Self-consistent mathematical model and simulation of carbon nanoparticle deposition from nonisothermal gas flow

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ABSTRACT

The new mathematical model of nanoparticle deposition and correlated nanostructured film growth has been developed. It is shown that the Brownian diffusion determines the final deposition rate but thermophoresis can drastically enhance it. Qualitative estimations and simulation results of nanostructured film growth are presented. For relatively small nanoparticles, some morphological parameters of the film are calculated.

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1. Introduction

The fundamentals of the Brownian diffusion of nanoparticles in a gas phase can be already found in many physics textbooks [1]. Technical applications of the Brownian diffusion have emerged only recently. In particular, considerable interest has focused on deposition of nanoparticles from gas flow on a reactor wall [2–6]. It is due to the importance of this process for wide variety of modern technologies concerned with coating and thin film production [7], nanoparticles transport [8], human health [9], membrane production [10], and others.

Therefore, insight into the physics of deposition process of nanoparticles, wherein the nanostructured film is formed, is really of great importance [10]. The formation and growth of a nanostructured film, resulting from nanoparticle deposition, depends on a variety of parameters. The temperature gradient in gas flow is one of the most important one. This temperature gradient affects the deposition due to the thermophoresis of nanoparticles.

The theoretical investigation of this complex process is based on the joint solution of the hydrodynamic problem for gas flow and the problem of heat transfer between the gas and the growing film, and subsequently, between the film and the reactor walls. Knowledge of the gas velocity profile and temperature profiles along the reactor is essential for solving the problem of convective

diffusion for nanoparticle distribution inside the reactor. After this we can determine the deposition rate of nanoparticles on the wall. It is obvious that a growing nanostructured film should change the conditions of the deposition process. This effect is especially important for a relatively thick deposited layer. Since the film grows relatively slowly, we consider the hydrodynamics and the heat transfer in the gas phase in quasi-steady state approximation. Also, it is worthy to note that experimental results depend on the shape of nanoparticles [11]. We consider only spherical nanoparticles in what follows. It is important to note that a vast majority of publications deal with relatively large nanoparticles with radii larger 100 nm [12]. Our consideration is limited to significantly smaller nanoparticles, less than 10 nm. Therefore we take into account the effects related with the Brownian diffusion. In particular, the Brownian diffusion of such nanoparticles determines the growth rate of a nanostructured film on the reactor wall.

For some industrial applications related with power engineering and chemical engineering special interest has dust deposition on tube surfaces from high speed compressible flows. There are many publications devoted to this problem. It is worthy to mention several reviews which consider this part of deposition problem [13–15].

The paper is structured the follows. First, we give a mathematical model of heat and mass processes that govern the deposition of nanoparticles. Then a self-consistent algorithm of the solution of our model is proposed and discussed. It is worthy to emphasize that we use the free molecular approximation for describing the

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Nomenclature

f	distribution function of nanoparticles in a reactor
n	number of all nanoparticles per unit volume in the reactor (m^{-3})
r	radial coordinate (m)
z	axial coordinate (m)
u	gas velocity (m s^{-1})
$\bar{u}$	average gas velocity (m s^{-1})
R_B	nanoparticle radius (m)
D_B	Brownian diffusion coefficient (m^2 s^{-1})
k	Boltzmann constant (J K^{-1})
T	temperature (K)
P	pressure (Pa)
m	mass of a gas molecule (kg)
b	mobility of nanoparticle (m/kg)
c_p	heat capacity at constant pressure of a gas (J kg^{-1} K^{-1})
h	thickness of a deposited film (m)
ε	porosity of a deposited film
j	deposition flow density (m^{-2} s^{-1})
r^*	half-width of an inlet nanoparticle beam (m)

l_t	thermal characteristic length (m)
l_B	characteristic length of the Brownian deposition (m)
p	relative number of nanoparticles in a deposited film
v	volumetric ratio of nanoparticles in a deposited film
s	relative surface area of nanoparticle group in deposited film
n^*	number density of nanoparticles on the reactor axes
K	efficiency of deposition of nanoparticles
η	dynamic viscosity of a gas (Pa s)
ρ	density of a gas (kg m^{-3})
v_{th}	thermophoresis velocity (m s^{-1})
λ	heat conductivity of a gas (W m^{-1} K^{-1})

Subscripts

in	value at the inlet of a reactor
w	reactor wall
c	deposited carbon film
i	*i*th group of particles

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interaction of nanoparticles with gas flow and at the same time we use approximation of continuous medium for describing of gas flow. In the next chapter we present a qualitative analysis of these equations and obtain qualitative analytical estimations of the nanostructured film. Then we present our simulation results. We summarize our results in the final section.

2. Mathematical model of the Brownian deposition of nanoparticles

The steady-state continuity equation for the distribution of the number density of nanoparticles f (r, z, R_B) via laminar flow in a cylindrical reactor is

$$\frac{\partial f u}{\partial z} + \frac{1}{r}\frac{\partial r v_{th} f}{\partial r} = \frac{1}{r}\frac{\partial}{\partial r}\left(r D_B \frac{\partial f}{\partial r}\right). \tag{1}$$

The number of all nanoparticles n per unit volume in the reactor is

$$n(r,z) = \int_0^\infty f(r,z,R_B)dR_B.$$

For a reactor with length L it is easy to show that the contribution of the longitudinal Brownian diffusion of nanoparticles could be neglected if the inequality $(R/L)^2 \ll 1$ is valid; this condition is true for most practical cases.

The second term on the left side of Eq. (1) takes into account the thermophoresis of nanoparticles in a radial direction. For the free molecular regime of the interaction of nanoparticles with gas flow the thermophoretic velocity v_{th} is determined by the expression [16]

$$\mathbf{v_{th}} = -\frac{3}{4}\frac{\eta}{\rho T}\nabla T = -\frac{3}{4}\frac{\eta k}{mP}\nabla T. \tag{2}$$

The Brownian diffusion coefficient is calculated by means of the Einstein relation [1]

$$D_B = kTb, \tag{3}$$

the mobility of a spherical nanoparticle b is calculated by means of the expression below

$$b = \frac{3}{16\pi R_B^2 P}\sqrt{\frac{2\pi kT}{m}}. \tag{4}$$

The field of gas temperature $T(r,z)$ along the reactor is governed by the advective heat diffusivity equation

$$\rho c_p u(r,z)\frac{\partial T(r,z)}{\partial z} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\lambda(T)\frac{\partial T(r,z)}{\partial r}\right). \tag{5}$$

We neglected a heat flow related to nanoparticles. Thus we cannot use Eq. (5) if the number density of nanoparticles is high.

For the gas velocity profile we use the Poiseuille profile:

$$u(r,z) = 2\bar{u}(z)\left(1 - \frac{r^2}{(R - h(z))^2}\right). \tag{6}$$

The averaged gas velocity $\bar{u}(z)$ is recalculated using the integral form of the continuity equation for mass gas flow rate in the reactor. It was shown earlier that such approximation for a velocity profile is good enough even in nonisothermal gas flow in a cylindrical reactor [17].

The equation for evolution of the film thickness is obtained by means of the mass conservation law for nanoparticles. In particular, for a monodisperse beam of nanoparticles in gas flow we have the following equation:

$$\frac{dh(z)}{dt} = -\frac{4}{3}\frac{\pi R_B^3}{(1-\varepsilon)}D_B\frac{\partial n(R-h,z)}{\partial r}. \tag{7}$$

It is important to emphasize that only the Brownian diffusion affects the nanostructured film growth because the size of nanoparticles is much smaller than the Knudsen layer. It is well known that the temperature gradient is absent in the Knudsen layer. We recall that thickness of the Knudsen layer is about two-three times larger than the mean free path of gas molecules. For atmospheric pressure the mean free path of air molecules is about 100 nm.

In a similar manner we obtain the flow of nanoparticles j on the reactor wall

$$j = D_B\frac{\partial n(R-h,z)}{\partial r}. \tag{8}$$

For a polydisperse beam of nanoparticles we have a more complicated equation for change of the film thickness:

$$\frac{dh(z)}{dt} = -\frac{4}{3}\frac{\pi}{(1-\varepsilon)}\int_0^\infty R_B^3 D_B\frac{\partial f(R-h,z)}{\partial r}dR_B. \tag{9}$$