Contents

Preface  ix
List of Contributors  xiii

Part I. Bénard and Thermocapillary Instabilities  1
1 Nonlinear Dynamics of Thin Evaporating Liquid Films Subject to Internal Heat Generation  3
ALEXANDER ORON
2 The Effect of Air Height on the Pattern Formation in Liquid–Air Bilayer Convection  15
D. JOHNSON, R. NARAYANAN, AND P. C. DAUBY
3 The Third Type of Benard Convection Induced by Evaporation  31
WEN-JEI YANG
4 Waves Generated by Surface-Tension Gradients and Instability  43
M. G. VELARDE, A. YE. REDNIKOV, AND H. LINDE
5 Thermocapillary-Coriolis Instabilities  57
ABDELFATAH ZEBIB AND CÉDRIC LE CUNFF

Part II. Shear and Pressure Driven Instabilities  71
6 Control of Instability in a Liquid Film Flow  73
S. P. LIN AND J. N. CHEN
7 Three-Dimensional Waves in Thin Liquid Films  85
A. A. NEPOMNYASHCHY
8 Modulation Wave Dynamics of Kinematic Interfacial Waves  99
H.-C. CHANG, E. A. DEMEKHN, R. M. ROBERTS, AND Y. YE
9 Multifilm Flow Down an Inclined Plane: Simulations Based on the Lubrication Approximation and Normal-Mode Decomposition of Linear Waves  112
C. POZRIKIDIS
10 Spatial Evolution of Interfacial Waves in Gas–Liquid Flows  129
M. J. MCCREADY
11 The Shear Breakup of an Immiscible Fluid Interface  142
GRÉTAR TRYGGVASON AND SALIH OZEN UNVERDI
12 Two-Fluid-Layer Flow Stability  156
S. ÖZGEN, G. S. R. SARMA, G. DEGREZ, AND M. CARBONARO
### Part III. Waves and Dispersion

<table>
<thead>
<tr>
<th>Chapter</th>
<th>Title</th>
<th>Authors</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>13</td>
<td>On Modeling Unsteady Fully Nonlinear Dispersive Interfacial Waves</td>
<td>THEODORE YAOTSU WU</td>
<td>171</td>
</tr>
<tr>
<td>14</td>
<td>Instabilities in the Coupled Equatorial Ocean–Atmosphere System</td>
<td>HENK A. DUKSTRA AND PAUL C. F. VAN DER VAART</td>
<td>179</td>
</tr>
<tr>
<td>15</td>
<td>Large-Amplitude Solitary Wave on a Pycnocline and Its Instability</td>
<td>DANIEL T. VALENTINE, BRIAN C. BARR, AND TIMOTHY W. KAO</td>
<td>198</td>
</tr>
<tr>
<td>16</td>
<td>Stability and Pattern Selection in Parametrically Driven Surface Waves</td>
<td>PEILONG CHEN AND JORGE VIÑALS</td>
<td>211</td>
</tr>
<tr>
<td>17</td>
<td>Deformation and Rupture in Confined, Thin Liquid Films Driven by Thermocapillarity</td>
<td>MARC K. SMITH AND DAVID R. VRANE</td>
<td>221</td>
</tr>
<tr>
<td>18</td>
<td>Linear and Nonlinear Waves in Flowing Water</td>
<td>CHIA-SHUN YIH AND WILLIAM W. SCHULTZ</td>
<td>234</td>
</tr>
<tr>
<td>19</td>
<td>Pinned-Edge Faraday Waves</td>
<td>DIANE M. HENDERSON AND JOHN W. MILES</td>
<td>246</td>
</tr>
<tr>
<td>20</td>
<td>Interfacial Shapes in the Steady Flow of a Highly Viscous Dispersed Phase</td>
<td>DANIEL D. JOSEPH AND RUNYUAN BAI</td>
<td>254</td>
</tr>
</tbody>
</table>

### Part IV. Multiphase Systems

<table>
<thead>
<tr>
<th>Chapter</th>
<th>Title</th>
<th>Authors</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>21</td>
<td>Interaction between Fluid Flows and Flexible Structures</td>
<td>WEI SHYY, HENG-CHUAN KAN, H. S. UDAYKUMAR, AND ROGER TRAN-SON-TAY</td>
<td>265</td>
</tr>
<tr>
<td>22</td>
<td>Numerical Treatment of Moving Interfaces in Phase-Change Processes</td>
<td>SURESH V. GARIEMELLA AND JAMES E. SIMPSON</td>
<td>278</td>
</tr>
<tr>
<td>23</td>
<td>Accuracy and Convergence of Continuum Surface-Tension Models</td>
<td>M. W. WILLIAMS, D. B. KOTHE, AND E. G. PUCKETT</td>
<td>294</td>
</tr>
<tr>
<td>24</td>
<td>Interaction of Convection and Solidification at Fluid–Solid Interfaces</td>
<td>L. BÜHLER, A. EHRHARD, AND U. MÜLLER</td>
<td>306</td>
</tr>
<tr>
<td>25</td>
<td>Interfacial Motion of a Molten Layer Subject to Plasma Heating</td>
<td>P. S. AYYASWAMY, S. S. SRIPADA, AND I. M. COHEN</td>
<td>320</td>
</tr>
<tr>
<td>26</td>
<td>The Fluid Mechanics of Premelted Liquid Films</td>
<td>M. G. WORSTER AND J. S. WETTLAUFER</td>
<td>339</td>
</tr>
<tr>
<td>27</td>
<td>Recent Advances in Lattice Boltzmann Methods</td>
<td>SHIYI CHEN, GARY D. DOOLEN, XIAOYI HE, XIAOBO NIE, AND RAOYANG ZHANG</td>
<td>352</td>
</tr>
<tr>
<td>28</td>
<td>Bubble Dynamics in Heterogeneous Boiling Heat Transfer</td>
<td>RENWEI MEI, JAMES F. KLAUSNER, AND GLEN THORNCROFT</td>
<td>364</td>
</tr>
</tbody>
</table>

### Part V. Complex Flows

<table>
<thead>
<tr>
<th>Chapter</th>
<th>Title</th>
<th>Authors</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>29</td>
<td>Heat, Mass, and Momentum Exchanges between Outer Flow and Separation Bubble behind a Single-Backward-facing Step with Gas Injection from One Duct Wall</td>
<td>TONG-MIN LIOU AND PO-WEN HWANG</td>
<td>381</td>
</tr>
<tr>
<td>30</td>
<td>A Moving Boundary Problem Arising from Stratigraphic Modeling</td>
<td>J. MARR, J. B. SWENSON, AND V. R. VOLLER</td>
<td>393</td>
</tr>
</tbody>
</table>
Contents

31 Convection Generated by Lateral Heating of a Solute Gradient: Review and Extension
   C. F. CHEN
   403

32 Heat Conduction from a Solid Particle and the Force on It in Stokes Flow in a Fluid with Position-Dependent Physical Properties
   ANDREAS ACRIVOS AND YONGGUANG WANG
   413

33 Radiation-Affected Ignition Phenomena with Solid–Gas Interaction
   SEUNG-WOOK BAEK AND JAE HYUN PARK
   427

34 Biomagnetic Fluid Dynamics
   YOUSEF HAIK, VINAY M. PAI, AND CHING-JEN CHEN
   439

   The Man I Know: Chia-Shun Yih, July 25, 1918–April 25, 1997
   YUAN-CHENG FUNG
   453

Index 459
Radiation-Affected Ignition Phenomena with Solid–Gas Interaction

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Nomenclature

\[ C \quad \text{specific heat} \]
\[ d_p \quad \text{particle diameter} \]
\[ G \quad \text{incident radiation} \]
\[ I \quad \text{radiative intensity} \]
\[ L \quad \text{characteristic length} \]
\[ n_p \quad \text{particle number density} \]
\[ q^c \quad \text{conductive heat flux} \]
\[ q^r \quad \text{radiative heat flux} \]
\[ Q_{\text{abs}} \quad \text{absorption efficiency} \]
\[ Q_{\text{ext}} \quad \text{extinction efficiency} \]
\[ Q_{\text{sca}} \quad \text{scattering efficiency} \]
\[ T \quad \text{temperature} \]

Greek Symbols

\[ \beta \quad \text{extinction coefficient \( (= \kappa + \sigma_s) \)} \]
\[ \varepsilon_p \quad \text{solid particle emissivity} \]
\[ \varepsilon_w \quad \text{wall emissivity} \]
\[ \theta \quad \text{polar angle} \]
\[ \kappa \quad \text{absorption coefficient} \]
\[ \lambda \quad \text{thermal conductivity} \]
\[ \mu \quad \text{direction cosine \( (= \cos \theta) \)} \]
\[ \sigma \quad \text{Stefan–Boltzmann constant \( = 5.670 \times 10^{-8} \text{ W/m}^2 \text{ K}^4 \)} \]
\[ \sigma_g \quad \text{gas concentration} \]
\[ \sigma_p \quad \text{particle concentration} \]
\[ \sigma_s \quad \text{scattering coefficient} \]
\[ \tau_L \quad \text{optical thickness \( (= \beta L) \)} \]
\[ \Phi \quad \text{scattering phase function} \]
\[ \phi \quad \text{equivalence ratio} \]
\[ \omega_s \quad \text{scattering albedo \( (= \kappa / \beta) \)} \]

Subscripts

\[ b \quad \text{blackbody} \]
\[ g \quad \text{gas} \]
\[ p \quad \text{particle} \]
33.1. Introduction

In the area of fire safety, the physical state under which a reactive two-phase mixture (solid particles/gas) can ignite and generate a heat release is one of critical problems still remaining to be elucidated in further detail. Basic phenomena involved are controlled by the balance between heat release and heat losses to the surroundings through conduction, convection, and radiation. Given a suitable ignition source, the reactive solid particles or gases can react exothermically with an oxidant if the heat removal is not sufficient.

Thermal ignition of a reactive material has been well reviewed by Merzhanov and Averson (1971). The thermal ignition by conduction has been examined (Shouman et al., 1974; Kordylewski, 1979). As far as the radiative heat transfer is concerned, Khalil et al. (1983) developed a model predicting the stationary thermal ignition of a heat-generating particle suspension bounded by emitting and diffusely reflecting walls. Smith et al. (1988) examined the thermal ignition phenomenon with a single homogeneous absorbing and emitting medium. Generally speaking, when the ignition heat source is the external radiation, the existing theoretical models for the ignition of a two-phase mixture can be divided into two types, solid or gas phase ignition, depending on the key exothermic reaction leading to ignition, which were investigated in detail by Baek (1990, 1992, 1994, 1996).

From this viewpoint, in this chapter the past works on the radiation-affected ignition phenomena with solid–gas interaction are presented and then further research work to be done is discussed.

33.2. Basic Formulation

In order to take account of radiation in the modeling of ignition, the divergence of radiative heat flux is required to be added into the conventional energy conservation equation (Viskanta and Menguc, 1987):

\[
\begin{align*}
\rho \frac{Dh}{Dt} &= \frac{dp}{Dt} - \nabla \cdot \bar{q} + \Phi + \dot{Q} + \rho \sum_{k=1}^{N} Y_i f_k \cdot \vec{V}_k, \\
\nabla \cdot \bar{q} &= \nabla \cdot (\bar{q}_C + \bar{q}_R) \\
&= -\lambda \nabla T + \kappa \left( 4\sigma T^4 - \int_{4\pi} I \, d\Omega \right),
\end{align*}
\]

where \( \Phi \) is dissipation by viscous stress, \( \dot{Q} \) is the heat generation, and the fifth term on the right-hand side represents the body-force work.

To derive the radiative heat flux and its divergence, it is inevitable to solve the radiative transfer equation (RTE), which is given by highly nonlinear integro-differential form with spectrally varying properties:

\[
\frac{dI}{ds} = -\left( \kappa + \sigma_s \right) I + \kappa I_b + \frac{\sigma_t}{4\pi} \int_{4\pi} \Phi(\hat{s}, \hat{s}_i) I(\hat{s}_i) \, d\Omega_i,
\]

where \( s \) is the path length along the pencil of rays. The terms on the right-hand side of Eq. (3) represent the attenuation of radiative intensity due to absorption and out-scattering and the augmentation through emission and in-scattering, respectively.
There have been many attempts to solve the RTE for application to various industrial high-temperature systems. Among others they are the Monte Carlo method, zone method, spherical harmonics method \( (P_n) \), discrete ordinates method, finite-volume method (Raithby and Chui, 1990), etc. In one-dimensional geometry, the exact formulation or a more simplified approach such as differential approximation and two-flux method are used. Methods other than the finite-volume method are summarized in the classical text about radiation in detail (Modest, 1993).

Particles are the principal contributors to the absorption and scattering of radiation. These effects can be well accounted for in the limiting cases of very small or very large concentrations. The radiative properties of a cloud consisting of the uniform-sized particles can be described by (Modest, 1993; Siegel and Howell, 1992)

\[
\sigma_s = \pi d_p^2 n_p Q_{sca}, \quad \kappa = \pi d_p^2 n_p Q_{abs}, \quad \beta = \kappa + \sigma_s = \pi d_p^2 n_p Q_{ext},
\]

where \( \sigma_s, \kappa, \) and \( \beta \) are scattering, absorption, and extinction coefficient, respectively. If the particles are mainly diffusely reflecting spheres, the scattering, absorbing, and extinction efficiencies in Eqs. (4) can be simplified to

\[
Q_{sca} = 1 - \varepsilon_p, \quad Q_{ext} = 1, \quad Q_{abs} = \varepsilon_p.
\]

### 33.3. Ignition of Solid Particles due to Absorption of Radiation

As schematically shown in Fig. 33.1, a mixture of carbon particles and air is contained between two parallel walls. The system is one dimensional, and the mixture is assumed to be quiescent. Two walls held at different temperatures \( T_1 \) and \( T_2 \) are assumed to be diffuse reflectors and emitters. The carbon particles are assumed to be spherical and uniformly monodispersed in the gaseous phase. The particle volume is neglected in comparison with the suspension volume. The particles are supposed to absorb and emit as well as isotropically scatter radiation and are also assumed to have constant monochromatic radiative properties. The inner particle temperature is considered to be uniform.

Under the above assumptions, the energy conservation equations for gas and particles can be represented by

\[
\rho_g C_g \frac{\partial T_g}{\partial t} = \lambda_g \frac{\partial^2 T_g}{\partial x^2} - n_p Q,
\]

\[
\sigma_p C_p \frac{\partial T_p}{\partial t} = n_p Q - \frac{\partial q_R}{\partial x} + H_p \Gamma_p,
\]

where \( H_p \) is the heat of combustion per unit mass of fuel particle and \( \rho_g \) and \( \sigma_p \) are the gas density and the particle concentration, respectively. The amount of heat transferred between gas and one particle, \( Q \), is expressed as

\[
Q = \pi d_p^2 h(T_g - T_p),
\]

where the heat transfer coefficient \( h \) can be calculated from the Ranz–Marshall correlation for the Nusselt number (Ranz and Marshall, 1952). The particle burning rate \( \Gamma_p \) is given
by

\[ \Gamma_p = n_p \pi d_p q, \] (9)

where \( q \) is the carbon burning rate per unit external geometric surface (Field et al., 1967).

In a one-dimensional geometry, the exact forms for the incident radiation, the radiative heat flux, and its divergence can be obtained as follows (Sparrow and Cess, 1970):

\[
G(\tau) = 2B_1 E_2(\tau) + 2B_2 E_2(\tau_L - \tau) + 2 \int_0^\tau \left[ (1 - \omega_s) \sigma T_p^4(\tau') + \frac{\omega_s}{4} G(\tau') \right] \cdot E_1(|\tau - \tau'|) d\tau',
\] (10)

\[
q_R(\tau) = 2B_1 E_3(\tau) - 2B_2 E_3(\tau_L - \tau) + 2 \int_0^\tau \left[ (1 - \omega_s) \sigma T_p^4(\tau') + \frac{\omega_s}{4} G(\tau') \right] \cdot E_2(\tau - \tau') d\tau' - 2 \int_\tau^{\tau_L} \left[ (1 - \omega_s) \sigma T_p^4(\tau') + \frac{\omega_s}{4} G(\tau') \right] \cdot E_2(\tau' - \tau) d\tau',
\] (11)

\[
-\frac{dq_R(\tau)}{d\tau} = 2B_1 E_2(\tau) + 2B_2 E_2(\tau_L - \tau) - 4(1 - \omega_s) \sigma T_p^4 - \omega_s G(\tau) + 2 \int_0^\tau \left[ (1 - \omega_s) \sigma T_p^4(\tau') + \frac{\omega_s}{4} G(\tau') \right] \cdot E_1(|\tau - \tau'|) d\tau',
\] (12)
\[ B_1 = \varepsilon_1 \sigma T_1^4 + 2(1 - \varepsilon_1) \]
\[ \times \left\{ B_2 E_3(\tau_L) + \int_0^{\tau_L} \left[ (1 - \omega_p) \sigma T_p^4(\tau') + \frac{\omega_p}{4} G(\tau') \right] \cdot E_2(\tau') \, d\tau' \right\}, \quad (13) \]

\[ B_2 = \varepsilon_2 \sigma T_2^4 + 2(1 - \varepsilon_2) \]
\[ \times \left\{ B_1 E_3(\tau_L) + \int_0^{\tau_L} \left[ (1 - \omega_p) \sigma T_p^4(\tau') + \frac{\omega_p}{4} G(\tau') \right] \cdot E_2(\tau_L - \tau') \, d\tau' \right\}, \quad (14) \]

where the exponential integral function \( E_n(x) \) is defined by
\[ E_n(x) = \int_0^1 \mu^{n-2} \exp \left( -\frac{x}{\mu} \right) \, d\mu. \quad (15) \]

### 33.3.1. Ignition Delay

The energy equation with the divergence of radiative heat flux is numerically solved with or without radiation. Figure 33.2 illustrates the temperature distributions near two walls. When the radiation is included, the carbon particle temperature rapidly increases with the temperature of radiatively transparent air almost unchanged. Evidently it is due to the radiative absorption of carbon particles. The increase in total heat flux due to radiation makes the ignition delay shorter, as shown in Fig. 33.3. Here, ignition delay is defined by the time interval from an initial exposure of the mixture to the radiation to the onset of ignition.

Temporal variations of gas and particle temperatures are presented in Fig. 33.4. In the vicinity of the hot wall, the conduction is more dominant than radiation because of the large gas temperature gradient; therefore, the air is always hotter than the particles. Contrary to this, the particle temperature is higher than air in the region far from the hot wall because of the far-reaching effect of radiation. This is a noteworthy characteristic of radiation.

The influence by scattering albedo was found to be negligible since the scattering redistributes only the radiative intensity, while the change in the hot wall emissivity \( \varepsilon_2 \) was more influential in predicting the ignition delays (Baek, 1992).

The effect of the particle size on the ignition delay is shown in Fig. 33.5. When the particle size increases at a fixed loading ratio, the particle number density decreases. This in turn leads to a reduction of extinction coefficient as well as a decrease in the total particle surface area available to the convective heat transfer between gas and particles. Consequently the particles are rendered less radiatively active and the increasing rate of particle temperature becomes much slower as the particle size increases. Finally, this results in longer ignition delay.

However, the carbon mass loading is known to affect the ignition delays negligibly because of two counterbalancing effects on the particle temperature. An appreciable change in the lower wall temperature makes no noticeable difference in ignition delays either.

### 33.3.2. Influence of Nonuniform Particle Temperature

The effect of nonuniform inner particle temperature was also taken into account (Baek, 1992). This can be accounted for by adopting the energy equation for a single particle, which replaces the energy equation for a particle cloud. The ignition delay was found to become shorter compared with the uniform particle temperature case. As the particle size increased, the ignition delay also increased because of the large volume-to-surface-area ratio.
33.4. Ignition of Combustible Gas by Radiatively Absorbing Inert Particles

The combustible gas with a sufficient amount of inert particles, which is exposed to external radiation, also poses a serious safety problem because of its inherent ignition risk. Its risk results from the fact that the inert particles absorbing the radiation have the possibility of directly
igniting the combustible gas by conduction and convection, even if the gas is not in immediate contact with the ignition source.

Hill et al. (1992) numerically and experimentally investigated the ignition of a hydrogen/air mixture by laser-heated coal particles. However, a very simplified radiation model has been adopted. Using a more rigorous radiation model and detailed chemistry, Baek (1994, 1996) explored the ignition of a suspension comprising inert particles and combustible gas (H$_2$/air, C$_3$H$_8$/air) in slab geometry.

### 33.4.1. Mathematical Modeling

As schematically shown in Fig. 33.6, a mixture of inert aluminum oxide particles and combustible gas is contained between two transparent walls that are heated by the external blackbody radiative heat source maintained at a high temperature. A one-dimensional open system is assumed, neglecting the gas expansion. The assumptions taken in Section 33.3 still apply here.

However, different from the model in Section 33.3, the reaction term appears in the gaseous energy equation, not in the particle energy equation:

$$
\rho_g C_g \frac{\partial T_g}{\partial t} = \lambda_g \frac{\partial^2 T_g}{\partial x^2} - n_p Q - \sum_{i=1}^{N} \dot{\omega}_i \Delta H_{g,i},
$$

where $\dot{\omega}_i$ is the reaction rate for the $i$th species.

In this problem, the ignition source is the external radiation so that the two-flux radiation model is preferred. The net radiative heat flux $q^R$ can be expressed in terms of $q^+$ and $q^-$ in the forward and the backward directions. The governing equations for $q^+$ and $q^-$ can be derived by the integrating RTE over a hemisphere (Modest, 1993; Siegel and Howell, 1992):
Figure 33.4. (a) Temperature variation of carbon particles and air for $\phi = 1.0$, $L = 0.05$ m, $d_p = 50$ mm, $T_1 = 300$ K, and $T_2 = 2000$ K; (b) magnified regime adjacent to the hot wall.

\begin{align}
q^R &= q^+ - q^- , \\
\frac{dq^+}{dx} &= -2(\kappa + \alpha_s)q^+ + 2\sigma_s q^- + 2\kappa \sigma T_p^4 , \\
\frac{dq^-}{dx} &= 2(\kappa + \alpha_s)q^- + 2\sigma_s q^+ - 2\kappa \sigma T_p^4 .
\end{align}

33.4.2. Discussions

In order to predict the ignition delay of a gas mixture accurately, a precise estimation of the reaction rate of each gas species is definitely required so that usage of the multiple-step chemical
Figure 33.5. Effect of particle size on ignition delays for $\phi = 1.0$, $L = 0.05$ m, and $T_1 = 300$ K.

Figure 33.6. Schematic diagram for the ignition of the mixture of combustible gas and inert particles.

kinetics is highly demanded. While for a mixture of H$_2$/air a system of 10 species (H$_2$, O$_2$, N$_2$, H, O, N, OH, HO$_2$, H$_2$O, NO) with 32 elementary reaction steps has been adopted (Baek, 1994), for a mixture of C$_3$H$_8$/air a system of 31 species (C$_3$H$_8$, O$_2$, OH, H, O, N$_2$, H$_2$, H$_2$O, HO$_2$, CO, CO$_2$, CH$_4$, CH$_3$, CH$_2$O, HCO, C$_2$H$_6$, C$_2$H$_5$, C$_2$H$_4$, C$_2$H$_3$, C$_2$H$_2$, CH$_2$CO, CH$_2$, CH, C$_2$H, HCCO, iso-C$_3$H$_7$, n-C$_3$H$_7$, C$_3$H$_6$, CH$_3$HCO, H$_2$O$_2$, CH$_3$CO) with 123 elementary reaction steps has been chosen (Baek, 1996). A mixture of CH$_4$/air has also been examined with a system of 16 species (CH$_4$, O$_2$, H, O, OH, H$_2$, H$_2$O, N$_2$, HO$_2$, CO, CO$_2$, CH$_3$, CH$_2$O, HCO, CH$_3$O, H$_2$O$_2$) and 35 elementary reaction steps. The reaction rate $\omega_i$ for the $i$th species is calculated from the CHEMKIN subroutines (Kee et al., 1980).

As shown in Fig. 33.7, the particles warm up first because of the absorption of radiation, and the particle temperature is always higher than the gas temperature before ignition. The onset of ignition is therefore defined as the time required for two temperature curves to cross each other, as revealed in the figure, in which $T_e$ is the temperature of external radiative thermal source. In Fig. 33.8 the ignition delays for hydrogen, methane, and propane with air are plotted on a linear scale against the inverse of the external source temperature. In general, the ignition
delay time increases with decreasing $T_r$. The ignition delay is shown to be the shortest in the order of propane, hydrogen, and methane.

The change of particle number density at a fixed particle size exerts a strong influence such that the smaller the particle number density, the longer the ignition delay. The decrease in particle size at a fixed particle number density leads to a decrease in the extinction coefficient.
as well as the particle concentration. The particles then become less radiatively active so that their temperature increase slower. This finally leads to a substantial increase in ignition delay.

33.5. Conclusions and Recommendations

This chapter presented an ignition phenomenon of a two-phase mixture that is exposed to the external radiation. The absorption of radiation by inert particles is found to play a significant role in directly igniting the combustible gas mixture. The combustible solid particles are also discovered to be very hazardous, even if no heat source other than external radiation is available. In deriving the results above, we made several assumptions so that several recommendations for further research can be made.

First, the radiation effects by gases as well as particles are recognized. In the previous results only the radiation effects by particles were assumed, neglecting gas radiation. However, gases such as CO, CO$_2$, O$_2$, NO$_x$, SO$_x$, and CH$_4$ are known to be involved in thermal radiation so that the deliberation of two-phase radiation (both gas and particle radiation) deserves further exploration.

For a gray mixture in which for the particles emission, absorption, and scattering are considered while the scattering for the gas is neglected, the RTE can be written as (Park et al., 1998; Denison and Webb, 1993)

$$\frac{dI}{ds} + (\kappa_g + \kappa_p + \sigma_s)I = \kappa_g I_{bg} + \kappa_p I_{bp} + \frac{\sigma_s}{4\pi} \int_{4\pi} I(\hat{s}) \Phi(\hat{s}, \hat{s}_r) \, d\Omega_s,$$

where $I_{bg}$ and $I_{bp}$ are the blackbody emissive powers corresponding to local temperatures of the gas and the particles, respectively. Now, the divergences of the radiative heat flux, $\nabla \cdot q^R$ for gas and $\nabla \cdot q^R_p$ for particles, which are required for gas and particle energy equations, can be denoted by

$$\nabla \cdot q^R = \kappa_g \left(4\pi I_{bg} - \int_{4\pi} I \, d\Omega\right),$$

$$\nabla \cdot q^R_p = \kappa_p \left(4\pi I_{bp} - \int_{4\pi} I \, d\Omega\right).$$

Thereby the two-phase radiation effects can be examined in the future. Park et al. (1998) have already shown that the gas radiation together with particle radiation can play a significant role in determining the gas and particle temperature variations compared with the case of particle radiation only.

Second, the nongray behavior is an intrinsic characteristic of a material (Modest, 1993). Buckius and his colleagues have investigated the nongray behavior of a solid–gas mixture by experiments (Skoczypiec et al., 1987; Walters and Buckius, 1991). For a gas-only medium, many reliable theoretical models, such as the narrow-band model, wide-band model, weighted sum of gray gases model, etc., were reported (Modest, 1993). In contrast, the concrete theoretical nongray model of a solid–gas two-phase medium has not been proposed yet.

Third, when particles were dealt with in previous works they were assumed to be homogeneous and spherical. While Hottel and Sarofim (1967) have shown that the effects of particle shape on radiative energy transfer are negligible in systems involving a distribution of particle sizes, the shape of a particle may severely affect the ignition characteristics (Vorsteveld and Hermance, 1987). For this reason, it is necessary to explore further the effects of particle size.
and polydispersion of particles. Even though the radiative properties alone for the polydisperse mixture have been extensively studied (Menguc and Viskanta, 1985), their effects in practical applications are yet to be reported.

Finally, a careful exploration and consideration of changes in geometry and boundary conditions also remain to be identified.

References