Reaction-diffusion fronts in media with spatially discrete sources

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The exact solution for a reaction-diffusion front propagating in a heterogeneous system of discrete, point-like sources is obtained without resorting to a representation of the sources by a spatially continuous function. When the reaction time is smaller than the characteristic diffusion time between neighboring sources, the front speed predicted by this discrete source model differs from the continuum theory based on the spatial averaging of the heterogeneities. Furthermore, when the sources are regularly distributed in space, discreteness introduces a limit and propagation beyond this limit is only possible in a system with randomly distributed sources via local fluctuations of the concentration. The discrete regime of front propagation is observed experimentally in suspensions of iron particles burning in oxygen-xenon mixtures.

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Propagating diffusion fronts in reactive, heterogeneous media consisting of two spatially separated phases are common in many fields, such as chemical kinetics, combustion, biology, etc. [1]. The reaction in such systems is localized within or at the phase boundaries; thus, the source term in the governing reaction-diffusion equation is not a continuous spatial function, i.e., it is discrete. Often, the approach used for modeling propagating fronts in such discrete systems is to average (homogenize) the source term with a spatially continuous function. However, a homogeneous representation of the sources is justified only if the two characteristic scales of the propagating diffusion front, the width of the reaction zone $l_R = vt_R$, and the width of the diffusion zone $l_D = D/v$, are much larger than the scale of the system heterogeneity l (here v is the front speed, t_R is the characteristic time of reaction, and D is the active component diffusivity) [2,3]. Because the front length scales are functions of the front speed, they cannot be estimated *a priori* from a solution that uses a yet unjustified source homogenization procedure [4].

To investigate the validity of the homogenization approach to model the diffusion front in a system with heterogeneous sources, we will obtain in this Brief Report an exact solution for the front speed without spatial averaging of sources and will compare it to a known solution derived from a mean-field theory approach. We will show that, in a system with regularly distributed sources, discreteness results in a front propagation speed v that is independent of the characteristic reaction time t_R. Moreover, we will show that discreteness leads to a propagation limit with nonzero front speed in a regular distribution of sources and that the front can propagate beyond this limit in a system with randomly distributed sources only through fluctuations in the local concentration. Finally, we will show experimentally that the continuous and the discrete regimes using the same reactive system can be achieved by changing the diffusivity of the active component.

In our analysis, we will consider the simplest possible discrete system where the sources of the diffusing component are points embedded in an inert continuum. We will also assume the source term to be a stepwise function in time: the source turns on when the diffusing component reaches some prescribed threshold T_i and then releases the active component with constant rate for a prescribed period of time t_R. In spite of its simplicity, such a discrete source model has a physical analog: a flame in suspensions of a nonvolatile solid fuel in a gaseous oxidizer [2], in which case the active component released by the particles is heat. Indeed, due to the density difference between the solid fuel and gas of more than three orders of magnitude, the distance between fuel particles in a combustible suspension is much larger than their diameter, such that the particles can be approximated as point sources. The ignition temperature T_i of particles is, in the first approximation, independent of flame speed. Furthermore, the reaction time t_R after ignition is controlled by the O_2 diffusion toward the particle surface [5], and, as a first approximation, the diffusivity can be assumed to be independent of the temperature. Using terminology common to combustion, the three dimensionless parameters characterizing the problem can be identified as the ignition temperature $\theta_i = c_p \rho T_i/(QB)$ (Q is the heat release per unit of fuel, B is the fuel concentration, c_p is the mixture specific heat, and ρ is the density of the media), the front speed $\eta = vl/D$ (v is flame speed, l is the distance between sources, and D is the thermal diffusivity of the media), and the combustion time $\tau_c = t_R D/l^2$.

The temperature field θ is governed by the reaction-diffusion equation with a source term

$$\frac{\partial \theta}{\partial t} = \nabla^2 \theta + F, \tag{1}$$

where for spatially continuous sources (i.e., space-averaged representation), the term F is defined as

$$F = \begin{cases} 0 & \text{if } x < 0 \text{ or } x > \eta\tau_c, \\ \frac{1}{\tau_c} & \text{if } 0 \leqslant x \leqslant \eta\tau_c, \end{cases} \tag{2}$$

where this source term is formulated in a reference frame attached to a steadily propagating front with the ignition surface located at $x = 0$. The expression defining η in a continuum with a stepwise reaction rate is obtained by matching the boundary conditions θ and $d\theta/dx$ at $x = 0$ [2] and can be written using dimensionless parameters as follows:

$$\theta_i = \frac{1 - e^{-\eta^2 \tau_c}}{\eta^2 \tau_c}. \tag{3}$$

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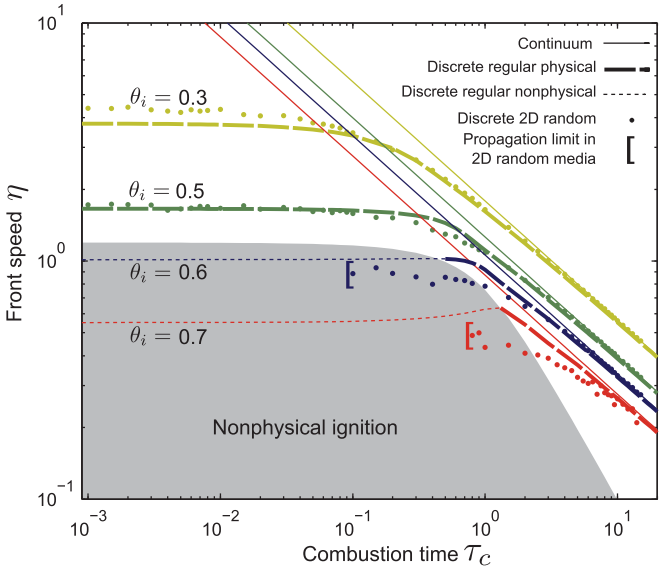

FIG. 1. (Color online) Dependence of the front speed η on the combustion time τ_c in a system with discrete heat sources and different ignition temperatures θ_i. The solid line is a continuum solution in which the sources have been homogenized with the inert media. The dashed line is the solution for regularly spaced sources. Points represent the average front speed measured from multiple simulations with randomly distributed sources. The shaded area represents the region where propagation is no longer possible in a system of regularly distributed sources.

Equation (3) contains only one combination of dimensionless parameters $\eta^2\tau_c$ that does not depend on l because the heat source term has been spatially homogenized. The dependence of η on τ_c is plotted in Fig. 1 as thin solid lines. By linearizing Eq. (3), it can be shown that η is proportional to $1/\sqrt{\tau_c}$. In other words, the continuum model predicts that η tends to infinity as $\tau_c \to 0$. This condition reflects the fact that, in the continuum model, the propagation of the diffusion front is controlled by the reaction rate of the sources.

To obtain an expression for the front speed in the same system without invoking source averaging, the source term F is expressed in the stationary, lab-fixed reference frame as an explicit function of the coordinates of the kth sources $\mathbf{x}_k$ and its ignition time τ_k:

$$F = \begin{cases} \sum_{k=1}^{N} g(\mathbf{x},\mathbf{x}_k)\delta(\tau-\tau_k) & \text{if } \tau_c = 0, \\ \sum_{k=1}^{N} \frac{1}{\tau_c} g(\mathbf{x},\mathbf{x}_k)\Theta(\tau-\tau_k)\Theta(\tau-\tau_k-\tau_c) & \text{if } \tau_c > 0, \end{cases} \tag{4}$$

where Θ is the Heaviside function, accounting for sources that are still burning at the moment of ignition, N is the number of ignited sources, and the function g is equal to 1 when $\mathbf{x} = \mathbf{x}_k$ or 0 otherwise. We initially assume that the distribution of sources forms a three-dimensional, cubic lattice, that the front propagates in the x direction from left to right, and all the sources in a z-y plane ignite simultaneously, i.e., the front is flat. Due to linearity of the simplified heat diffusion equation, the temperature of a plane of sources θ_s just about to be ignited ($m = 0$) can be found by linear superimposition of the contributions from all reacted and still reacting sources on the left side of the domain:

$$\theta_s = \sum_{m=1}^{\infty}\sum_{n=-\infty}^{\infty}\sum_{p=-\infty}^{\infty} \Delta\theta_{mnp}. \tag{5}$$

When $\tau_c \ll 1$, all sources on the left side of the igniting plane are already reacted at the moment of ignition. In this case, reacted sources can be approximated by δ functions not only in space but also in time and $\Delta\theta_{mnp}$ is the Green's function for an individual source:

$$\Delta\theta_{mnp} = \frac{1}{(4\pi m\Delta\tau)^{3/2}} \exp\left(-\frac{(m^2+n^2+p^2)}{4m\Delta\tau}\right). \tag{6}$$

We now assume that consequent planes of sources are ignited at regular time intervals $\Delta\tau$ so the time elapsed from the ignition of the mth row can be written as $m\Delta\tau$ [2,6]. By combining Eqs. (5) and (6), η can be expressed for $\tau_c = 0$ as

$$\theta_i = \frac{1}{(4\pi)^{3/2}} \sum_{m=1}^{\infty}\sum_{n=-\infty}^{\infty}\sum_{p=-\infty}^{\infty} \left(\frac{\eta}{m}\right)^{3/2} \times \exp\left(-\frac{(m^2+n^2+p^2)\eta}{4m}\right). \tag{7}$$

For $\tau_c \geqslant 0$, Eq. (7) can be written in the general form using the integral over time of Green's functions:

$$\theta_i = \frac{1}{(4\pi\tau_c)^{3/2}} \sum_{m=1}^{\infty}\sum_{n=-\infty}^{\infty}\sum_{p=-\infty}^{\infty} \int_{(\frac{m}{\eta}-\tau_c)\Theta(\frac{m}{\eta}-\tau_c)}^{\frac{m}{\eta}} \tau^{-3/2} \exp\left(-\frac{m^2+n^2+p^2}{4\tau}\right) d\tau, \tag{8}$$

where τ is an integration variable. Unlike the continuum approximation given by Eq. (3), the front speed defined by Eqs. (7) and (8) explicitly depends on the structure of the media, i.e., on the intersource spacing l. For the limiting case of $\tau_c = 0$ given by Eq. (7), it is inversely proportional to l, but the dependence diminishes with increasing combustion time τ_c. Therefore, τ_c is the measure of the system's departure from the continuum case: the system demonstrates discrete properties when $\tau_c \ll 1$ and becomes a continuum when $\tau_c \gg 1$. The identification of the discreteness parameter τ_c now provides a quantitative tool that determines when spatial averaging is a valid procedure [2,7]. Just as spatial averaging may not be justified in certain systems, the existence of heterogeneities in the media through which the reactive front propagates is not sufficient grounds to reject the use of spatial averaging. Only by examining the discreteness parameter τ_c can the appropriateness of spatial averaging be evaluated.

The front speed η predicted by Eq. (8) is plotted in Fig. 1 as a thick dashed curve. For values of $\tau_c \gg 1$, the continuum solution with continuous heat release [i.e., Eq. (1)] merges with the discrete source model. For $\tau_c \ll 1$, however, the front speed obtained by the discrete model becomes insensitive to τ_c and departs from the continuum solution. This reflects the fact that, as $\tau_c \to 0$, the propagation of the front is dictated by the diffusion of heat across the intersource spacing, a feature that is absent in the continuum solution given by Eq. (1). Whereas

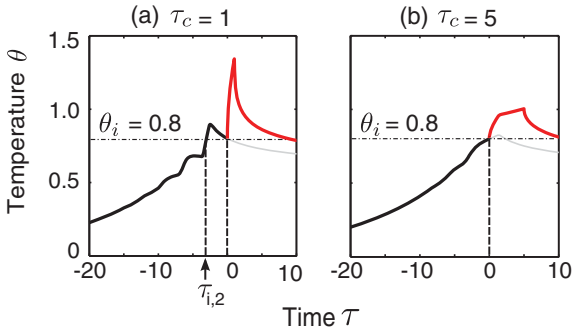

FIG. 2. (Color online) Time-temperature histories of a source igniting under physically (a) invalid and (b) valid conditions. The time $\tau = 0$ corresponds to the ignition of the source and the thin gray line originating at $\tau = 0$ corresponds to θ if ignition is omitted. In (a), the first intersection between θ and θ_i is denoted as $\tau_{i,2}$.

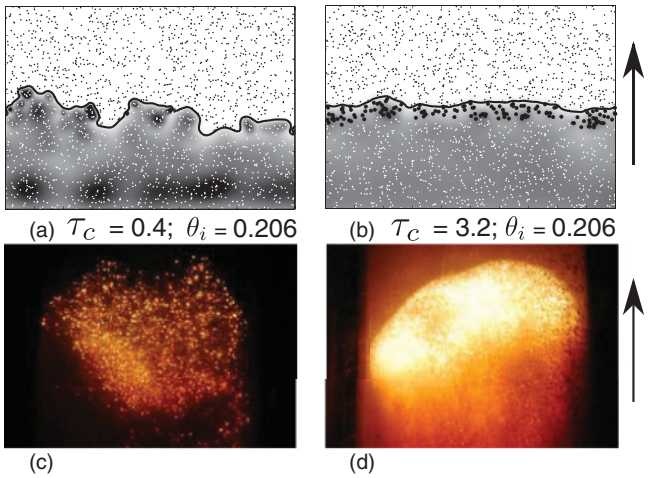

FIG. 3. (Color online) Two-dimensional numerical simulations and photographs of the flame in iron suspensions in Xe-O_2 [(a) and (c)] and He-O_2 [(b) and (d)] mixtures. The upward arrows indicate the propagation direction.

the continuum model fails when $\tau_c \to 0$, a finite front speed is predicted by Eq. (8) because the discrete model accounts for the intersource heat diffusion.

The discrete source solution with regular spacing exhibits nonphysical behavior for $\theta_i > \theta_{i,\mathrm{cr}}$, where $\theta_{i,\mathrm{cr}} \approx 0.568$. Examination of the time-temperature history of a source igniting under nonphysical conditions, as shown in Fig. 2(a), indicates that Eqs. (7) and (8) demand that the sources ignite as their temperature is *decreasing*, having previously crossed the ignition temperature θ_i without igniting [3]. The time-temperature history of a source igniting under physically valid conditions is shown in Fig. 2(b). The nonphysical branch of Eq. (8) is plotted in Fig. 1 as a thin dashed line for $\theta_i = 0.6$ and 0.7, where the front speed η decreases with a decreasing combustion time τ_c (a result that goes against physical intuition). Moreover, the shaded region in Fig. 1 represents the η-τ_c parameter space, where ignition will always occur under nonphysical conditions. If the condition of a fixed delay between sources is relaxed, and the sources are allowed to ignite upon first encountering the ignition temperature, then the sequence of ignition events is disrupted and the front promptly quenches in this nonphysical regime. Thus, this condition is associated with a propagation limit deriving from the discrete nature of the media. This limit, which occurs at finite front speed and in the absence of heat losses, is unique in combustion theory [3,8]. The solution shown here for a three-dimensional system of regularly spaced sources can also be shown to apply to a one-dimensional system of equally spaced planar sources and a two-dimensional regular array of line sources if we note that the summation in the j and k components in Eqs. (7) and (8) can be approximated by integrations over planar and line sources, respectively. Thus the propagation limit and front speed behavior found here apply equally to systems with regularly spaced sources for any system dimensionality.

To explore the effect of randomizing the spatial positioning of the sources (in contrast to the solution with the regular lattice presented previously), simulations in systems with randomly distributed point sources were investigated by the method of superimposing Green's function of individual sources. Computer simulations were performed in two-dimensional (2D) square domains containing 6400 sources. While numerical simulations were required to examine a statistically significant number of randomly generated systems with a large number of sources, the method of solution remained based on the superposition of Green's functions and no finite difference or other numerical approximations were introduced. Periodic boundary conditions were applied to the side boundaries by placing images of reacting sources outside the domain, while the front and back boundaries remained free. The initiation of the propagating front was performed by a forced ignition of a layer of sources. Snapshots of the resulting propagating fronts are shown in Figs. 3(a) and 3(b) for the discrete and continuous regimes, respectively. Significant roughening of the front can be observed for the discrete regime of propagation. The front speed η was obtained by linearly fitting the x positions and the corresponding ignition times of reacting sources. The average front speeds (plotted as points in Fig. 1) were obtained from multiple simulations performed with identical parameters but different random spatial distributions of the sources. The agreement between Eq. (8) and the front speed obtained from the simulations performed with random distributions is quite good. The front is capable of propagating successfully through sources randomly distributed under conditions for which propagation with regularly spaced sources is not possible, due to the quenching phenomenon discussed above. Propagation beyond the limit associated with a regular distribution of sources is only possible through the exploitation by the front of concentration fluctuations inherent to randomly distributed sources.

Both continuous and discrete propagation regimes were realized experimentally in suspensions of iron dust particles in a gaseous oxidizer. At moderate O_2 concentrations, iron particles in suspensions react completely heterogeneously without any gaseous products matching the model assumption of point sources. By replacing N_2 in air first by He and then by Xe, the thermal diffusivity can be drastically altered, changing the value of the dimensionless combustion time τ_c by almost an order of magnitude from 3.2 (predominantly continuum regime, $\tau_c > 1$) to 0.4 (predominantly discrete

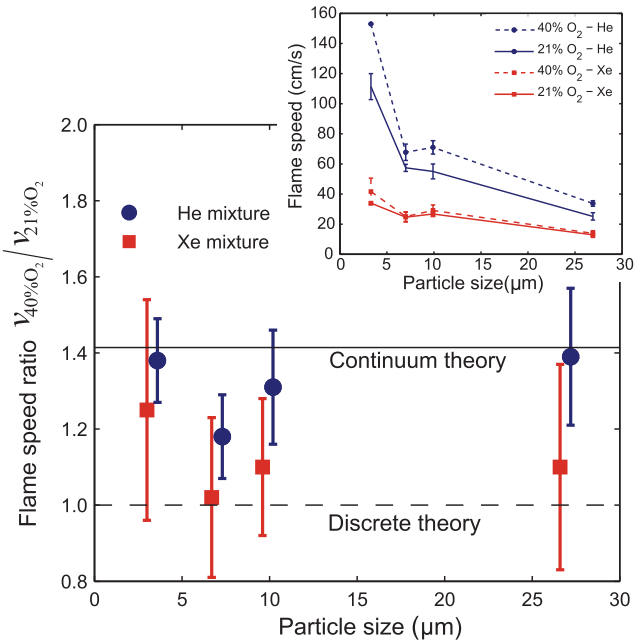

FIG. 4. (Color online) Ratio of flame speeds between two different O_2 concentrations in He and Xe mixtures. The inset shows experimental measurements of the flame speed in iron suspensions in 21% and 40% O_2 in He and Xe.

regime, $\tau_c < 1$) without changing the reaction chemistry or adiabatic flame temperature. These values of τ_c were used in the simulations shown in Figs. 3(a) and 3(b). The experimental procedure consisted of producing a fuel-rich suspension of iron particulates inside a glass tube and igniting the mixture at the open end of the tube. A laminar flame was initiated and a high-speed camera recorded the propagation of the flame along the tube. The experiments were performed in reduced gravity created inside an aircraft flying a parabolic trajectory at gravity levels below 0.05 g, which eliminated particle settling and natural convection, allowing observation of low speed flames ($v \approx 5$ cm/s) characteristic for 25-μm iron particles in Xe-O_2 mixtures. The particle-to-particle nature of the discrete regime for iron suspensions in Xe-O_2 was manifested by the rough front structure, whereas the flame front in He-O_2 mixtures was smooth, characteristic of the continuous regime (see Fig. 3). A similar difference in front appearance for He and Xe mixtures was demonstrated by the numerical model, as shown in Fig. 3.

Besides the flame appearance, the insensitivity of the flame speed to particle combustion time corroborates the discrete propagation regime in Xe-O_2 mixtures, as shown in Fig. 4. Flames in Xe mixtures are less sensitive to changes on the O_2 concentration C_{O_2}, or the particle reaction time ($t_R \sim \frac{1}{C_{O_2}}$), suggesting that the propagation mechanism is limited by particle-to-particle heat diffusion, characteristic of discrete flames, whereas flames in He mixtures varied with the O_2 concentration in better agreement with the continuum theory ($v \sim \frac{1}{\sqrt{t_R}}$).

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