Numerical Simulation of Laser-Induced Detonation in Multi-Phase Mixtures

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Abstract: Simulation of interaction of laser pulse with inert and combustible gas-particle and gas-droplet mixtures plays an important role in environmental and engineering applications. The injection of metal particles with low evaporation temperature and ionization potential or liquid droplets causes optical breakdown on individual particle or droplet, and leads to drop of detonation minimum pulse energy of the mixture. The physical and mathematical models and up-to-date numerical methodology for computer modeling are developed and validated. Laser-induced detonation in aerosol systems is studied, and possibilities of the new methodology are demonstrated.

Key–Words: Droplet, Particle, Combustion, Detonation, Optical Breakdown, Two-Phase Flow

1 Introduction

The subject of the study stems from the increasing role of particulate reacting flows appearing in industry, technology and environment. Interaction of laser pulse with gas-particle mixtures plays an important role in different applications including environmental monitoring of high-risk industrial objects and enclosed spaces (coal mines, oil storages and others), measurements of flammability and explosibility limits in particulate reacting substances and propagation of laser radiation through explosive mixtures, laser-induced volumetric explosion for application to fire mitigation, design of air-breathing pulse detonation engines. Use of laser pulse allows to create desired temporal and spatial distributions of ignition centres and to perform a homogeneous ignition within the sub-microsecond interval. Laser ignition has the potential to replace the conventional electric spark plugs in engines that are required to operate under much higher compression ratios, faster compression rates, and much leaner fuel-to-air ratios than engines today.

The role of particles and droplets in environmental and engineering applications is two-fold. On the one hand, particles and droplets may pose potential hazard for human activity (erosion of surface and deposition of aerosols in human lungs). On the other hand, they can be successfully used in engineering solutions (to induce working processes in energy systems and to suppress acoustic instabilities of thermal processes). Processes that control transport and combustion of particles and droplets remain unresolved, and introduce significant uncertainties into modeling and simulation. One of the most important parameters for practical applications is the minimum pulse energy (MPE) required to induce ignition and detonation of the mixture.

When a power laser pulse \( I_s \sim 10^{11} \text{ W/cm}^2 \) interacts with a gas, the gas breaks down and becomes highly ionized \([1,2]\). This process is always accompanied by a light flash and generation of sound. The development of electron cascade requires the existence of initial free electrons in a gas.

Particles or droplets, trapped by a laser beam, considerably influence results of the process \([3]\). It is well known from experiments that for every particle size of any material there is threshold intensity at which the particle material converts into the metastable condition and its intense evaporation leads to heat destruction of the particle, either by means of local jetting of the essential part of the particle mass or due to explosion of the particle (optical breakdown).

The injection of metal particles with low evaporation temperature and low ionization potential (e.g., aluminium) leads to drop of detonation MPE \( (I \sim 10^9 \text{ W/cm}^2) \) due to optical breakdown on individual particles. Vapour aureole around metal particle is a source of free electrons, and optical breakdown in the gas-particle mixture comes for lower energy of laser pulse than in pure gas.

Many experimental, theoretical and numerical studies have been performed for the past years \([3–9]\). However, some fundamental and practical problems...
are yet to be resolved. They include qualitative and quantitative description of processes around individual particle and droplet, knowledge in particle microphysics and optical properties of particles, sub-models of heating and evaporation, transport of aggregates of complex morphology, threshold values of optical breakdown, dependence of MPE on the contributing factors (laser pulse, composition of gas mixture, shape of particles).

Physical and mathematical models of optical breakdown on individual particle and droplet, and up-to-date numerical methodology for computer modeling are developed. Laser-induced detonation in gas-particle and gas-droplet mixtures is studied, and advantages of the new methodology are demonstrated. The in-house computer code has been developed, and contribution of parameters of laser pulse and composition of the mixture is studied. Comparison of some numerical results with experimental data is made.

2 Laser Pulse

The time of laser pulse, its shape and intensity define the interaction of laser pulse with individual particle or droplet and mixture.

The intensity of laser pulse is represented as a product of the maximal intensity, \( I_{m0} \), the function describing the time distribution of the intensity, \( f_1(t) \), the function taking into account the spatial distribution of the intensity, \( f_2(r) \), and the function describing absorption of radiation in the medium, \( f_3(z) \). The intensity of laser pulse is

\[
I(t, r, x) = I_{m0} f_1(t) f_2(r) f_3(z),
\]

where \( t \) is time, \( r \) is radial coordinate, and \( z \) is coordinate indicating direction of propagation of laser beam.

The theoretical peak intensity of laser pulse at any radial point is calculated for given power and degree of focus. The laser does not reach its peak operating power at the moment when it is turned on. It requires a short time to ramp up to its peak output. For a laser pulse which lasts 8 \( \mu \)s, the laser output reaches its peak intensity in about one fourth of a pulse duration and will have dropped to roughly three fourth of its peak value when the laser is shut off. The laser model includes a ramp time parameter during which time the laser’s output increases linearly to a maximum (Figure 1).

The time distribution of the intensity is represented by a continuous piecewise-linear function

\[
f_1(t) = \sum_{k=1}^{N-1} \left[ I_k + (I_{k+1} - I_k) \frac{t - t_k}{t_{k+1} - t_k} \right] \times \phi(t_k, t_{k+1}),
\]

where \( t_k \) and \( I_k \) are time and intensity of laser pulse, and \( N \) is a number of ramp points. The function \( \phi(t_k, t_{k+1}) \) is given by

\[
\phi(t_k, t_{k+1}) = \frac{t - t_k + |t - t_k|}{2|t - t_k| + \varepsilon} - \frac{t - t_{k+1} + |t - t_{k+1}|}{2|t - t_{k+1}| + \varepsilon},
\]

where \( \varepsilon \) is the small value used to avoid division by zero.

A continuous piecewise-linear representation of pulse shape is used to compute the temporal characteristic

\[
S = \int_0^\infty f_1(t)dt = \frac{1}{2} \sum_{k=1}^{N-1} \frac{I_{k+1} + I_k}{t_{k+1} - t_k}.
\]

In a plane normal to the direction of laser pulse, the spatial distribution of the intensity is described by the normal distribution (Figure 2)

\[
f_2(r) = \exp \left( -\frac{2r^2}{R^2} \right),
\]

where \( r \) is a radial distance from centreline of the laser beam, and \( R \) is a radius of laser spot.

The absorption of radiation is described by Bouguer–Lambert–Beer law

\[
f_3(z) = \exp (-\mu z),
\]

where \( \mu \) is the absorption coefficient depending on the nature, state and fraction of particles as well as on the wave length of laser radiation.
Figure 2. Intensity of laser pulse as a function of radial coordinate (radius of laser spot is 5 mm)

The total energy of laser pulse is related to its intensity

\[ Q = \int_0^{2\pi} \int_0^\infty \int_0^1 I_0 f_1(t) f_2(r) r \, dr \, d\varphi \, dt, \]

where \( \varphi \) is a polar angle.

3 Breakdown mechanism

Physical model of optical breakdown provides qualitative description of the processes, particularly interaction of laser pulse with individual metal particle and liquid droplet.

3.1 Metal particle

A chain of processes leading to explosion and optical breakdown of individual metal particle was developed (Figure 3). These processes depend on optical properties of particle, its shape and ratio of particle size to radius of laser spot.

![Figure 3. Optical breakdown on metal particle](image)

The particle is heated up to high temperature, melting and evaporation start (Figure 3a). Evaporation of a particle leads to the formation of a vapour aureole around the particle. Free electrons are generated in the vapour aureole as a result of thermal emission from particle surface (if \( T < T_b \)) and isothermal ionization in vapour aureole (if \( T > T_b \)). This leads to collisions of electrons with ions and atoms and electron–electron collisions. Ionization of vapour aureole due to reverse drag effect leads to development of electron avalanche and formation of micro-plasma spots around the particle (Figure 3b). The cascade ionization process is significant at high pressure and longer laser pulse because under these conditions, electron–atom or electron–ion collisions have sufficient time to occur during the laser pulse [2]. Micro-plasma spots are expanded due to thermal diffusion of electrons and ionisation of molecules and atoms of surrounding gas. Micro-plasma spots are merged, and plasma fireplace is formed around the ensemble of particles (Figure 3c). The plasma fireplace absorbs laser radiation, and contributes to development and propagation of self-sustaining shock wave in the gas-particle mixture (Figure 3d).

3.2 Liquid droplet

Compared to metal particle, heating and evaporation of liquid droplet are delayed due to weak absorption of laser radiation. Concentration of free electrons in vapour aureole is insufficient for development of electron avalanche. In this case, the key mechanism of development of optical breakdown is explosive evaporation of droplet (Figure 4).

![Figure 4. Optical breakdown on liquid droplet](image)

The laser radiation focuses inside a droplet near its shadow side (Figure 4a). In this region, overheating conditions arise, and liquid is in meta-stable state
in which its temperature exceeds the temperature of saturated vapour at given temperature. Internal vapour cavity is formed, and liquid boils off in this cavity (Figure 4b). Increase in pressure in the vapour cavity creates conditions for internal micro-breakdown. Internal micro-plasma spot is appeared and absorbs laser radiation (Figure 4c). Further increase in pressure in the vapour cavity forms shock wave expanding inside droplet (Figure 4d). Expansion of shock wave induces thermal ionization of surrounding gas on shock wave front (Figure 4e). Free electrons that have appeared on shock wave front induce chain mechanism of breakdown, receiving their energy due to reverse drag effect (Figure 4f). Intense vaporization of droplet leads to the thermal destruction of the droplet either by means of local jetting of essential part of droplet mass or by its explosion.

4 Mathematical Model

The mathematical formulation of the problem is divided into low-level and high-level models. Low-level models correspond to the processes in the volume occupied by an individual particle or droplet. The high-level models correspond to the processes in the volume occupied by multi-phase mixture.

4.1 Basic assumptions

The diffusion processes define the evolution of the mixture on the time scales, which are much longer than the time of laser pulse. In calculations, the time of laser impulse is 8 μs, and the diffusion is not taken into account in equations for gas phase. In other cases, when long laser pulses are considered, the account for molecular diffusion is very important for accurate calculation of properties and evolution of multi-phase mixture.

To justify these assumptions, it is necessary to carry out the estimation of time scales of the problem considered. The time scales of the problem considered are the following:

\[ \tau_1 \sim \frac{\rho_p r_p^2}{Y D}, \quad \tau_2 \sim \frac{r_p^2}{a_p}, \quad \tau_3 \sim \frac{r_p^2}{a_g}, \]

\[ \tau_4 \sim \frac{r_p^2}{D}, \quad \tau_5 \sim \frac{c_p \rho_p r_p^2}{\chi_g(T)}, \quad \tau_6 \sim \frac{r_p}{u_s}, \]

where \( \tau_1 \) is the time of burn-out of a particle, \( \tau_2 \) and \( \tau_3 \) are the transient period of temperature inside and outside a particle, \( \tau_4 \) is the transient period of concentration field of oxidant and combustion products, \( \tau_5 \) is the time of heating of a particle up to temperature \( T \) under the laser radiation, and \( \tau_6 \) is the time of gas-dynamical processes for scale of particle size.

The results of time scales estimations for spherical aluminium particle in air are shown in the Figure 5 using logarithmic scales, where heat conductivity of gas is 0.026 W/m K, thermal conductivity of gas is \( 2.10 \cdot 10^{-5} \) m²/s, density of particle is 2700 kg/m³, thermal conductivity of particle is \( 6.68 \cdot 10^{-5} \) m²/s, specific thermal capacity of particle is 880 J/kg K, oxidant mass fraction is 0.3 kg/m³, diffusion coefficient of oxidant is \( 1.8 \cdot 10^{-5} \) m²/s. These values are typical for the problem considered.

![Figure 5. Typical time scales of the problem](image)

For metallic particle, for example aluminium, the time scales \( \tau_1, \ldots, \tau_6 \) are of next lower order than the time of laser pulse \( \tau_1 \). It is opposed to water droplets for which \( \tau_1 \approx \tau_5 \). and the model of thermal thick particle should be used to simulate their heating and evaporation [7]. In later case, the non-stationary equation of thermal conductivity is solved inside and outside the particle with corresponding boundary conditions on the phase interface. If temperature of particle reaches boiling temperature and evaporation begins, the problem is complicated because phase interface is moved (in this case, coordinate transformation is introduced to simplify numerical solution of thermal conductivity equation).

4.2 Low-level models

Low-level models describe melting, heating, evaporation and formation of vapour aureole, appearance of free electrons due to thermal ionisation on front of shock wave, and development of electron avalanche due to reverse drag effect.

To compute optical properties of particle (e.g., absorption efficiency of laser radiation), semi-empirical data are used. There are detailed data about temperature dependencies of optical, thermal and physical
properties of aluminium, because it often occurs in practice.

Heating model is based on numerical solution of unsteady heat diffusive equation [7, 10].

The equations describing electron avalanche in the vapor aureole include the equation of heating of vapor aureole due to electron–atom collisions, the equation of warming-up of electrons, the ionization kinetic equation of vapor as a result of electron impact, and the equation of particle mass.

The plasma in vapor aureole is considered as an ideal gas. The Euler equations are used for the simulation of gas dynamical processes in vapor aureole.

The detailed chemistry model is not important to develop the low-level model. Use of detailed chemistry model in the volume occupied by an individual particle requires high computational costs. A simple model of one-step chemical reaction is used in order to reproduce explosion of individual particle.

Threshold value of optical breakdown on an individual particle is computed as a result of the solution of low-level models. Low-level models are incorporated in high-level models that describe detonation of the mixture.

### 4.3 High-level models

The multi-phase mixture consists of some gas and particulate components (Figure 6). Gas phase consists of a combustible component (fuel), an oxidant component, a component that is combustion product, and a neutral component. Particulate phase represents ensemble of metal particles and liquid droplets. Vapour phase is a product of thermal decomposition of metal particles, and it consists of atoms, ions and electrons. Condensed phase represents metal oxide that is a result of vapour condensation.

![Figure 6. Composition of multi-phase mixture](image)

The Eulerian approach is used to formulate governing equations describing detonation of the mixture. The governing equations are written for all gas and particulate components of the mixture. The diffusion processes are ignored because they define the evolution of the mixture on the time scales, which are much longer than the time of laser pulse.

The simplified kinetic model is used (e.g., combustion model of methane–air mixture includes 15 equations).

### 4.4 Source terms

The data obtained from solution of low-level problems are used to calculate source terms in the governing equations describing high-level problem. It is assumed that particles are uniformly distributed in the domain. Some volume of the mixture depending on particle volume fraction is associated with each particle (individual reactor of a particle). The model of unsteady well-stirred reactor is used to calculate physical quantities in this volume.

The solution of high-level problem provides fraction of particles and volume occupied by a particle.

### 5 Computational Procedure

The problem considered is multi-physical and multi-scale. The main feature of the problem is correlation and interference of physical, gas dynamics and chemical processes, and a wide range of temporal and spatial scales. Metal particles have a non-spherical shape.

The in-house computer code has been developed. The equations are solved numerically based on finite volume method, splitting scheme on physical factors, piecewise parabolic method and Chakravarthy–Osher scheme for inviscid fluxes. Approximate Riemann solver is employed to calculate the interfacial fluxes on the first fractional time step. Chemical reactions and interphase exchange of mass, momentum and energy are considered at the second fractional time step. The internal time step is used to ensure stability of numerical scheme. Pseudo-gas of particles does not have internal pressure, so artificial pressure is introduced to design similar computational procedures for gas and particulate components.

### 6 Results and Discussion

The results concern processes near individual particle and droplet, and detonation of multi-phase mixture. Particle location relative to centreline of laser beam, energy, time and shape of laser pulse vary in calculations. The output quantities are threshold value of optical breakdown and detonation MPE.

#### 6.1 Metal particle

The Figure 7 shows heating of metal particle up to the boiling temperature. The particle temperature de-
pends on total energy of laser pulse and distance from particle to centreline of laser beam.

![Figure 7. Temperature of metal particle](image)

Development of electron avalanche in the form of dependence of degree of ionization of vapour aureole on time is shown in the Figure 8. The particle is located on the centreline of laser beam. The electron avalanche is developed in 0.68 µs from the laser pulse started, and ionization takes place within a short time interval (it is about 0.04 µs).

![Figure 8. Development of electron avalanche](image)

The degree of ionisation as function of time and energy of laser pulse is shown in the Figure 9. Micro-plasma spots around the particle are formed at energy of 1.03 J. Pre-breakdown conditions are sensitive to small change of energy of laser pulse. The threshold value of plasma formation is defined as a part of power passed up to the beginning of breakdown.

Interaction of laser pulse with individual metal particle is related to one of the following stages.  
- Pre-threshold energy of laser pulse ($Q < 1$ J). Energy of laser pulse is not enough to ionize vapour aureole around particle. Evaporation of particle exists but degree of ionisation is small, and vapour aureole is transparent for laser radiation.  
- Near-threshold energy of laser pulse ($Q = 1–2$ J). Degree of ionisation changes from some percents to 100%.  
- Post-threshold energy of laser pulse ($Q > 2$ J). The process proceeds at completely ionized vapour aureole around particle. Ionisation has an avalanche character within short time interval.

6.2 Liquid droplet

The temperature field in a water droplet and surrounding is shown in the Figure 10. The laser beam falls on the droplet from the right to the left. A local temperature rise is observed on the exposed surface of droplet. A thin thermal boundary layer is formed in the vicinity of the droplet. Increase in temperature inside droplet corresponds to the centre of internal vapour cavity which is located near shadow side of a droplet. The droplet is superheated and water is in the meta-stable state. Line 7 corresponds to the start of explosive process at temperature of 698 K. Time of explosive transformation of a droplet is 1.54 µs.

The plasma spot is non-transparent to radiation. Increase in pressure and temperature induces expansion of shock wave. Its intensity decreases with increase in distance from the centre of vapour cavity.

The Figure 11 shows the threshold value as a function of droplet radius. The time of optical breakdown is a result of the competition between 3 factors: (i) time of droplet heating to the temperature of explosive transformation (at low pulse energy, the large droplets do not have enough time for being heated, and the small droplets exchange heat intensively with the surrounding), (ii) intensity of the shock wave contributing to the thermal ionization of vapour (for massive droplets, the shock wave is weak), (iii) time of
development of an electron avalanche.

Figure 11. Threshold value on optical breakdown

The size of the laser spot has a significant impact on the threshold value of optical breakdown. For droplets of 5 µm, a breakdown takes place at energy of 10 J. Increase in droplet radius leads to increase in time of droplet heating and decrease in degree of ionization. No electron avalanche develops at low intensity of laser pulse, and threshold value of optical breakdown increases.

6.3 Minimum pulse energy

The minimum pulse energy is a function of radius of laser spot, mass fraction of particulate component and volume fraction of oxidant.

The results obtained are presented in the Figure 12 for fish-plate aluminium particles in the acetylene–oxygen–nitrogen mixture. Volume fraction of acetylene is 15%. Volume fraction of oxygen changes from 15% to 35%. Mass fraction of particles is 1 g/m³. Wave length of laser beam is 4.2 µm, radius of laser spot is 1.5 cm, and time of pulse is 2.6 µs [11, 12].

Figure 12. Minimum pulse energy of detonation

At \(Q = 150\) J combustion of fuel takes place in small region adjacent to shock wave front. At \(Q = 200\) J the temperature and pressure in shock wave front increase, and volume fraction of fuel decreases on 20–30% for time of laser pulse. Further energy supply to mixture leads to considerable increase in temperature and pressure in the mixture, and development of unsteady gas dynamics processes in vapor aureole. At \(Q = 300\) J about 60% of fuel is used, and at \(Q = 300\) J about 95% of fuel burns beyond the shock wave front. The energy of laser pulse 350–400 J is the MPE of detonation.

6.4 Specific fronts

The Figure 13 shows the depth of penetration of laser pulse into multi-phase mixture and locations of melting front (line 1), evaporation front (line 2) and plasma formation front (line 3) as functions of mass fraction of particles. The melting, evaporation and plasma formation take place under the corresponding lines.

When the mass fraction of particles increases, the regions of melting, evaporation and plasma formation reduce.

7 Conclusion

The chain of events leading to the optical breakdown on the individual metal particle of non-spherical shape and liquid droplet, and detonation of gas-particle mixture has been developed.

The mathematical formulation of the problem was divided into low-level models and high-level models. The low-level models correspond to the processes in the volume occupied by an individual
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References:


