Deposition efficiency of barium hexaferrite by aerosol deposition

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ABSTRACT

We present results of barium hexaferrite powder mass consumption across a wide range of starting powder quantities and deposition times. From these results we develop a transfer efficiency figure of merit to describe deposition efficiency and growth rates applicable to aerosol deposition and similar spray deposition techniques. We find that the transfer efficiency of barium hexaferrite was 0.082% and the transfer efficiency rate coefficient was 0.056 min^{-1} with a decay factor of -0.773. As a means to further understanding the deposition efficiency we present flow simulations of an aerosol deposition system using different particle sizes and standoff distances. We find that impact with the substrate strongly depends on the particle size, particle location within the nozzle, and standoff distance. We find that the values in the simulation are consistent with those used to produce films with the aerosol deposition system used at the Naval Research Laboratory and consistent with values found in the literature. We find that to improve the transfer efficiency nozzle design must be optimized, particle size must be carefully selected, standoff distance must be selected, and the powder in the aerosol chamber must be delivered at an optimal rate. These factors may be individually tuned to contribute to the final transfer efficiency figure of merit that can be used to assess the efficiency of the aerosol deposition process.

1. Introduction

Aerosol deposition (AD) is a technique that rapidly produces dense polycrystalline films that are several microns thick. One hallmark of AD is that the entire process occurs at room temperature, thus enabling integration of high-melting-temperature materials with low-melting-temperature materials. The process was developed in the late 1990s by Jun Akedo et al. [1] building on earlier work in the 1980s [2,3]. The application arena for AD films are numerous; some include, piezoelectrics [4], biomedical implants [5], abrasion coatings [6], electromagnetic interference shielding [7,8], rf circuitry [9], and corrosion resistance [10]. Additional background can be found in reviews of AD [11,12]. As AD develops from the laboratory scale to the industrial scale a detailed understanding of the system properties and mechanisms of the growth process become increasingly important for maximizing deposition efficiency and in tuning the parameters for each material growth. This motivates the need to develop a fundamental understanding of the process parameters and to develop a figure of merit (FoM) for comparison within the field of AD and between other spray deposition techniques.

There have been some efforts on simulating and measuring the gas and particle flow from nozzles in AD and cold spray. Gas and particle velocities for cold spray were estimated using an isentropic gas flow model that provides a framework for analytical calculations of gas velocities [13,14]. Reports specific to AD include an experimental apparatus to measure the impact velocity of 30–70 nm silver particles and Pb-Zr-Ti-O (PZT) particles. The velocities were found to be 650 m/s and 200 m/s, respectively indicating that ceramic particles require much less velocity to deposit than metallic particles [15]. Estimates of the impact velocity based on the carrier gas were also reported to vary between 150 and 500 m/s for α-Al$_2$O$_3$ and PZT particles [12]. More recent work using numerical simulation of Al$_2$O$_3$ particles between 0.5 and 15 μm in size suggests that particle velocity can be tuned both by the particle size and pressure in the deposition chamber [16]. Simulations of gas flow using different nozzle sizes, chamber pressures, and substrate standoff distances also suggest the importance of tuning these parameters to optimize the gas flow [17]. Later, these parameters were correlated with experimental results of deposition of 3 μm diameter TiO$_2$ particles with a standoff distance of 15 mm [18]. More recently, computational fluid dynamic CFD simulations of gas flow with 0.5 μm diameter Al$_2$O$_3$ particles were performed for a range of standoff distances and pressures. They found that the average impact velocity of the particles were between 335 and 346 m/s for standoff distances between 1 and 7 mm, respectively. They compared experimental results

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Nomenclature

A	Coefficient of transfer efficiency
AC	Aerosol chamber
AD	Aerosol deposition
BaM	Barium hexaferrite
C_s	Stokes drag coefficient
CFD	Computational fluid dynamic (simulation)
D	Diameter
DC	Deposition chamber
E	Total specific energy (internal plus kinetic)
FCT	Flux-corrected transport
FoM	Figure of merit
GSV	Granule spray in vacuum
$\mathbf{I}$	Identity matrix
KIMS	Korean Institute of Materials Science
NRL	Naval Research Laboratory
P	Pressure
PZT	Pb-Zr-Ti-O compounds
Re	Reynolds number
T	Temperature
T	Thickness
TE	Mass transfer efficiency
f	High velocity correction term
l	Length
m	Mass
rf	Radio frequency
t	Time
v	Gas-phase velocity
w	Width
x	Longitudinal position
y	Axial position
β	Exponent of the time dependence of transfer efficiency
γ	Ratio of the specific heats
λ	Thermal conductivity
μ	Gas-phase viscosity
ρ	Density
τ	Viscous stress tensor

Subscripts and superscripts

b	Boundary
f	Final condition
$film$	Properties pertaining to the film
i	Initial condition
j	Particle index in simulation
p	Particle
rel	Relative
$stock$	Spray feedstock
sub	Substrate
$sweep$	Nozzle sweep
sym	Symmetric part of tensor
$'$	Effective value

that found that the film thickness decreased with increasing standoff distance [19]. These efforts suggest that particle type, particle size, chamber pressure, nozzle design, and standoff distance all play an important role in understanding and controlling the deposition process.

There has been very little reported on the effect of powder mass flow through the nozzle. Studies of milling and heat treatment of the powders have been done [20,21], however no fundamental study of the mass flow in AD systems has been found in the literature. In this work, we report experimental results that relate powder mass consumption and film thickness and we use these results to develop a mass transfer efficiency FoM. We include results of simulations of particle flow near the substrate that suggest particle impact is strongly governed by standoff distance and particle size. We synthesize these results to suggest that the mass transfer rate is also an important parameter in film formation of AD systems.

2. Materials & methods

2.1. Numerical procedure

The numerical simulations focus on the particle paths as they pass through the nozzle and impinge on the substrate in the deposition chamber. Because of that, we only model the deposition chamber, as well as the channel between the aerosol chamber (AC) and deposition chamber (DC).

For these simulations, the volume fraction is assumed to be low enough that we have only one-way coupling of the gas-phase to the dispersed-phase. The gas-phase follows the viscous Navier-Stokes equations in conservative form for a compressible gas:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \rho v = 0$$

$$\frac{\partial \rho v}{\partial t} + \nabla \cdot \rho v \otimes v = -\nabla P + \nabla \cdot \tau$$

$$\frac{\partial \rho E}{\partial t} + \nabla \cdot (\rho E + P) v = \nabla \cdot \lambda \nabla T$$

The viscous stress tensor, τ, is defined as

$$\tau = \mu (\nabla v)_{\text{sym}} - \frac{2}{3} \mu (\nabla \cdot v) \mathbf{I}$$

The Sutherland expression is used for expressing viscosity as a function of temperature,

$$\mu(T) = 1.458 \times 10^{-5} \left(\frac{T^{3/2}}{110 + T} \right).$$

We assume the Prandtl number is 1 to calculate the thermal conductivity of the gas. To close these equations, we assume a calorically perfect gas and the ideal gas relation:

$$\rho E = \frac{P}{\gamma - 1} + \frac{1}{2} \rho v^2.$$

Single particles are tracked using a Lagrangian particle-tracking procedure. The particle is characterized by its mass $m_{p,j}$, diameter $D_{p,j}$, location $x_{p,j}$ and velocity $v_{p,j}$. The single-particle equation of motion is

$$m_{p,j} \frac{dv_{p,j}}{dt} = 3\pi D_{p,j} \mu (v - v_{p,j}) f(\text{Re}_{p,j})$$

where v is the gas-phase velocity, μ is the gas-phase viscosity, and $\text{Re}_{p,j}$ is the Reynolds number based on the slip velocity, $v_{\text{rel},j} = (v - v_{p,j})$. The high velocity correction term, $f(\text{Re}_{p,j})$ is the ratio of the actual coefficient of drag to the Stokes drag coefficient, $C_{s,j} = 24/\text{Re}_{p,j}$ and has been the subject of extensive research. The simplest high velocity correction term is the Schiller and Naumann law [22], expressed as

$$f(\text{Re}_{p,j}) = (1 + 0.15 \quad \text{Re}_{p,j}^{0.687})$$

This correlation works well for Reynolds numbers under 800. For particles on the order of one micron, the Reynolds number will always be well below 800.

For the gas-phase, the flux-corrected transport (FCT) algorithm of Boris and Book [23] is used, which is especially suited for high-speed