Response of the Particle Ignition Dynamics on External Periodic Temperature Perturbation: Supercritical and Critical Regimes

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Key words: ignition, dynamic regimes

Introduction

An ignition of a single reacting heterogeneously metal particle in a surrounding gas medium is usually described as transition from kinetically controlled regime of reaction to diffusion controlled one [1]. As a result of nonlinear on temperature Arrhenius reaction rate, diffusion of oxidizer from surrounding gas medium to the reacting particle and the thermal exchange between the surrounding and the particle, a qualitative description of the particle temperature dynamics by bistable potential, $V\left(T_{particle}\right)$, is extensively used [2]. Each minimum in this potential is due to the reaction which may be either kinetically controlled or diffusion controlled. Investigating the temperature dynamics at low temperatures, when the reaction is kinetically controlled $(k/\beta \ll 1)$, potential can be reduced to the onestable potential which is characterized by having two equilibrium points, one stable and one unstable (for subcritical state). Ignition takes place when the potential loses its stable state at a critical temperature of surrounding (critical state). Unstable value of the temperature of the particle at that critical condition is well known as a critical one.

The situation described above could be fully complete if the temperature of surrounding medium periodically changes with time. The temperature dynamics of the particle becomes a complex function. Close to the critical temperature of particle the temperature dynamics is sensitive to the temperature perturbation in the surrounding. In reality, even for the system in the supercritical state (reduced potential has no equilibrium points) external fluctuations complicate dynamics of ignition and lead to a noticeable variation of the ignition delay time.

The main objective of this article is to analyse the dynamics of the ignition process of the particle in dynamic temperature conditions by moments of time when the particle temperature passes through a fixed value. The response of temperature dynamics to an imposed external periodic perturbation is defined as strong when it can temporarily and some times periodically decrease the temperature of particle during ignition.

Model and two representations of dynamics

Periodic external temperature conditions for a particle are set as a harmonic function, $T_\infty = \overline{T}_\infty + A\sin\left(\omega t + \phi_0\right)$, where $\overline{T}_\infty$ is the time average value, A is the amplitude, ω denotes the frequency and ϕ_0 is the initial phase. Consider a Newtonian thermal exchange at low-frequency change of temperature in the surrounding, when thermal waves in the particle are not generated, the temperature dynamics of that particle will be described by the conservation energy equation written in dimensionless form here as

$$\frac{d\theta}{d\tau} = \psi_c e^{\theta} - \theta + \lambda + \varepsilon \sin\left(\Omega\tau + \phi_0\right) \tag{1}$$

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with initial condition $\theta\left(\tau=0\right)=\theta_0$.

Here θ is the dimensionless temperature of the particle, ε is the dimensionless amplitude of the surrounding temperature perturbation. λ denotes dimensionless difference between the critical temperature of surrounding at static regime, $\varepsilon=0$, and average value of the surrounding temperature at the dynamic regime, $\varepsilon\neq 0$. ψ_c is the Semenov parameter which has the critical value, e^{-1}, corresponding to the static regime of reaction. Time and frequency are represented in the thermal relaxation time scale: $\tau=t/\tau_r$; $\Omega=\omega\tau_r$. Since the mean magnitude of the perturbation may have different values then there are three dynamical regimes of ignition in reality. Namely: subcritical, $\lambda<0$, critical, $\lambda=0$, and supercritical, $\lambda>0$.

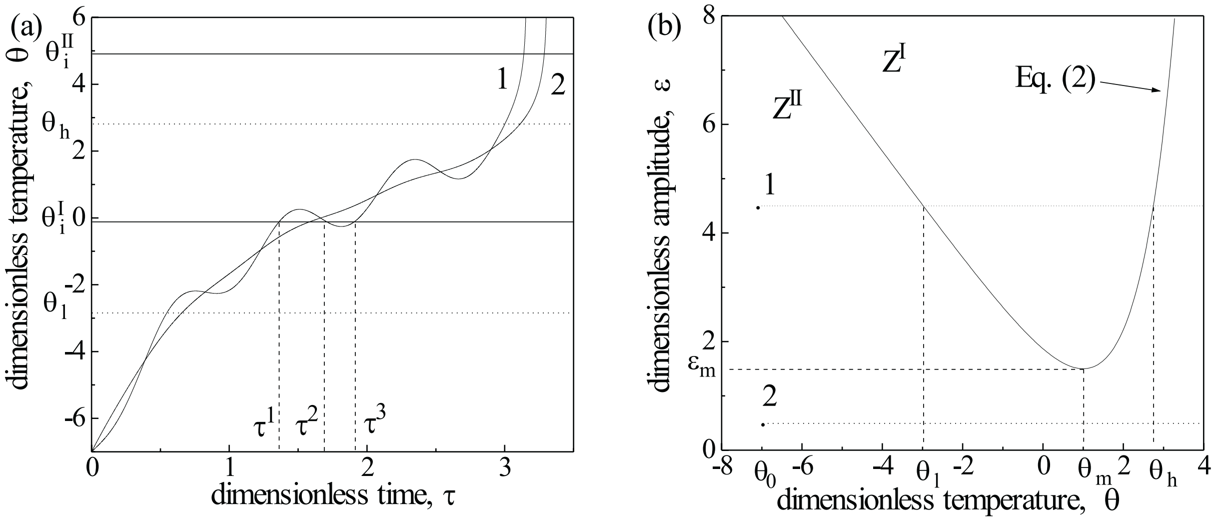

Figure 1. (a) Own representation of dynamics and (b) parametrical representation of dynamics for $\varepsilon=4.5$ (1), $\varepsilon=0.5$ (2). Z^I is the region of strong response to perturbation. Z^{II} is the region of weak response. The parameters are: $\theta_0=-7$, $\lambda=1.5$, $\Omega=1.2$, $\phi_0=-\pi/2$. θ_l and θ_h are the temperature limits for strong response.

Examples of the supercritical temperature dynamics of the particle are shown in Fig. 1(a) for two values of amplitude perturbation, ε. As it can be seen from this figure, response of the particle temperature to the external perturbation is strong within some temperature interval, $\theta_l<\theta<\theta_h$, only for the large amplitude case. The limiting values of this interval, θ_l, θ_h, depend on the parameters of the perturbation, ε, λ, and may be estimated analytically. Equating the right hand side of Eq. (1) with zero and minimizing it on the phase, we can found the next relation

$$\varepsilon=\psi_c e^{\theta}-\theta+\lambda \tag{2}$$

which is shown in Fig. 1(b). This curve divides $\theta-\varepsilon$ plane into two regions: a region of strong response to the external perturbation, Z^I, and a region of weak response, Z^{II}. The minimum of this function is located at the point $(\theta_m,\varepsilon_m)=(1,\lambda)$. It can be easily verified that Eq. (2) has two solutions, θ_l and θ_h, when the amplitude of the external perturbation is larger than the value of the minimum of this function, $\varepsilon>\varepsilon_m$, one solution, $\theta_l=\theta_h=\theta_m$, when $\varepsilon=\varepsilon_m$, and no solutions when $\varepsilon<\varepsilon_m$. Using the parametrical dependency calculated by Eq. (2) and represented in Fig. 1(b), the temperature dynamics can be described qualitatively by a point moving from the coordinates (θ_0, ε) to (∞, ε). If during ignition the temperature of the particle passes through region Z^I, $d\theta/d\tau$ changes its sign and therefore temperature dynamics is similar to curve 1 between θ_l and θ_h in Fig. 1(a). After some time, when the temperature of the particle becomes equal to θ_h, the chemical reaction controls the ignition process and the periodic temperature perturbation plays minor role. For $\varepsilon<\varepsilon_m$, the temperature response to external perturbation is not strong, and the particle temperature increases continuously with time. This case is well known in literature as a small amplitude limit (see, for example, Ref. [4] and references therein).

For the critical regime of ignition ($\lambda=0$), a strong response is realized for all values of the amplitude of the external perturbation. Applying perturbation with negligible small amplitude, we can estimate the temperature interval where strong response occurrs, $\theta_h-\theta_l=2\sqrt{2\varepsilon}$.

Phase diagram for the passage time

The approach described in the previous section does not show all the information about the time history of the particle temperature. It gives only the conditions and the temperature interval in which the response of temperature dynamics to the external perturbation is strong. It is easy to understand that the evolution of the particle temperature depends not only on the amplitude and the time average value of the perturbation, ε, λ, but also depends on the phase parameters, Ω and ϕ_0. To investigate the dynamics at different frequencies, we will be interested in moments of time, when the temperature of the particle passes the value θ_i, which can lie within Z^I or Z^{II} region. As it follows from Fig. 1(a), for the large amplitude case, $\varepsilon > \varepsilon_m$ the particle temperature crosses the value θ_i^I at several moments of time: τ_i^1, τ_i^2, τ_i^3. However, the temperature of the particle reaches the temperature θ_i^{II} only once. Moreover, at the moment τ_i^2 we have $d\theta/d\tau < 0$, but for odd values (τ_i^1 and τ_i^3) we can see that $d\theta/d\tau > 0$. The values of these moments and the number of intersections are determined by the frequency of the perturbation. For small amplitudes, $\varepsilon < \varepsilon_m$ one point of intersection of the particle temperature with θ_i^I occurs as well as with θ_i^{II}.

Let $F(\tau, \Omega; \theta, \theta_0, \lambda, \varepsilon, \phi_0) \equiv 0$ be the implicit solution of Eq. (1) written at the moment τ for the particle temperature θ. Fixing the value of θ, $\theta = \theta_i$, all the values of τ_i (τ_i^1, τ_i^2, τ_i^3...) that satisfy the implicit solution of Eq. (1) can be found numerically. Assuming that $F(..;...)$ is a continuously differentiable function at each point, then, in accordance with the implicit function theorem [5], the derivative of $\tau_i(\Omega)$ with respect to the frequency is

$$\frac{d\tau_i}{d\Omega} \equiv \frac{d\theta_i/d\Omega}{d\theta_i/d\tau_i} = -\frac{\partial F/\partial\Omega}{\partial F/\partial\tau_i} \tag{3}$$

where the implicit solution, $F(..;...)$ is taken in the point τ_i, θ_i. As it follows from the last equation, $d\tau_i/d\Omega$ is equal to infinity when $d\theta_i/d\tau_i$ (or $\partial F/\partial\tau_i) = 0$. This condition is only possible for $(\theta_i, \varepsilon) \in Z^I$ and (θ_i, ε) satisfying Eq. (2).

For $(\theta_i, \varepsilon) \in Z^I$ there are two solutions for the phase expressed as

$$\left.\begin{array}{lcl} (\Omega\tau_i)_n^+ + \phi_0 & = & +\arcsin\left(-(\psi_c e^{\theta_i} - \theta_i + \lambda)/\varepsilon\right) + 2\pi n, \\ (\Omega\tau_i)_n^- + \phi_0 & = & -\arcsin\left(-(\psi_c e^{\theta_i} - \theta_i + \lambda)/\varepsilon\right) + 2\pi(n + 1/2). \end{array}\right\} \tag{4}$$

Substituting θ_i and ε that satisfy Eq. (2) into Eqs. (4) yields

$$(\Omega\tau_i)_n + \phi_0 = \frac{3\pi}{2} + 2\pi n, \tag{5}$$

where $\Omega\tau_i$ must be greater than zero and $n = 0, 1, 2, ...$

For $(\theta_i, \varepsilon) \in Z^{II}$ there are no frequencies possible at which the derivative $d\tau_i/d\Omega = \infty$. In fact, the phase in Eq. (5) is the bifurcation point for the phases described by Eqs. (4). Examples of the solution, $\tau_i(\Omega)$, of $F(\tau_i, \Omega; ...) \equiv 0$ for these three different cases are shown in Fig. 2(a), see next page. It is easy to see from this figure that the solution, τ_i, strongly depends on the frequency of the perturbation.

The results obtained above are in good agreement with the experimental data represented in Ref. [3]. As it is shown in Fig. 2(b) by solid squares, discontinuity of the experimental dependency of τ_i against Ω are observed. The experimental parameters taken is used for simulations and have the values: $\lambda = 1.5$, $\varepsilon = 11.99$ and $\theta_i = -0.52$. It is easy to check, (θ_i, ε) corresponds to region where the response to perturbation is strong, $(\theta_i, \varepsilon) \in Z^I$, which results in the discontinuity of the experimental dependency of the ignition delay time on frequency. According to the numerical simulations and Eqs. (4), strong response leads to the multiplicity of $\tau_i(\Omega)$ in frequency bands, width of which greatly depends on the kinetic constants of the chemical reaction and inertia properties of the particle.

Actually, casual temperature fluctuations of the surrounding medium exist during experiments and make a contribution to the ignition process. They both change the frequency band width of the multiplicity when $(\theta_i, \varepsilon) \in Z^I$ and result in an induction of the multiplicity of the ignition delay time when (θ_i, ε) lies in the region Z^{II} near the curve given by Eq. (2). In addition, using an analytical approximate solution of Eq. (1) written for the low temperature stage of ignition, $\theta < 0$ (when reaction rate is small but not negligible), the envelope functions for solution and the experimental data are obtained. For the more simple case – inertia heating, the lower envelope function is plotted as a smooth dependency of time on frequency, $\tau^-(\Omega)$, in Fig. 2(b).

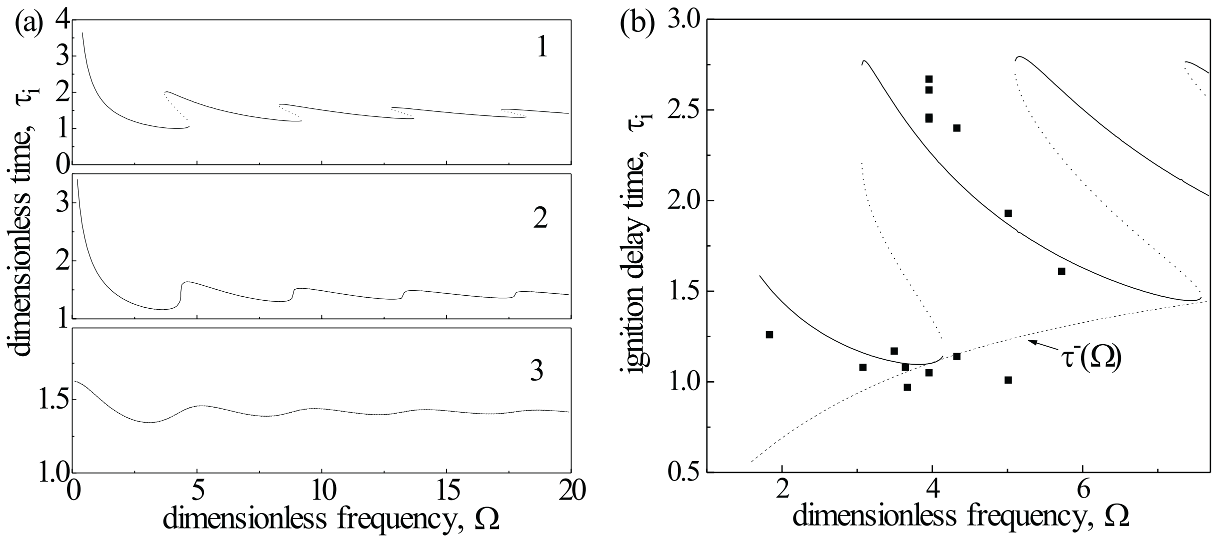

Figure 2. (a) Solutions for implicit function $F(..;..) \equiv 0$ as dependency of the time on frequency. $(\theta_i, \varepsilon) \in Z^I$ (1), (θ_i, ε) satisfies Eq. (2) (curve 2), $(\theta_i, \varepsilon) \in Z^{II}$ (3). (b) Comparison of the numerical simulations with experimental data. Dotted curves correspond to τ_i at which $d\theta/d\tau < 0$, solid curves correspond to τ_i at which $d\theta/d\tau > 0$. Solid squares – experimental data. Smooth dependency $\tau^-(\Omega)$ is the lower envelope function for solution $\tau_i(\Omega)$ of implicit function $F(\tau_i, \Omega; ..) \equiv 0$ (case of inertia heating).

Conclusion

In this article we have studied the ignition of the single particle in dynamic temperature conditions. Using passage time concept for supercritical and critical regimes of ignition we can conclude that

- Temperature limits for strong response of the temperature dynamics of the particle to the external periodic perturbation are fixed by parameters of perturbation, λ and ε.
- The ignition delay time dependency on frequency, $\tau_i(\Omega)$, is also controlled by the temperature of ignition, θ_i, and may be both smooth and discontinued function.
- Making use of the analytical approximate solution of Eq. (1), smooth envelope functions, $\tau^-(\Omega)$ and $\tau^+(\Omega)$, between which the particle temperature passes through the temperature of ignition, θ_i, are estimated.

Acknowledgment *The author gratefully acknowledges Dr. Eugene N. Kondratyev for helpful discussions and notes, Pavel L. Kachka for hardware support and Dr. H. Mousa for critical remarks to the text.*

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