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# A 3D numerical simulation of laser-induced incandescence of soot particles in coal combustion products*

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**Abstract:** Laser-induced incandescence (LII) has received increasing attention as a potentially powerful technique for in-situ measuring of the volume fraction and primary size of soot particles in combustion systems. In this study, a 3D Monte Carlo simulation combined with a Mie equation was developed to analyze the influence of spectral absorption and scattering on the measured LII flux emitted by soot particles. This paper represents a first attempt to analyze soot measurement using the LII technique in coal combustion products. The combustion products of gases ($CO_2$, $N_2$), soot, and fly-ash particles, present between the location of laser-excited soot and the LII flux receiver. The simulation results indicated that an almost Beer-Lambert exponential decrease in LII flux occurred with an increase in the volume fraction of soot particles, while a nearly linear decrease occurred with an increase in the volume fraction of fly-ash particles. The results also showed that scattering effects of both soot and fly-ash particles on the LII flux could be neglected. Compared with the absorption of gases, a decrease of 20% of LII flux was observed with soot particles, and a decrease of 10% with fly-ash particles.

**Key words:** Laser-induced incandescence (LII), Soot, Fly-ash, 3D Monte Carlo, Scattering, Absorption
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## INTRODUCTION

Recent studies indicate that dispersed fine particles are a component of $PM_{2.5}$ emissions from coal-fired power plants (Stanmore *et al.*, 2001; Veranth *et al.*, 2000; Zheng *et al.*, 1993). The volume fraction of fine particulate correlates directly with respiratory and cardiovascular problems leading to mortality (Li *et al.*, 2003). The goal of ensuring global public health has led to an increased demand for stricter power plant emission standards and a need for measuring fine particle size distributions to quantify the $PM_{2.5}$. In addition, precise time-resolved measurements are needed for investigating combustion processes and for maximizing the collection efficiency of particulate control devices (Knutson and Whitby, 1975).

The laser-induced incandescence (LII) technique has been considered as a potential in-situ technique for measuring fine particle distribution in a non-intrusive way with extremely high time resolution (Roth and Filippov, 1996; Schulz *et al.*, 2006; Snelling *et al.*, 1997). The LII technique involves rapidly heating soot particles to vaporisation temperature by means of a high energy laser pulse. This results in enhanced thermal radiation, i.e., incandescence. Recent studies (Axelsson *et al.*, 2001; Dasch, 1984; Snelling *et al.*, 2000) indicated that the detected LII signal could be used to infer volume fraction and primary particle size of soot particles in flames and stack emissions.

However, the incident LII flux at the detector is not the *true* value emitted by laser-heated soot particles because of the interference of absorption, scat-

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tering, or radiative emission from participating media such as gases, reactive species, and solid particles. A study of the bias generated by light scattering and probe-volume heterogeneity on the measured soot signal was reported for a 1D on-line measurement (Murphy and Shaddix, 2005). The results indicated that in-scattering augment to the measured LII intensity could be approximately balanced by out-scattering attenuation. In addition, the derived soot temperature was strongly weighted towards the properties of the "hot" soot in a bimodal probe volume. Numerical results (Chen *et al.*, 2007) indicated that soot temperature derived from the ratio of uncorrected LII fluxes at two detecting wavelengths could be lower than the *true* value because a greater decrease in the spectral flux occurs at shorter wavelengths. Dauen *et al*.(2008) demonstrated this phenomenon by simulating the scattering of aggregated soot particles using a backward Monte Carlo method.

To the best of our knowledge, the LII signal bias of soot particles in coal combustion products, including gases and multicomponent particles, has not been investigated, except in a few recent studies (Chen, 2005; Murphy and Shaddix, 2005; Chen *et al*., 2007; Dauen *et al*., 2008). Such knowledge could be used directly to reduce the bias in measurements of soot particles determined in cases of media with mixed gases and soot particles. The present study focuses on the signal trapping analysis of soot measurement using the LII technique within the media of gases, soot, and fly-ash particles. A 3D Monte Carlo method combined with a Mie equation is developed to simulate LII emission transportation of laser-heated soot particles in a geometry containing complex coal combustion products exhibiting spectral dependent absorption and two kinds of scattering phenomena. The different levels of scattering and the absorption effects on the detected LII signal are discussed. The detected LII fluxes show the influence of particles and assumptions relating their absorption and scattering behaviors to different particle sizes and volume fractions are presented.

## THEORETICAL MODEL

An idealized soot particle measurement system using the LII technique in a cubical enclosure laden with coal combustion products is shown in Fig.1. The media of the combustion products are composed of gases ($CO_2$, $N_2$), soot and fly-ash particles at 300 K. The side length of the enclosure is 50 mm. A fixed reference frame ($x$, $y$, $z$) is defined, and the origin is set at the centre of the enclosure. An incident square laser beam with 2 mm sides is assumed to traverse the media along the $y$-axis, and illuminates soot particles suspended in the middle of the cubic enclosure. The LII radiation emitted by laser-heated soot particles is collected by a detector along the $x$-axis. Outside of the enclosure, the walls constitute an ideal blackbody resulting in no influence by the external media.

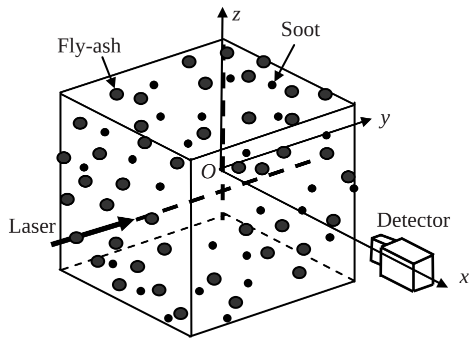


**Fig.1 A schematic configuration of an idealized system for detecting soot particles in coal combustion products using the LII technique**

To avoid interference of the incident laser beam on LII flux, we chose to simulate the beginning of the cooling phase of the heated soot particle, just after the laser pulse. In addition, all heated particles were assumed to be at a uniform temperature of 4000 K along the laser beam path, neglecting the possibility of on-line extinction of the laser excitation. Such an assumption can greatly simplify the numerical model but becomes a potential shortcoming when the loading of soot or fly-ash is high.

For an absorbing, emitting and scattering medium in an enclosure in local thermodynamic equilibrium, the equation that describes the light propagation can be written as (Howell, 1998)

$$\frac{\partial I_\lambda(\boldsymbol{x},\boldsymbol{\Omega})}{\partial x} = (k_{\text{ext},\lambda} - k_{\text{sca},\lambda})I_0 - k_{\text{ext},\lambda} I_\lambda(\boldsymbol{x},\boldsymbol{\Omega}) + k_{\text{sca},\lambda}\int_{W'=4\pi} I_\lambda(\boldsymbol{x},\boldsymbol{\Omega}') f_\lambda(\boldsymbol{x},\boldsymbol{\Omega}'\to\boldsymbol{\Omega})\,\mathrm{d}\boldsymbol{\Omega}', \quad (1)$$

where $I_\lambda(\boldsymbol{x}, \boldsymbol{\Omega})$ represents the spectral intensity at the location $\boldsymbol{x}$ along the direction $\boldsymbol{\Omega}$, $I_0$ is the spectral blackbody intensity, $k_{\text{sca},\lambda}$ and $k_{\text{ext},\lambda}$ are the spectral coefficients of scattering and extinction, respectively, and $f_\lambda(\boldsymbol{\Omega}'\rightarrow\boldsymbol{\Omega})$ is the scattering phase function of the coal combustion products in this study.

The solution of Eq.(1) here is based on a Monte Carlo approach. The LII method gives essentially time resolved information. Taking this time-resolved information, the Monte Carlo approach considers a snapshot in time during the process, and therefore is "time frozen".

The cubical enclosure is divided into several sub-volumes, in each of which a local reference frame ($x'$, $y'$, $z'$) is defined (Fig.2). The origin of the local reference frame is assumed to be in the centre of each sub-volume. The emissive spectra of laser-heated soot particle are divided into several uniform wavelength intervals defined as $[\lambda_i, \lambda_i+\Delta\lambda]$ ($i$=1, 2, …). Large numbers of photons or energy bundles are assumed to launch from each wavelength interval in each sub-volume of the laser-heated zone.

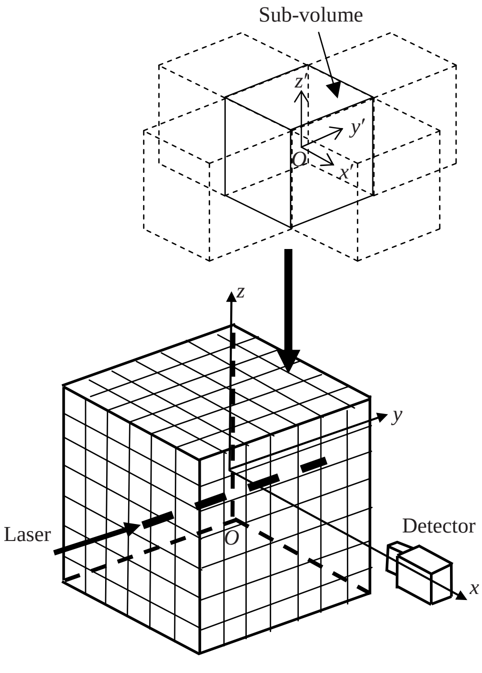


**Fig.2 Sub-volume division and local reference frame definition for numerical simulation**

Fig.3 shows the main flow chart of the 3D Monte Carlo method for each photon launched from the laser-heated zone in the coal combustion products, from the emissive source to the detector. The launch location $\boldsymbol{p}$ and direction $\boldsymbol{x}_k$ for each photon are described by Chen *et al*.(2007). The initial energy carried by each photon $w_0$ is calculated by

$$w_0 = 4a_\text{p}\sigma V_{ijk}F_\lambda T_\text{soot}^4 / N_\text{phot}, \tag{2}$$

where $a_\text{P}$ is the Planck mean absorption coefficient of soot particles, $\sigma$ is the Stefan-Boltzmann constant, $V_{ijk}$ is the volume of the sub-volume ($i$, $j$, $k$), $F_\lambda$ (Modest, 1992) is the blackbody radiation of laser-heated soot particles in the wavelength interval $[\lambda, \lambda+\Delta\lambda]$, $T_\text{soot}$ is the temperature of the laser heated soot particles in the sub-volume ($i$, $j$, $k$), and $N_\text{phot}$ is the number of photons launched in each wavelength interval of the sub-volume ($i$, $j$, $k$).

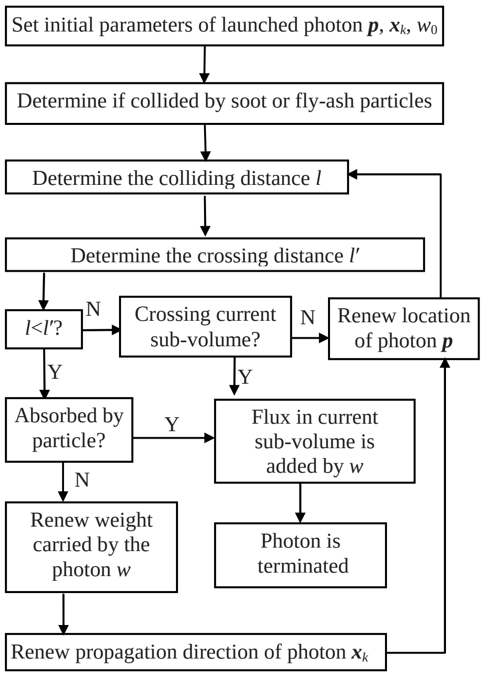


**Fig.3 Main flowchart of the 3D Monte Carlo simulation for coal combustion products**

The ratio of extinction coefficients of solid particles is defined by

$$A = \frac{k_\text{soot, ext}}{k_\text{soot, ext} + k_\text{fly-ash, ext}}, \tag{3}$$

where $k_\text{soot, ext}$ and $k_\text{fly-ash, ext}$ represent the extinction coefficients of soot and fly-ash particles, respectively. An equidistributed random number between 0 and 1,

$R$, is chosen and compared with the value, $A$. If the value of $R$ is less than $A$, the photon is collided by soot particles; otherwise it is collided by fly-ash particles. The travelling distance for the photon before colliding, $l$, is determined by

$$l = -\frac{\ln R}{k_{\text{ext}}}, \tag{4}$$

where $k_{\text{ext}}$ is the extinction coefficient of the collided particles. The random number, $R$, is compared to the scattering albedo to decide whether the photon is absorbed or scattered by the collided particle. If $R$ is greater than the value of the albedo, the photon is absorbed by the particle, otherwise it is scattered. The direction of the photon after scattering is determined by the scattering phase function of the collided particle. The energy carried by the photon is renewed by

$$w = w'\left[1 - \exp(-k_{\text{g, abs}} l)\right], \tag{5}$$

where $w'$ is the energy carried by the photon before the collision event and $k_{\text{g, abs}}$ is the absorption coefficient of the gases.

## RESULTS AND DISCUSSION

During the coal combustion process, soot and fly-ash particles are localized in fuel rich hydrocarbon zones in the presence of polynuclear aromatic hydrocarbons (PAHs), which may be absorbed on the solid surface of the soot. When using optical diagnostics to characterize soot particles, the selection of collected wavelengths is generally performed avoiding the PAHs fluorescence and $C_2$ emissions. The value of 0.405 μm (±0.005 μm) is selected as the detecting wavelength bandwidth of the LII signal (Schulz *et al*., 2006).

### Optical properties of coal combustion products

$CO_2$ gas produces a highly varying spectral absorption profile with extremely strong absorption bands in the detecting wavelength bands (Chen, 2005). In addition, $N_2$ does not participate in radiative transport.

The soot and fly-ash particles in the coal combustion products were assumed to be spherical and dispersed. The optical properties of soot particles depend on temperature, wavelength, morphology, etc. The value of the complex refractive index of soot particles, $m_{\text{soot}}$, has been discussed in numerous publications (Dalzell and Sarofim 1969; Lee and Tien, 1980; Charalampopoulous and Chang, 1988; Krishnan *et al*., 2001). In this study, a constant value of 1.57–0.56i was chosen for $m_{\text{soot}}$, and 1.5–0.005i for $m_{\text{fly-ash}}$. Both of these values can be modified easily as computation parameters.

The Mie theory was adopted for calculating the cross sections of extinction and scattering, and the phase function for a single particle with the mean size, $d_p$ (Howell, 1998). The calculated results of the scattering phase functions (Fig.4) showed that soot particles had isotropic scattering while fly-ash particles had high forward scattering.

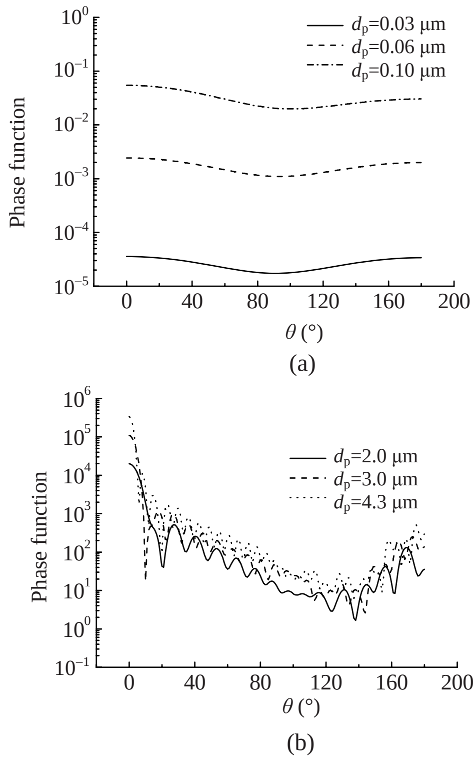


**Fig.4 The scattering phase functions calculated using the Mie equation for (a) soot particles; (b) fly-ash particles**

### Influence of coal combustion products

Typical chosen values were adopted for the cubic domain at atmospheric pressure: (1) At the laser-heated control zone: the mean diameter of the soot particles was $d_{\text{soot}}$=0.03 μm with the volume fraction of $1.40\times10^{-7}$, and no fly-ash particles existed; (2) Outside the laser-heated control zone: the diameter of the soot particles was $d_{soot}$=0.03 μm with $1.41\times10^{-6}$, the diameter of the fly-ash particles was $d_{fly\text{-}ash}$=2 μm with $8.37\times10^{-6}$; (3) The value of the gases absorption coefficients $k_{abs, gas}$ was assumed to be 0.001 $m^{-1}$ in the entire domain.

As a base case, the cube contained only the gases $CO_2$ and $N_2$. Soot and fly-ash particles were progressively introduced to incrementally increase the complexity. Fig.5 shows the simulated results as a function of distance along the *x*-axis with the different assumed coal products. The values of the LII flux decrease with increasing path distance. This phenomenon accords with the Beer-lambert law (Siegel and Howell, 1981). A typical decrease in the wall flux is observed when soot particles are added to the flue gas. Similarly, a decrease in the wall flux occurred when fly-ash particles were added with the same order extinction to the media of gases and soot particles.

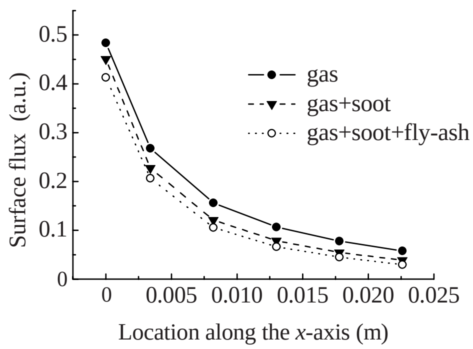


**Fig.5 Predicted LII wall flux in the assumed media of coal combustion products with the mean diameter of particles: $d_{soot}$=0.03 μm with the volume fraction of $1.40\times10^{-7}$ in the laser-heated control zone; $d_{soot}$=0.03 μm with $1.41\times10^{-6}$, and $d_{fly\text{-}ash}$=2 μm with $8.37\times10^{-6}$ outside the laser-heated control zone; $k_{abs, gas}$=0.001 $m^{-1}$ in the whole domain**

## Effect of particle size

To investigate the particle size effect on LII signal propagation, the numerical simulation was made when either soot or fly-ash particles existed outside the laser-heated control zone. Fig.6 shows the calculated surface flux at the positive *x*-axis with soot particle sizes between 0.001 and 0.1 μm and the volume fraction less than $1.8\times10^{-5}$. The obtained surface fluxes decrease exponentially with the increase in volume fraction of soot particles. A similar tendency is shown for the fluxes with increasing particle size.

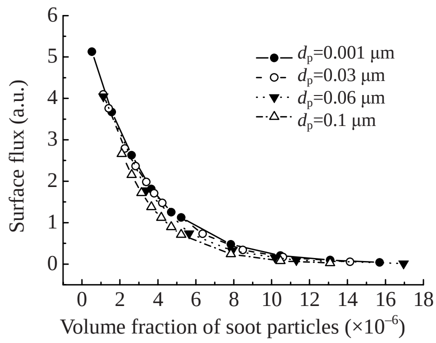


**Fig.6 The calculated result of LII flux arriving on the surface of medium at the positive *x*-axis vs the volume fraction of soot particles**

Fig.7 shows the results of the calculated surface flux at the positive *x*-axis for different sizes of fly-ash particles between 1 and 4.3 μm. The results show that the surface LII flux decreased with an increase in the volume fraction of the fly-ash particles. At the same volume fraction, the results show that the value of the surface flux increased with an increase in the size of the fly-ash particles.

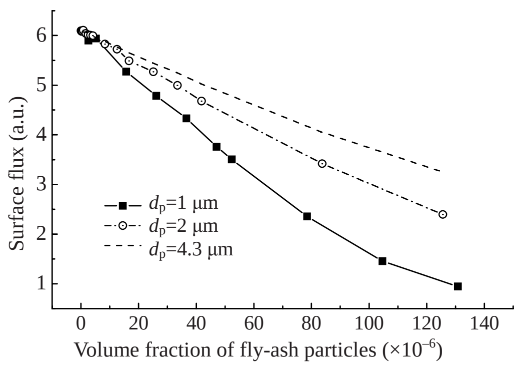


**Fig.7 The calculated result of LII flux arriving on the surface of medium at the positive *x*-axis vs the volume fraction of fly-ash particles**

## Scattering influence of solid particles

For each set volume fraction of solid particles, the numerical calculations were made considering isotropic, anisotropic and neglected scattering, respectively. The results are presented in Fig.8 and Fig.9 for the soot and fly-ash particles, respectively.

The surface flux decreased exponentially with an increase in the volume fraction of soot particles at the mean diameter of 0.03 μm (Fig.8). The scattering effect on the fluxes showed a similar tendency. This phenomenon is explained by the strong absorption characteristics of soot particles. The relation of flux and volume fraction can be calculated according to the Beer-lambert law. From Fig.8, it is clear that the scattering effect of soot particles can be neglected.

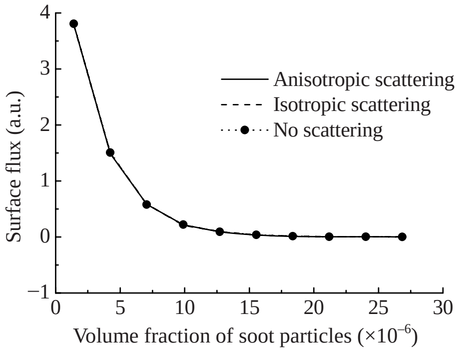


**Fig.8 Different scattering effects of soot particles on the measured surface LII flux**

Compared with soot particles, larger discrepancies among the curves corresponding to anisotropic, and isotropic scattering occurred for the fly-ash particles (Fig.9). In addition, the curve for calculations based on non-scattering is closer to that for anisotropic scattering than it is to the curve for isotropic scattering. This is explained by the strongly forward scattering behavior of the fly-ash particles, as mentioned above.

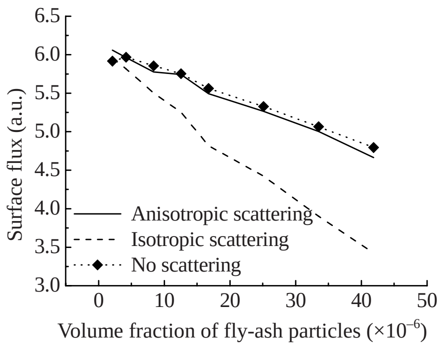


**Fig.9 Different scattering effects of fly-ash particles on the measured surface LII flux**

## CONCLUSION

The effects of absorption and scattering of coal combustion products on the LII signal of soot particles were investigated using the 3D Monte Carlo method combined with the Mie theory.

The effect of soot particle sizes on the surface LII flux was investigated using mean diameters ranging from 0.001 to 0.1 μm, and volume fraction of less than $1.8\times10^{-5}$. The calculated results indicated that the effect of soot particles of different sizes in this range on the detected LII flux was almost the same.

With an increase in the volume fraction of fly-ash particles, the surface LII flux decreased for several particle sizes. At the same volume fraction of particles, the simulated results showed that the value of the surface flux increased with an increase in the size of the fly-ash particles.

The simulated results showed that real scattering of soot particles can be assumed to be isotropic or neglected. Therefore, under the present simulated conditions, both neglecting of scattering and isotropic scattering for soot particles are reasonable approximate approaches. This conclusion will reduce the complexity of analysis during LII measurement of coal combustion products.

For the fly-ash particles, neglecting scattering gave results closer to anisotropic scattering than to isotropic scattering.

All the computations and analyses in this study were used to investigate soot particles in coal combustion products using the LII technique. However, the developed Monte Carlo method could be extended to forecast the measurement accuracy of laser-based diagnostics in denser media including three or more participating ingredients.

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