International Journal of Heat and Mass Transfer 73 (2014) 595–599

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

The effects of density on thermal conductivity and absorption coefficient for consolidated aluminum nanoparticles

Shawn C. Stacy [a], Xin Zhang [b], Michelle Pantoya [a,*], Brandon Weeks [b]

[a] Department of Mechanical Engineering, Texas Tech University, Lubbock, TX 79409-1021, United States
[b] Department of Chemical Engineering, Texas Tech University, Lubbock, TX 79409-3121, United States

ARTICLE INFO

Article history:
Received 20 November 2013
Received in revised form 17 February 2014
Accepted 17 February 2014
Available online 15 March 2014

Keywords:
Radiation absorption coefficient
Thermal conductivity
Aluminum powder
Aluminum compacts
Nanoparticles

ABSTRACT

Thermal conductivity and absorption coefficients were determined for a range of bulk densities for consolidated aluminum (Al) nanoparticle pellets by a laser flash analysis technique. Samples where heated on their front face using a Nd:YAG laser (1064 nm wavelength) and the temperature was measured on the rear face as a function of time. COMSOL Multiphysics® was used to model the experimental setup to determine the thermal conductivity and absorption coefficients. Thermal conductivity was found to increase with density from 0.2 to 1 W/(mK) for densities from 1.1 to 2.3 g/cm^3 due to increase surface contact between particles. The absorption coefficient was found to range from 0.30 to 0.54 and increase with density for the range of bulk densities investigated. These results provide a fundamental understanding of properties influencing ignition mechanisms in aluminum containing materials.

1. Introduction

Aluminum (Al) particles are a key reactant in many composite energetic material formulations. Powders of fuel and oxidizers are combined and often consolidated by compressing the mixture to various bulk densities. Determining thermal transport properties and absorption coefficients for individual reactants is the first step toward modeling their laser ignition behavior by defining fundamental a heat flux boundary condition. Thermal transport properties, such as thermal conductivity and thermal diffusivity, are important for determining ignition delay. Also, the absorption coefficient quantifies how much of the incident laser energy is absorbed by the surface. For composites consisting of Al nanoparticles, thermal transport properties generally increase with density [1] and absorption coefficients for a packed bed of Al particles have been difficult to obtain [2].

Thermal diffusivity is commonly measured by laser flash analysis (LFA) techniques. In the early 1960s, Parker et al. developed a method to determine thermal diffusivity, α, from the time it takes for the rear face of a sample to reach 50% of the maximum temperature increase, $t_{50\%}$, from an energy pulse on the front face using Eq. (1) [3]. The sample thickness is defined by H. This method is accurate for small temperature increases (i.e., less than 500 °C) based on the assumption of minimal radiation and convection loses.

$$\alpha = \frac{1.38H^2}{\pi^2 t_{50\%}} \quad (1)$$

Thermal conductivity can then be determined by applying Eq. (2) and using a mass weighted average specific heat capacity, C_p and a bulk density, ρ.

$$k = \alpha C_p \rho \quad (2)$$

Composite energetic materials such as Al nanoparticles combined with molybdenum trioxide (MoO_3) at various densities have been studied with LFA and show thermal conductivity and thermal diffusivity increase with density [1]. Thermal conductivity was found to gradually increase from 0.2 to 1 W/mK for a density change of 1.07 to 2.5 g/cm^3 [1].

In addition to the thermal transport properties, determining the absorption coefficient for reactive composites is critical for understanding laser ignition by accurately predicting how much energy is absorbed by the surface [4]. For an evaporated film of Al, less than 10% of the incident 1064 nm wavelength light will be absorbed [5]. For very small particles, there will be scattering that is dependent on wavelength and particle size. The Mie solution to Maxwell equations can be used to described the scattering of light by a sphere [6]. Using the Mie scattering code, MiePlot, and the optical constants [7] for Al nanoparticles, 81 nm in diameter, show that more than 27.5% of the light is absorbed by the particle [8]. The Al optical constants, for a wavelength of 1064 nm are: index of refraction, $n = 1.38$ and extinction coefficient, $k = 1.02$ [7].

* Corresponding author. Tel.: +1 806 742 3563; fax: +1 806 742 3450.
E-mail address: michelle.pantoya@ttu.edu (M. Pantoya).

http://dx.doi.org/10.1016/j.ijheatmasstransfer.2014.02.050
0017-9310/

Nomenclature

a	absorption coefficient	Ra_L	Rayleigh number
C_p	heat capacity, J/gK	T	temperature, K
$f(t)$	smoothed square pulse	$t_{50\%}$	time to reach 50% of the maximum temperature, S
H	sample thickness, mm	T_{amb}	ambient air temperature, K
$\bar{h}$	mean heat transfer coefficient, W/(m^{2}K)	T_s	surface temperature, K
k	thermal conductivity, W/mK	ρ	bulk density, g/cm^3
L	length scale for free convection, m	%TMD	percent theoretical maximum density, %
Pr	Prandtl number	α	thermal diffusivity, mm^2/s
Q	constant laser energy, J		
$q(t)$	time dependent laser power, W		

The amorphous alumina shell on Al particles [9] is transparent to 1064 nm wavelength light [10]. This can easily be observed for visible light due the fact that alumina, Al_2O_3, nanoparticles are white while most other nanoparticles are grey or black due to scattering and absorption. The Al_2O_3 shell will still have a slight effect on absorption though. For 632.8 nm wavelength light, the oxide coating on a single 34 nm diameter Al nanoparticle will slightly increase absorption from 99.06% to 99.075% [2]. It should be noted that this is for a single particle and there will be diffraction and interference effects between consolidated particles. It is difficult to accurately model the laser interactions of randomly packed particles; therefore, experimental analysis provides insightful observations of this behavior [2].

An experimental method to measure laser light reflection and transmission is to have an incident laser beam aligned perpendicular to a flat, optically thin sample and measure the scattered light at various angles around the sample. Begley and Brewster determined the amount of light (632.8 nm wavelength, HeNe Laser) scattered from confined and consolidated 34 nm diameter, Al nanoparticles to be 35–38% [2]. They found Al difficult to work with and only obtained results from two successful experiments due to possible cracking of the sample [2]. The effects of density were not reported with their setup.

Another method for determining absorption is by using an integrating sphere. Integrating sphere experiments consist of a hollow sphere with a diffuse, reflective coating inside, small ports for a light source and light sampling, and the sample suspended in the center. A portion of the incident light on the sample will be equally reflected to all points on the sphere. Absorption coefficients can be obtained by taking the ratio of reflected to incident light and accounting for the ratio of the area of the measuring port and the surface area inside the sphere. Integrating sphere experiments provide a scalar quantity for absorption, but lose the spatial information determined by the reflection and transmission experiments used by Begley and Brewster [2]. Also, integrating sphere experiments have limitations with sensitive energetic composites. If accidentally ignited, they will damage the diffusion coating.

Tolochko et al. conducted an integrating sphere study to determine the absorption coefficients for a large number of materials used for laser sintering [11]. Results showed that for metal powders, Nd-YAG lasers (1.064 μm wavelength) absorb 1.5–2.5 times more energy than CO_2 lasers (10.6 μm) [11]. Metal oxides and polymers were found to absorb very little energy from the Nd:YAG laser, but almost all of the energy from the CO_2 laser. This study also showed that due to scattering between particles, loose powders absorb significantly more energy than packed powders [11]. These results indicate that studying Al alone is important for composite energetic materials because most of the energy will be absorbed by the Al fuel particles and not the oxidizers.

Absorption coefficient can also be determined by modeling the heat transfer of laser heating through a pellet and curve fitting an absorption coefficient to correspond with a temperature measurement on the opposite of the heating side of the specimen. This approach has been applied before and boundary conditions should be validated by matching heating and cooling curves [12]. Monagheddu et al. used this modeling and iteration method to show that 12–19% of the incident energy CO_2 laser is absorbed by micron Al+Ni [12]. Carbon/graphite sprays or paint have been used to coat samples to increase absorption and maintain a constant absorption coefficient between samples [13,14]. The absorption coefficient determined by this modeling and curve fitting method for coatings is greater than fully consolidated Al+MoO_3 [13], but at higher heating rates the coating will act as a thermal insulator slowing ignition delays on the order of magnitude of the laser pulse [14].

Laser ignition models use estimates for the absorption coefficient that are available in the literature [15] or by curve fitting thermocouple results to a model [12,13]. Laser ignition studies of Al+Ni [15] and Al+MoO_3 [16] pellets using a CO_2 laser (10.6 μm wavelength) cite integrating sphere experiments for Al+nitrocellulose with a wavelength of 1.053 μm [17] for an estimation of absorption coefficient. While these approximations are close [12], different wavelengths will change the optical properties [7] and scattering [6].

All of these studies highlight the importance of experiments that elucidate the properties of aluminum nanoparticles and contribute toward more accurate models. The objective of this study is to examine thermal transport properties and absorption coefficients for Al powder compacts as a function of bulk density. This objective was accomplished through experiments coupled with modeling.

2. Materials

Aluminum powder was obtained from NovaCentrix (formally Nanotechnologies) with a mean particle diameter of 81 nm, 2.1 nm thick Al_2O_3 shell and 80% active aluminum content. The theoretical maximum density (TMD), calculated as the volume average density of Al and amorphous Al_2O_3, was found to be 2.88 g/cm^3 for the aluminum powder. The Al powder was sieved through a 325 mesh to reduce the size of clusters. Sieved powder was then hand-pressed into cylindrical pellets with a 4.7 mm diameter and approximately 2 mm thickness. The pellet densities ranged between 1.1 and 2.3 g/cc.

3. Methods

Pressed pellets were heated on the top surface by a Nd:YAG laser (1064 nm wavelength, 10 ms pulse length, and 1 J pulse power) and temperature as a function of time was measured on the bottom surface using a type E thermocouple, 75 μm junction diameter. Thermal paste was used to ensure contact between the