

Available online at www.sciencedirect.com



International Journal of Heat and Mass Transfer 47 (2004) 3643-3648

www.elsevier.com/locate/ijhmt

International Journal of

HEAT and MASS TRANSFER

Measurement of light extinction constant of JP-8 soot in the visible and near-infrared spectrum

Jinyu Zhu^{a,*}, Andrea Irrera^{b,1}, Mun Young Choi^b, George W. Mulholland^c, Jill Suo-Anttila^d, Louis A. Gritzo^d

^a Transportation Technology R&D Center, Argonne National Laboratory, 9700 S. Cass Ave, Argonne, IL 60439, USA

^b Mechanical Engineering and Mechanics, Drexel University, Philadelphia, PA 19104, USA

^c Building and Fire Research Laboratory, NIST, Gaithersburg, MD 20899, USA

^d Fire Science and Technology, Sandia National Laboratories, Albuquerque, NM 87185, USA

Received 12 January 2003; received in revised form 14 April 2004

Abstract

The dimensionless extinction constant, K_e , was measured using the NIST large agglomerate optics facility (LAOF) for soot produced from JP-8 flames. Measurements were performed using light sources ranging from 632.8 to 1565 nm. These experiments represent the first measurement of dimensionless extinction constant for soot produced from JP-8 flames. The K_e values did not display significant spectral variations that were observed for pure fuels such as acetylene and ethene. The measured K_e values ranged from 9.75 to 9.95 in the wavelength range between 632.8 and 1565 nm. Measurements provide a more reliable value of K_e for use in optical-based soot diagnostics for soot concentration and temperature measurement as well as for soot radiation analysis.

© 2004 Elsevier Ltd. All rights reserved.

Keywords: Soot; Extinction constant; Radiative properties; Light extinction; Two-wavelength pyrometry

1. Introduction

Since 1996, JP-8 has become the primary fuel used for military aviation and battlefield operations. There are many benefits of using JP-8 over its predecessor, JP-4, including its higher flashover temperature (leading to reduced hazards involved in its storage and transport) and lower levels of benzene and other carcinogenic aromatic content (leading to reduced health risks associated with exposure and inhalation). It is estimated that more than 4.5 billion gallons of JP-8 are used each year by the United States military. Due to increased importance as a primary aviation fuel, the hazards associated with cata-

*Corresponding author. Tel.: +1-630-252-9920; fax: +1-630-252-3443.

E-mail address: jzhu@anl.gov (J. Zhu).

¹ Present Address: Saipen (ENI), Italy.

strophic events leading to JP-8 fueled fires must be evaluated. Under such situations, the thermal hazard to the critical armament cargo and/or occupants posed by the fire is dominated by radiative emission from the hot soot particles [1,2]. Thus, accurate measurements of the soot concentration and temperature to be used in the radiative heat transfer formulation are critical in gaining a better understanding of the burning behavior of large JP-8 fires.

Soot concentration and temperature measurements in flames and fires are typically measured using nonintrusive, light extinction and or emission techniques. For light extinction measurements, the dimensionless soot extinction constant, K_e , is used to determine soot concentrations through Bouguer's Law:

$$\frac{I}{I_0} = \exp\left(-K_{\rm e}\frac{f_{\rm v}}{\lambda}L\right) \tag{1}$$

Bouguer's Law relates the ratio of the transmitted (I) and incident (I_0) light intensities to the concentration of

0017-9310/\$ - see front matter @ 2004 Elsevier Ltd. All rights reserved. doi:10.1016/j.ijheatmasstransfer.2004.04.013

Nomenclature				
$C_2 \ d_{ m p} \ f_{ m v} \ f_{ m vg}$	Planck's second constant, 1.4388 cm*K diameter of the soot primary particle soot volume fraction soot volume fraction measured using gravi- metric technique	m $m_{\rm s}$ $M_{\rm s}$ n_{λ} $x_{\rm p}$	soot refractive index, $m_{\lambda} = n_{\lambda} - ik_{\lambda}$ mass of filter collected soot mass concentration of soot real part of the refractive index of soot optical size parameter	
$I \\ I_0 \\ K_e \\ k_\lambda$ L	transmitted laser intensity incident laser intensity dimensionless extinction constant imaginary part of the refractive index of soot pathlength of light extinction	Greek λ Θ_{s} ρ_{s}	symbols wavelength of light mass specific extinction coefficient density of soot	

soot (f_v , volume fraction), and the light pathlength through the soot, *L*. A common method for soot temperature measurement is the two-wavelength pyrometry technique [3,4] in which emission from the hot soot is simultaneously monitored at two different wavelengths, λ_1 and λ_2 (wavelengths are usually centered in the nearinfrared spectrum). The measured ratio of the spectral emission intensity is used to calculate the temperature with the following relationship [5]:

$$\frac{I_{\lambda_1}}{I_{\lambda_2}} = \left(\frac{E(m_1)}{E(m_2)}\right) \left(\frac{\lambda_2}{\lambda_1}\right)^6 \left(\frac{\exp(C_2/\lambda_2 T) - 1}{\exp(C_2/\lambda_1 T) - 1}\right)$$
(2)

where E(m) is a function of the refractive index of soot, *m*, and C_2 is the Planck's second constant.

Determination of soot concentration from light extinction measurements and temperature using two wavelength pyrometry require accurate and wavelengthdependent values of K_e . Often the values of K_e are calculated using the Rayleigh-limit solution of the Mie analysis (see Eq. (3)) with complex refractive index, *m*, obtained from the literature [6–9]:

$$K_{\rm e} = \frac{36\pi n_{\lambda}k_{\lambda}}{\left(n_{\lambda}^2 - k_{\lambda}^2 + 2\right)^2 + 4n_{\lambda}^2k_{\lambda}^2}$$
(3)

where n_{λ} and k_{λ} are the real and imaginary parts of the complex refractive index. There are significant uncertainties in this method of determining K_e values [10]. In 1995, Choi et al. [11] performed accurate K_e measurements using a simultaneous gravimetric sampling and light extinction technique (GSLE) for soot produced from premixed acetylene/air flames. They reported values that were nearly a factor of two larger than the K_e calculated using Eq. (3) (with m = 1.57-0.56i [6,9]). Subsequent experiments were performed using visible and near-infrared light sources for soot produced from laminar and turbulent diffusion flames of acetylene and ethene [12,13]. These measurements revealed significant fuel and wavelength dependent behaviors which tend to discount the utility of a universal value of K_e (one that applies for soot produced from all fuels and wavelengths ranging from visible to the near-infrared spectrum).

In this study, we report accurate measurements of K_{e} in the visible and infrared spectrum for soot produced from JP-8 flames. The results can be used for more reliable soot concentration and temperature measurements and radiative heat transfer analysis. There are several important reasons for obtaining soot optical properties of JP-8 in the near-infrared spectrum. First, recent experiments indicate that there is significant variation in the measured K_e values due to differences in fuel type. Thus, accurate interpretation of soot concentration measurements in JP-8 fires will require direct measurement of optical properties of JP-8 soot. Second, there is likely to be lower levels of attenuation, which can extend the pathlength/concentration range of the light extinction measurements which is especially important for heavily sooting situations encountered in JP-8 fires. Third, soot optical properties in the near-infrared will be required for two wavelength pyrometry applications to measure soot temperature in JP-8 fires. These experiments represent the first reported measurement of $K_{\rm e}$ for JP-8 soot in the visible and near-infrared spectrum.

2. Experimental descriptions

Experiments were performed using the NIST large agglomerate optics facility (LAOF). Fig. 1 displays a schematic of the experimental apparatus including the LAOF and custom made JP-8 burner. Light extinction measurements were performed using a 10 mW He–Ne laser operating at 632.8 nm, a 30 mW diode laser operating at 856 nm, a 3 mW diode laser operating at 1314 nm, and a 1.5 mW diode laser operating at 1565 nm. Measurements were performed using a combination of the 632.8 nm, 1314/632.8 nm, and 1565/632.8 nm)



Fig. 1. Schematic of the large agglomerate optics facility (LAOF).

to minimize experiment-to-experiment variation on the spectral dependence of K_e . A rotating beam block was attached in front of each light source to selectively monitor the wavelength of interest. The beam was then directed through the cell to silicon photodiode detectors using gold-coated mirrors for the 856/632.8 nm experiment. For the 1314/632.8 nm and 1565/632.8 nm experiments, both the silicon and InGaAs photodiode detectors were used. An infrared pass filter was placed in front of the InGaAs detector to discriminate against the 632.8 nm light.

Soot from JP-8 fuel was produced using a burner constructed of 3/8" copper tube (see Fig. 2). A brass Swagelok union tee connecting the burner tube, fuel inlet, and nitrogen inlet was filled with a fiberglass material (thin-fiber 'angel hair') to enhance evaporation of the fuel. The fuel was metered into the burner at a rate of 0.3 ml/min using a syringe pump. Nitrogen flow ranging from 35 to 50 cc/min was metered into the tube assembly to control the flame height and sooting propensity. The Swagelok assembly and the copper tubing were heated using an electrical resistance heater to a temperature of 450 K. A thin-mesh screen was placed around the flame to minimize the disturbances from the



Fig. 2. Schematic of the laminar JP-8 burner.

ambient surroundings. The position of the burner was adjusted with respect to the hood to minimize the



Fig. 3. Laser transmittance through the LAOF cell using a 632.8 nm laser.

disturbance from ventilation flow. A sampling probe was used to collect soot from fume hood and to introduce the soot into the transmission cell.

Prior to introducing soot into the transmission cell, the incident intensity, I_0 (at 632.8, 856, 1314, or 1565) nm), was monitored for approximately 60 s beginning at time denoted by A in Fig. 3. Soot produced from the burner was then introduced into the transmission cell through suction from a vacuum pump attached at the lower end of the tube (note that the soot and gas exhaust bypasses the collection filter) at time denoted by B. The filling of the transmission cell with soot decreases the transmitted laser intensity (beginning at time denoted by B) until steady state conditions are obtained (denoted by time C). After attaining steady transmitted laser intensity measurements, the exhaust flow containing soot was directed through a glass fiber filter assembly to collect the soot for a period of approximately 5 min. after which clean air was used to purge the transmission cell of the soot. The incident intensity measurement was again performed (at time D) to ascertain whether soot was deposited on any of the optical components during the collection period. The mass of the collected soot, m_s (collected on the glass fiber filter), was then weighed using a microbalance (with 2-3 µg sensitivity). The gravimetrically-determined soot volume fraction, f_{yg} , can be obtained using the following relationship:

$$f_{\rm vg} = \frac{m_{\rm s}}{\rho_{\rm s}} V \tag{4}$$

where V is the volume of collected gas, m_s is the mass of collected soot and ρ_s is the density of soot (1.74 g/cc [11]). Noting that the soot volume fractions measured using light extinction technique, f_v , and the gravimetric

technique, are equal (since they were measured simultaneously for the soot in the transmission cell), f_v can be replaced with f_{vg} in Eq. (1) to calculate the value of K_e that provides correspondence. For each experiment, the dimensionless extinction constant was then determined using the measured intensity ratio and the gravimetrically-determined soot volume fraction:

$$K_{\rm e} = \frac{-\ln\left(\frac{I}{I_0}\right)\lambda}{f_{\rm vg}L} = \frac{-\ln\left(\frac{I}{I_0}\right)\lambda\rho_{\rm s}}{m_{\rm s}L}V \tag{5}$$

Absolute calibration experiments for this measurement technique were performed using an aerosol of monodisperse polystyrene spheres. The comparison of the measured K_e for the calibration aerosol to the actual value (using the known refractive index of the polystyrene spheres of 496 nm in diameter) indicates an error of less than 4%.

3. Results and discussions

Fig. 4 and Table 1 display the measured K_e for JP-8 soot as a function of wavelength as well as values for acetylene and ethene soot for the same wavelength range [13] and JP-8 soot [14] at 632.8 nm measured in previous studies. The uncertainty limits for these experiments correspond to the estimated total expanded uncertainty (95% confidence level) based on both Type B and Type A uncertainties [15]. The Type B uncertainties are based on scientific judgments rather than statistical means and equal 2.7% of the mean value. The Type A uncertainties (which are evaluated by statistical methods) based on



Fig. 4. K_e measurement for JP-8, ethene and acetylene at 632.8, 856, 1314 and 1565 nm.

Table 1 Measured K_e values of JP-8 soot from present study and K_e values of acetylene and ethene soot from previous investigation [11]

Wavelength (nm)	JP-8 <i>K</i> _e	Ethene $K_{\rm e}$	Acetylene $K_{\rm e}$
632.8	9.87	9.65	8.12
856	9.95	9.35	8.83
1314	9.75	9.17	9.12
1565	9.76	9.19	10.0

five repeat measurements on each of three days equal about 2-5% of the mean value.

As shown in Fig. 4, the K_e for JP-8 soot was nearly constant as a function of wavelength. The values of $K_{\rm e}$ for JP-8 soot are generally higher than the values for ethene and acetylene (with the exception of the 1565 nm value for acetylene). The spectral dependence of K_e for JP-8 soot is also uniquely different from that of acetylene and ethene soot, which further discounts the use of a universal value of K_e for interpretations of light extinction measurements. The increase of $K_{\rm e}$ with wavelength observed for acetylene was attributed to the diminishing influences of beam shielding at the higher wavelengths [13,16]. The reduction in ethene soot K_e with wavelength may be caused by the smaller magnitude of scattering at the higher wavelengths. The differences in $K_{\rm e}$ for the various fuels and their dependence on wavelength may be attributed to differences in the refractive indices and the physical and fractal properties of the soot. Additional experiments involving chemical analysis and TEM analysis and modeling efforts [16] will need to be performed to provide insights into these effects.

The measured values of K_e for JP-8 soot are also significantly higher than the calculated values traditionally used in the combustion and fire community [6]. The calculated values range from 4.89 to 6.10 (for wavelengths ranging from 632.8 to 1565 nm using the refractive indices of Dalzell and Sarofim [6]) whereas the measured K_e range from 9.75 to 9.95 for the same spectrum. The potential errors introduced into the soot volume fraction measurements when using the calculated K_e (instead of the measured values) are related to the ratio of the measured and calculated K_e values. For example, at 632.8 nm wavelength, the use of K_e equal to 4.89 over predicts the soot volume fraction by more than 100%.

The magnitude and the spectral variation of the K_e values can also influence the temperature measurements using the two wavelength pyrometry technique (see Eq. (2)). As the wavelength is increased into the near-infrared spectrum, the scattering contribution to the total extinction is expected to decrease, leading to the Rayleigh-limit relationship for the extinction constant of $K_e = 6\pi E(m)$. Using this relationship, the term $E(m_1)/E(m_2)$ in Eq. (2)

can be replaced by K_{e1}/K_{e2} . For comparison, at λ_1 equal to 632.8 nm and λ_2 equal to 856 nm, the measured K_{e1}/K_{e2} is equal to 9.87/9.95 whereas the calculated value is equal to 4.89/5.20. The errors in the temperature resulting from the use of the calculated K_e versus the measured K_e are muted (resulting from the temperature appearing in the exponential term) compared to the errors introduced into the soot volume fraction. However, the use of the measured values of K_e in the near-infrared will produce more reliable temperature information.

Complications arising from the spectral dependence of radiative emission and absorption from gaseous and particulate components produced in flames and fires dictate a simpler approach to estimate the radiative heat transfer behavior. A commonly used method to simplify the radiative heat transfer formulation is to introduce the Planck-mean absorption coefficient for optically thin environments ($K_eL \ll 1$) [17,18]:

$$K_{\rm p} = 3.83 f_{\rm v} K_{\rm e} T / C_2 \tag{6}$$

As in the case of light extinction measurements, the measured values of K_e will significantly increase the absorption coefficient compared to the Rayleigh-limit solution calculation. The use of the measured K_e will increase the Planck-mean absorption coefficients by more than a factor of two.

4. Conclusions

The dimensionless extinction constant, K_e , was measured in the visible to near-infrared spectrum for soot produced from JP-8 flames. These experiments represent the first measurement of K_e for soot produced from JP-8 flames. The K_e values did not display significant spectral variations that were observed for pure fuels such as acetylene and ethene. The measured K_e values ranged from 9.75 to 9.95 in the wavelength range from 632.8 to 1565 nm. These measurements provide more reliable values of K_e for use in optical-based soot diagnostics for soot concentration and temperature measurement as well as for soot radiation analysis.

Acknowledgements

The authors would like to acknowledge financial support from SANDIA National Laboratory through grant BF-9295. The authors wish to acknowledge the helpful discussions provided by Dr. Christopher Shaddix at Sandia National Laboratories, Livermore, California, and the assistance in measurements provided by Mr. Marco Fernandez of NIST.

References

- L.A. Gritzo, Y.R. Sivathanu, W. Gill, Transient measurement of radiative properties, soot volume fraction and soot temperature in a large pool fire, Combust. Sci. Technol. 139 (1998) 113–136.
- [2] J.M. Suo-Antilla, P. Drozda, L.A. Gritzo, M.Y. Choi, Characterization of soot morphology and optical properties from large pool fires, ASME IMECE Paper 33972, 2002.
- [3] M. Klassen, J.P. Gore, Temperature and soot volume fraction statistics in toluene-fired pool fires, Combust. Flame 93 (1993) 270–278.
- [4] M.Y. Choi, A. Hamins, G.W. Mulholland, T. Kashiwagi, Simultaneous optical measurement of soot volume fraction and temperature in premixed flames, Combust. Flame 99 (1994) 174–186.
- [5] Y.R. Sivathanu, G.M. Faeth, Soot volume fractions in the overfire region of turbulent-diffusion flames, Combust. Flame 81 (1990) 133–149.
- [6] W.H. Dalzell, A.F. Sarofim, Optical constants of soot and their application to heat-flux calculations, Transaction of the ASME, J. Heat Transf. 91 (1969) 100–104.
- [7] S.C. Lee, C.L. Tien, Optical constants of soot in hydrocarbon flames, in: Proceedings of the Eighteenth Symposium (International) on Combustion, 1981, pp. 1159–1166.
- [8] Z.G. Habib, P. Vervisch, On the refractive index of soot at flame temperature, Combust. Sci. Technol. 59 (1988) 261– 274.
- [9] K.C. Smyth, C.R. Shaddix, The elusive history of m = 1.57-0.56i for the refractive index of soot, Combust. Flame 107 (1996) 314–320.

- [10] I.M. Kennedy, Models of soot formation and oxidation, Progr. Energy Combust. Sci. 23 (1997) 95–132.
- [11] M.Y. Choi, G.W. Mulholland, A. Hamins, T. Kashiwagi, Comparisons of soot volume fraction using gravimetric and light extinction techniques, Combust. Flame 102 (1995) 161–169.
- [12] S.S. Krishnan, K.C. Lin, G.M. Faeth, Optical properties in the visible of overfire soot in large buoyant turbulent diffusion flames, ASME J. Heat Transf. 122 (2000) 517– 524.
- [13] J.Y. Zhu, M.Y. Choi, G.W. Mulholland, S.L. Manzello, L.A. Gritzo, J.M. Suo-Antilla, Measurement of visible and near-IR optical properties of soot produced from laminar flames, in: Proceedings of the Twenty Ninth Symposium (International) on Combustion, The Combustion Institute, Sapporo, Japan 2002, pp. 2367–2374.
- [14] A.M. Irrera, Measurement of Dimensionless Extinction Constant for Soot Produced from Various Liquid Fuels, M.S. Dissertation, Department of Mechanical Engineering and Mechanics, Drexel University, 2002.
- [15] B.N. Taylor, C.E. Kuyatt, Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results, NIST Technical Note 1297, Gaithersburg, MD, 1994.
- [16] G.W. Mulholland, R.A. Mountain, Coupled dipole calculations of extinction coefficient and polarization ratio for smoke agglomerates, Combust. Flame 119 (1999) 56– 68.
- [17] R. Siegel, J.R. Howell, Thermal Radiation Heat Transfer, fourth ed., Taylor and Francis-Hemisphere, Washington, 2001.
- [18] M.F. Modest, Radiative Heat Transfer, McGraw-Hill, Inc., New Jersey, 1993, pp. 421–432.