5)

where \( r, \theta, \) and \( z \) are the axes of a cylindrical coordinate, \( p, \nu, g \) are pressure, kinetic viscosity and gravity acceleration assumed constant properties, respectively, and \( \rho \) is the density that was calculated by the function for temperature and pressure. The heat transfer rate of radiation \( \nabla \cdot q' \) is provided as follows:
\[ \nabla \cdot q' = \int_0^\infty \nabla \cdot q'_\lambda d\lambda \]  
(6)

where \( \lambda \) indicates wavelength.

**Numerical Procedures**

A schematic diagram of the thermal CVD reactor that we selected for the numerical analysis is shown in Figure 1 [16]. The reactor is made of quartz. Its diameter is 18 cm, the entire length is 90 cm, and the diameter of the inlet and outlet of gas mixture is 3/4 of an inch. The reactor is heated with a 20 cm long halogen lamp that surrounds the reactor.

The substrate, having a size of 20 (w) × 30 (l) mm is set at 45 to the horizontal with a constant pitch (d). The substrate is composed of three materials: Ni, TiN, and Si. A titanium nitride (TiN) thin film is deposited on the Si and a Ni thin film that acts as a catalyst is deposited on TiN/Si. The gas mixture is composed of argon (Ar) and acetylene (C\(_2\)H\(_2\)) that flows into the reactor at a ratio of 5 : 1. Ar is supplied at 1000 sccm (standard cubic centimeter per minute) and C\(_2\)H\(_2\) at 200 sccm. The total chamber pressure is 5.5 torr. Ar gas flows into the reactor until the temperature reaches the reaction temperature, then the C\(_2\)H\(_2\) gas passes into the reactor.

The governing equations are discretized using finite
volume method (FVM) and a monotone advection and reconstruction scheme (MARS) with second-order accuracy [15]. The pressure term coupled with velocity was achieved through the SIMPLE algorithm. Convergence was declared when the maximum normalized sum of the absolute residual sources was less than $10^{-6}$.

The discrete beam method (DBM) [17] is adopted as the radiation heat transfer model because the inside of the reactor is maintained at a very high temperature. The emissivity, reflectivity and transmissivity of quartz chamber are 0.4, 0.6, and 0.0, respectively. The values for the substrate on the Ni backside are 0.7, 0.3, and 0.0, and on the Si front side and bottom surface are 0.2, 0.8 and 0.0, respectively [18,19]. The view factor calculation for each surface is performed as follows [15].

$$\begin{align*}
F_{ij} &= \sum_{k=1}^{N_{ij}} \alpha_{ik} f_{ij} \\
\alpha_{ik} &= \begin{cases} 
1 & \text{if the beam strikes } j \text{ or zero otherwise.} 
\end{cases}
\end{align*}$$

where, $i$ and $j$ are each surface, and $f_{ij}$ is the view factor for a single beam. The coefficient $\alpha_{ik}$ is equal to 1 if the beam strikes $j$ or zero otherwise.

The grid structure for the thermal CVD reactor is shown in Figure 2. On the whole, the grid structure is coupled with two types - structured hexahedral and unstructured tetrahedral. That is, to simulate the flow and the thermal fields focused around the multiple substrates, hexahedral element is set in general, and then tetrahedral element is set denser than hexahedral grid around the substrates.

A velocity boundary condition is applied at the inlet based on the flow rate of gas mixture. The value of the velocity is determined from following equation:

$$\nu_{5.5\text{torr}} = \frac{p_{\text{standard}}}{p_{5.5\text{torr}}} \times \nu_{\text{standard}}$$

where $p_{5.5\text{torr}}$ and $p_{\text{standard}}$ are the pressures at 5.5 torr and standard, respectively.

Figure 2. Grid structure for the numerical analysis.

A pressure boundary condition is applied at the outlet in order to maintain the 5.5 torr inside of the reactor. Constant temperature condition given by experiments and thermal resistance was applied at the heated wall zone. For the rest of the wall, we applied an adiabatic boundary condition. Also, to take advantage of the computation, a symmetry boundary condition is applied at the center.

Results and Discussion

Figures 3 and 4 show the thermal and flow fields for a single substrate. The temperature and velocity around the substrate are very stable, and the stable zone is wider at the back of substrate than at the front of substrate. This stability of temperature and velocity at the back of the substrate is a primary factor for the growth of CNTs. Also, the stability of the whole area at the back of the substrate is directly concerned with the mass production of CNTs [13]. In this paper, we increase the number of substrates to three, and then compare the effect of radiation heat transfer to a single substrate.

Thermal and Flow Fields Analysis for Multiple Substrates

Figures 5 and 6 show the temperature and velocity magnitude distributions for the three substrates with the increase of the substrate distance. From Figure 5, the constant temperature film is observed around the multiple substrates when the substrate distance is relatively close. This is due to the fact that the radiation heat transfer affects each substrate directly and the gas flow does not infiltrate between each substrate’s back and front side as
shown in Figure 6. This constant temperature film disappears when the substrate distance is wider. Also, when the substrate distance is wider than 40 mm, the characteristics of the temperature and flow distributions at each substrate are similar to that of a single substrate.

Figure 7 shows the average temperature at the back and front of each substrate. From Figure 7(a), the average temperature decreases with the increase of the substrate distance. The decreasing rate of the average temperature at the front of substrate is larger than at the back of substrate. This is due to the fact that the first substrate is closer to the low gas flow region as the substrate distance is wide. When the substrate distance is at 10 mm, the average temperature of the back side agrees well when compared with single substrate. Figure 7(b) shows that the tendency of the average temperature for the second substrate is similar to the first substrate. When the substrate distance is close, the low temperature gas flow does not infiltrate to the back and front side of the second substrate. Thus, the average temperature is higher than, or equal to the single substrate for both sides. The average temperature decreases with the increase of the substrate distance because the infiltration rate of the low temperature gas flow increases and the radiation heat...
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Figure 7. Comparison of average temperatures on the backside of each substrate with respect to the substrate distance.

(a) Left substrate (the first substrate for streamwise)  (b) Middle substrate (the second substrate for streamwise)  (c) Right substrate (the third substrate for streamwise)

Figure 8. Comparison of average velocity magnitude on the backside of each substrate with respect to the substrate distance.

(a) Left substrate (the first substrate for streamwise)  (b) Middle substrate (the second substrate for streamwise)  (c) Right substrate (the third substrate for streamwise)

transfer effect for each substrate decreases. When substrate distance is wider than 40 mm, the average temperature of the back side is lower than when compared with a single substrate. From Figure 7(c), when the substrate distance is relatively close, the average temperature of the front side is higher than that of the back side, unlike the tendency of the first and second substrates. This result is attributed to the radiation heat transfer effect, that is, when the substrate distance is close the temperature of the front side rises because of the radiation heat transfer effect between the back side of the second substrate and the front side of the third substrate. This effect decreases with the increase of the substrate distance, then the average temperature of the front side decreases continuously. For the back side of third substrate, the average temperature increases with the increase of the substrate distance at the close distance region and reaches a maximum point at 40 mm, then decreases as the substrate distance further increases. This parabolic characteristic of the average temperature profile is due to the fact that the temperature of the back side slightly increases because of the decrease in the low temperature gas flow as the substrate distance increases. However, the average temperature suddenly drops because of the decrease of the radiation heat transfer effect from the halogen lamp when there is an increase of the substrate distance.

The velocity magnitude at the direction of the substrate length (l) is plotted in Figure 8 for the back side of each substrate. From Figure 8(a) and (b), we can notice that the low temperature gas infiltrates the back side of the substrate and increases with the increase of the substrate distance. This fact demonstrates the decrease of the average temperature with the increase in the substrate distance as discussed. For the second substrate, when the substrate distance is at 40 mm, the velocity magnitude is also agrees well when compared to the single substrate value. From Figure 8(c), the velocity magnitude decreases with the increase of the substrate distance up to 40 mm, and the velocity magnitude is almost the same over 40 mm. The velocity magnitude of the third substrate is lower than the single substrate for all substrate distances. This result is attributed to the fact that the location of the third substrate is behind the location of the single substrate for all cases.
Conclusion

The radiation heat transfer effect was studied numerically for the multiple substrates with respect to the substrate distance. The following results were obtained.

1) When the substrate distance is close, the temperature and flow films were observed due to the radiation heat transfer effect of each substrate.

2) As the substrate distance became wider, the radiation heat transfer effect of each substrate decreased, then the average temperature decreased and the velocity magnitude increased.

3) When the three substrates are set to grow the CNTs, the substrate distance did not exceed 40 mm.

4) The back side of the first substrate, the back and front sides of the second substrate and the front side of the third substrate could be used to grow the CNTs at the three substrate conditions.

Acknowledgment

This work was supported by the University of Seoul (2005).

References