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Two-dimensional numerical study of solid-solid combustion

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A detailed two-dimensional numerical study of the highly exothermic nature of synthesis of refractory materials is presented. The process is modeled by a two-dimensional, pseudohomogeneous model. The model equations, which are very stiff and exhibit varying time and length scales, are implemented in the commercially- available computational fluid dynamics code, FLUENT using user-defined-functions (UDFs). The implementation of the new model is validated using a sample problem solved earlier by Degreve *et al*. [1]. The new model correctly captured the complex behavior of the combustion front.

Key words: gasless combustion, FLUENT user-defined-functions, two-dimensional combustion front.

Introduction

A certain class of ceramic and metallic materials can be manufactured by the self-propagating high-temperature synthesis method. Even though by now this process has included reaction systems of the solid-gas and solid-liquid types, the original work was directed towards the reactions investigated where not only the reactants but also the final products formed in the solid phase [2, 3]. These reactions are therefore generally referred to as gasless, solid-solid or condensed phase combustion reactions. Borides, carbides, sulfides, silicides of metals and intermetallic compounds can be synthesized by this way. These materials are extensively used in high temperature applications, such as corrosion and wear-resistant coatings, thermal insulators and so on, because of their desirable physical and chemical properties.

Because of the obvious practical implications as well as from the standpoint of basic research, scientific investigation of this combustion processes is very important. Therefore, it can safely be said that the modeling and analysis of these systems continue to pose real challenges. The full power of analytical mathematical techniques and numerical simulation needs to be brought out to reveal the intricate complex dynamic behavior that can be displayed by these physical systems [4, 5, 6].

In the present study, the numerical simulation of a combustion wave propagating through a sample of compressed powder is considered. The powder consists of a mixture of two components, which upon reaction release a substantial amount of heat. After ignition of the sample, a combustion wave propagates through the reactant mixture, converting unburned reactants to products. Due to the high exothermicity of the chemical reaction, the combustion process is self-sustaining, needing no additional energy expenditure after the initial start-up period.

Experimental observations, numerical simulations and mathematical analysis have revealed that the propagation of a reaction front in the condensed phase can occur in a variety of different ways. One-dimensional behavior, where the front remains planar, is displayed as either steady state (constant velocity, constant pattern) or pulsating (oscillatory velocity)combustion. The pulsations are not necessarily periodic, but can become quite complex in nature [7]. In addition, nonplanar modes of propagation, where the front breaks up in the transverse direction, have been observed and analyzed. One or more hot spots may move along the surface of a cylindrical sample, following a screw-like path (spinning combustion), or one or more luminous points can be seen to appear, disappear and reappear repeatedly on the sample surface (multiple point combustion). The different types of behavior do not necessarily relate to different physical systems. As the amount of heat release is controlled by diluting the initial mixture of reactants with reaction products or as the initial temperature is changed by uniform preheating of the sample, one particular system can display the varying modes of dynamic behavior mentioned above.

It is hoped that the study of condensed phase combustion will lead to an understanding necessary to control the process itself and the properties of the product formed as well.

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Mathematical Model

We only briefly discuss the mathematical model for gasless combustion since the assumptions and derivation can be found in several publications [8, 9, 10]. The governing equations describing the reacting system under adiabatic conditions are:

energy balance

$$\rho C_p \frac{\partial T}{\partial t} = \lambda\left[\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2}\right] + k_0 C(-\Delta H)\exp\left(-\frac{E}{R_g T}\right) \quad (1)$$

and

mass balance

$$\frac{\partial C}{\partial t} = -k_0 C \exp\left(-\frac{E}{R_g T}\right) \quad (2)$$

The initial conditions (t=0) are given by:

$$T = T_0,\ C = C_0 \quad \text{for} \quad 0 \le y \le R,\ 0 \le x \le L \quad (3)$$

and the boundary conditions (t > 0) by:

for $t_H > t > 0$ and x = 0, 0 < y < R

$$T = T_H \quad (4a)$$

for $t > t_H$ and x = 0, 0 < y < R

$$T = T_0 \quad (4a)$$

for x = L and y = R

$$\frac{\partial T}{\partial n} = 0 \quad (4c)$$

It is more convenient to rewrite the model Eqs. (1) and (2) in dimensionless form as follows,

energy balance:

$$\frac{\partial \theta}{\partial \tau} = \frac{\partial^2 \theta}{\partial \xi^2} + \frac{\partial^2 \theta}{\partial \zeta^2} + (1-\eta)\exp\left(\frac{\theta}{1+\beta\theta}\right) \quad (5)$$

mass balance:

$$\frac{\partial \eta}{\partial \tau} = \gamma(1-\eta)\exp\left(\frac{\theta}{1+\beta\theta}\right) \quad (6)$$

The variables θ and η denote the dimensionless temperature and the conversion of the limiting component of the reaction, respectively. The behavior of the reaction front propagation in the condensed phase can be governed by two parameters:

$$\beta = \frac{R_g T_0}{E}, \quad \gamma = \frac{c_p R_g T_0^2}{(-\Delta H)E} \quad (7)$$

where β represents the inverse of the dimensionless activation energy and γ the inverse of the dimensionless temperature rise. The reaction-diffusion system is subject to the initial and boundary conditions given by

$\tau = 0,\ 0 \le \xi \le L_x$ and $0 \le \zeta \le L_y$

$$\theta = \theta_0 \text{ and } \eta = 0 \quad (8)$$

$\tau = 0,\ 0 \le \xi \le L_x$ and $0 \le \zeta \le L_y$

$$\theta = \theta(\zeta, \tau) \quad (9)$$

$\xi = L_x$ and $0 \le \zeta \le L_y$

$$\frac{\partial \theta}{\partial \xi} = 0 \quad (10)$$

$\zeta = 0,\ \zeta = L_y$ and $0 \le \xi \le L_x$

$$\frac{\partial \theta}{\partial \zeta} = 0 \quad (11)$$

This formulation describes the ignition process and subsequent reaction front propagation for a cylindrical sample of material. All sides except one have been assumed insulated. After sufficient heat has been supplied through the side at $\xi = 0$ by raising the temperature locally, ignition will occur and a reaction front will propagate through the material till it has consumed all the available reactants.

Model Implementation

A brief description of the user defined functions developed in this work is given below.

Description of "combustion.h"

There are a total of one user-defined-scalar (UDS) and four user-defined-memory variables (UDMs) defined in this problem. In the header file "combustion.h" all the parameters required for setting up the problem are given as inputs. The variable names and comment lines together give details of the units of different inputs.

Description of "comb-solv.c"

The function DEFINE_INIT (initialize_function, domain) is used for initializing all the user-defined-memory variables. This function is called when the problem is initialized in FLUENT.

The functions DEFINE_ADJUST (adjust_function, domain) is written for calculating θ and η during transient calculations. This function is called before each iteration. The comment statements before different pieces of this function explain what each part is doing.

The function DEFINE_SOURCE (theta_source, c, t, dS, eqn) sets the source/sink term in the dimensionless temperature.

The function DEFINE_UDS_UNSTEADY (UDS_ unsteady_condition, c, t, i, apu, su) sets transient terms for the user-defined scalar, UDS-0 and the user-defied-memory, UDM-1.

Description of "comb-prop.c"

The function DEFINE_DIFFUSIVITY (UDS_diffusion_coeff, c, t, i) computes the diffusivity value to be used in the user-defined-scalar equation.

Description of "comb-bc.c"

The function DEFINE_PROFILE (convective_bc_uds0, thread, i) is used for setting the flux boundary condition on different boundaries.

Results and Discussion

To test the performance of the FLUENT user-defined-function when used in conjunction with the numerical simulation of a physical problem, gasless or solid-solid combustion was a good choice. The reasons are as follows: First of all, the governing equations themselves are relatively simple in nature. Changes in temperature are governed by a partial differential equation containing only the Laplacian operator to account for heat conduction and a nonlinear term describing heat generation to the chemical reaction. Variations in conversion are obtained at each location by the integration of an ordinary differential equation. Simulation runs can therefore easily be constructed and are not unnecessarily made complicated by the presence of a multitude of governing equations and transport variables. The simplicity of the mathematical formulation in fact hides a lot of the difficulties encountered when actually trying to solve these equations numerically. Time scales in solid-solid and just plain combustion problems vary widely. Some regions in the physical domain are quite active and undergoing extremely rapid changes, whereas other regions appear dead. The biggest cause for worry though is the small length scale associated with the chemical reaction zone, and this is the second reason for choosing gasless combustion as a test case.

Since it is not the purpose here to perform an exhaustive study of gasless combustion, we have chosen to present results for only one set of dimensionless parameters governing this type of system. A nice choice is made by taking $\beta = 0.03$, $\gamma = 0.08$, $\theta_0 = -9.5$. The resulting system is known to exhibit periodic oscillations in one-dimensional simulations. Figures 1a and 1b show the calculated dimensionless temperature and conversion profiles at different times. The length of the physical system in the x-directions are L_x=200.0, and in the y-direction L_y=100.0. This represents quite a large system. The number of intervals chosen for the discretization of the domain in the x- and y-direction is 200 and 100, respectively. The computations for the results presented here relate to the simulation of a freely propagating solid flame. The initial guess for temperature and conversion profiles of the freely propagating flame was obtained as a natural transition between simulation on a fixed physical domain and

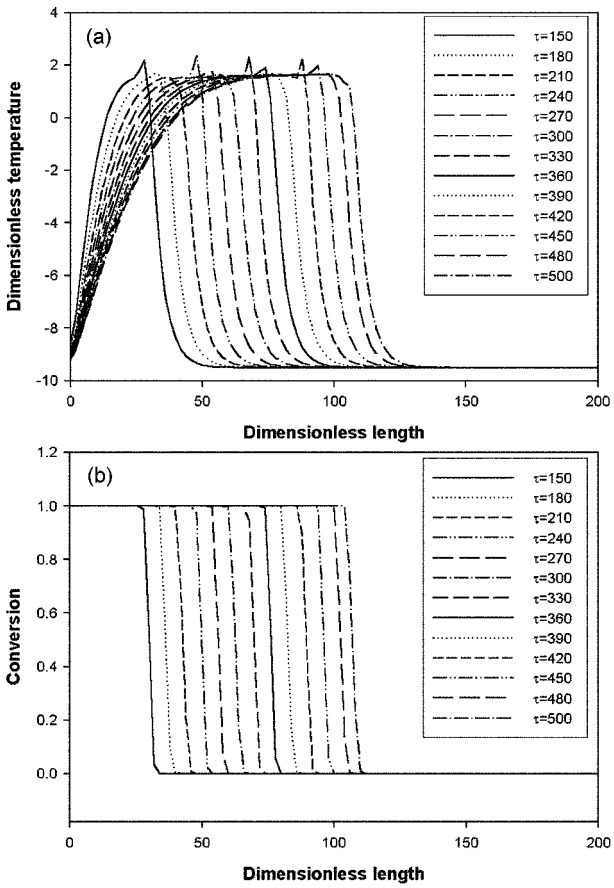

Fig. 1. (a) Dimensionless temperature profiles at different times, (b) Dimensionless conversion profiles at different times.

inside a reactor. The first part of the computations (not shown here) therefore consisted in igniting an initially cold slab of material at $\xi = 0$ using a high temperature source at $\theta = 3.0$ (the adiabatic temperature for this system) and watching the reaction front propagate. Upon reaching the axial position, $\xi = L_x/2$ the free flame option of the computer program was switched on, and thereafter we found that the system exhibited mild temperature oscillations, see Fig. 1a. Figure 1b gives the conversion profiles. In Fig. 2, the combustion

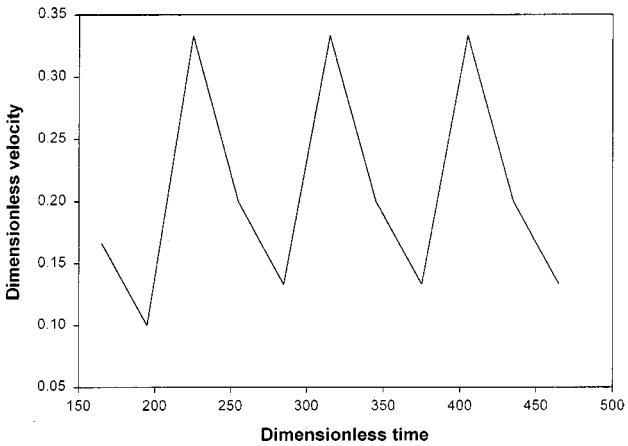

Fig. 2. Combustion front velocity vs. dimensionless time.

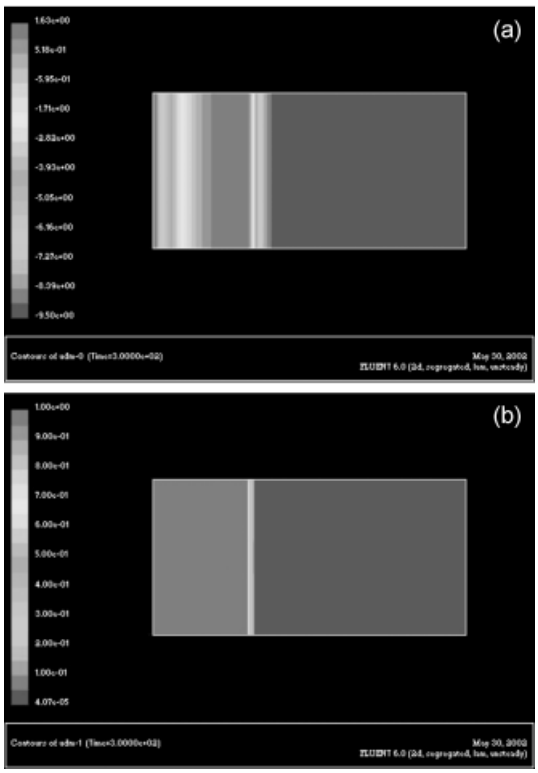

Fig. 3. (a) Temperature contour for freely propagating solid flame (t=300), (b) Conversion contour for freely propagating solid flame (t=300).

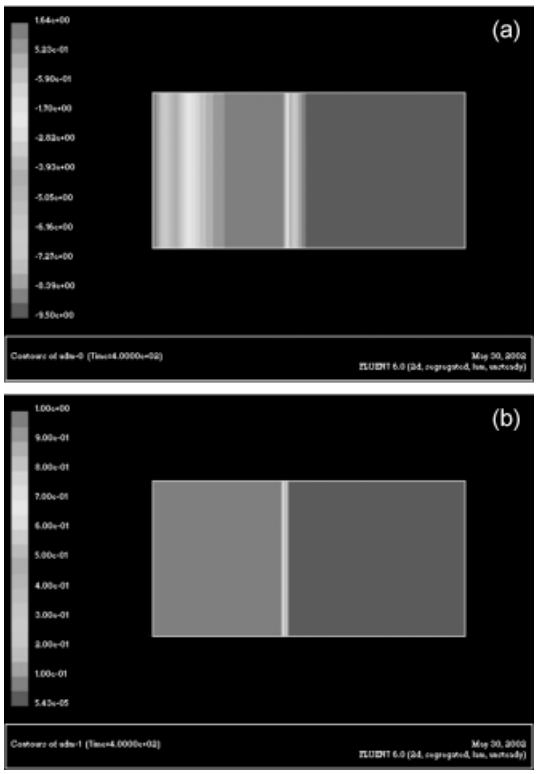

Fig. 5. (a) Temperature contour for freely propagating solid flame (t=400), (b) Conversion contour for freely propagating solid flame (t =400).

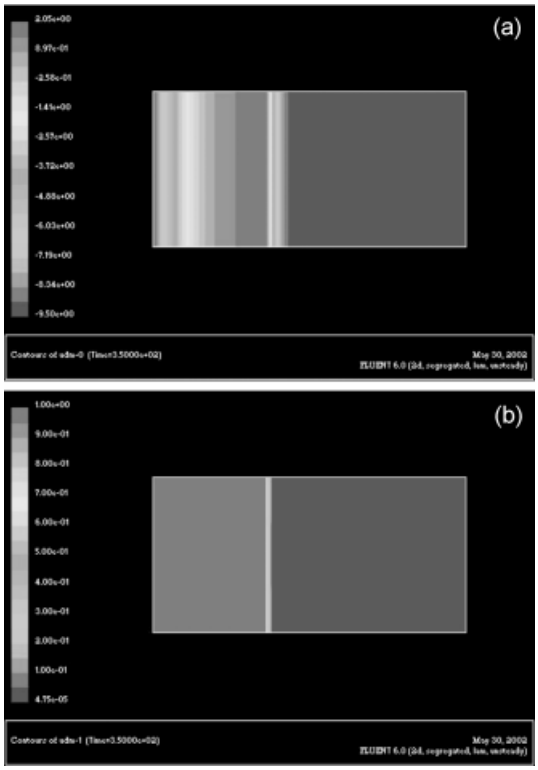

Fig. 4. (a) Temperature contour for freely propagating solid flame (t =350), (b) Conversion contour for freely propagating solid flame (t =350).

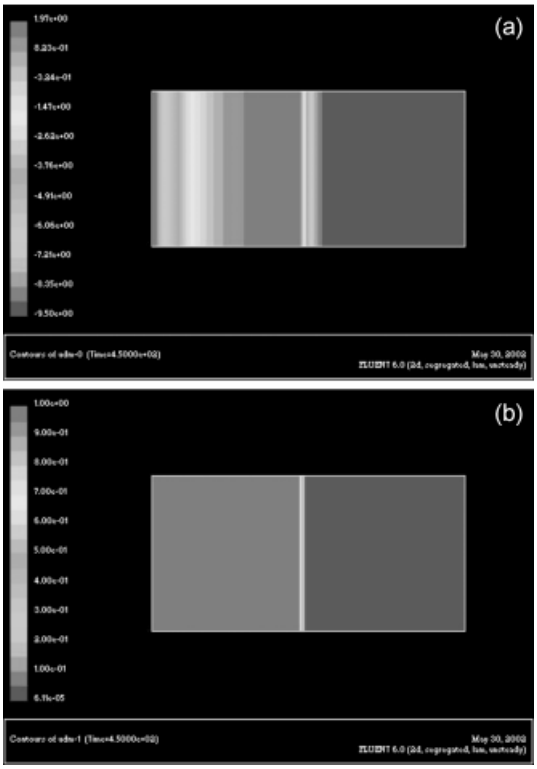

Fig. 6. (a) Temperature contour for freely propagating solid flame (t=450), (b) Conversion contour for freely propagating solid flame (t =450).

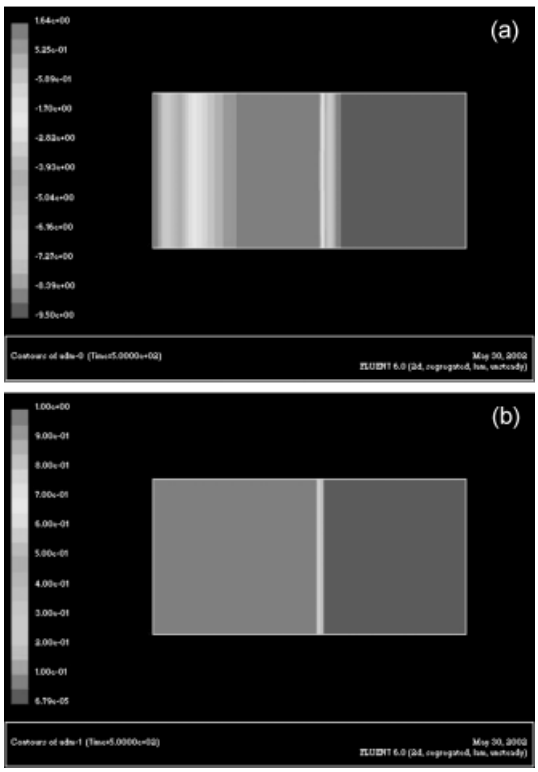

Fig. 7. (a) Temperature contour for freely propagating solid flame (t=500), (b) Conversion contour for freely propagating solid flame (t =500).

front velocity vs. dimensionless time is shown. After dimensionless time τ greater than 200 the combustion front velocity shows a periodic oscillation pattern. Figures 3a to 7b show the contours of two-dimensional temperature and conversion, respectively. The reaction zone is given by those regions in space where the conversion suddenly jumps from a value near zero to one as shown in Figs. 3b to 7b. It is interesting to observe that the thickness of the reaction zone increases and decreases as the combustion front propagates into the unreacted region in Figs. 3a to 7a. This behavior resembles what is observed during experiments where luminous regions appear, disappear and reappear, again.

Summary

In this work, a mathematical model describing solid-solid combustion in condensed system has been implemented. This model is validated by solving a sample problem given by by Degreve *et al*. [1]. The new model successfully captured the complex combustion front during the process. The predictions of the model are in close agreement with the results reported by by Degreve *et al*. [1].

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