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Effects of disordered microstructure and heat release on propagation of combustion front

Tarun Bharath Naine1 and Manoj Kumar Gundawar1*

Abstract: Numerical experiments for diagnosis of combustion of actual heterogeneous systems are performed on a one-dimensional chain. The internal microstructure of actual heterogeneous systems is apriori unknown, various distributions like uniform, beta, and normal have been considered for distributing neighboring reaction cells. Two cases, for the nature of distribution of heat release of reaction cells are taken into account, one with identical heat release and the other with disordered heat release. Role of different random distributions in describing heterogeneous combustion process is established in present paper. Particularly, the normal distribution of arranging neighboring reaction cells has been found to be powerful methodology in explaining the combustion process of an actual heterogeneous system at higher ignition temperatures for both cases of distributing heat release. Validation of the developed model with the experimental data of combustion of the CMDB propellants, gasless Ti + xSi system, and different thermite mixtures is performed. Our results show that the experimental burning rates at higher ignition temperatures (ε > 0.32) of the heterogeneous system are better reproduced theoretically with the present model. We have also shown that different combustion limits for different thermite systems are the consequences of disordered heat release. Experimental data for thermite systems that have lower inflammability limits are

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Manoj Kumar Gundawar is working as an assistant professor at university of Hyderabad, Hyderabad, India. His primary research interests are experimental work on laser-induced breakdown spectroscopy. His group is also interested in the modeling aspects of the heterogeneous combustion, particularly, randomizing the differences in positions of the combustion cells. The most important discovery from his group shows that the random structure of the microheterogeneous system plays a crucial role in the dynamical and statistical behavior of the system, a direct consequence of the nonlinear interaction of the structure of the system with the thermal wave. Present manuscript further extends the idea of random distributions to other distributions which is apriori unknown parameter. The experimental burn rates can be associated with one of the distributions by matching them with the numerical burn rates. This kind of strategy enables the predication of the burn rates for future studies and different amounts of mixtures.

PUBLIC INTEREST STATEMENT

Modeling and simulation of heterogeneous combustion can overcome the difficulties of experimental investigations, has inherent safety and wide operational features. The actual heterogeneous mixtures while packing involves improper mixing, percentage of diluters, and binding agents. These situations have their own impact on internal microstructure, modes of combustion front, and combustion limit. The present manuscript explains the effect of these situations on combustion process by considering various distributions like uniform, beta, and normal for distributing neighboring reaction cells. Two cases, for the nature of distribution of heat release of reaction cells are taken into account, one with identical and the other with disordered heat release. Our results show that the normal distribution of arranging neighboring reaction cells can be the powerful methodology in explaining the combustion process of an actual heterogeneous system. We have also shown that different combustion limits for different thermite systems are the consequences of disordered heat release.
analyzed in the view of disordered heat releases of cells. The model developed in the view of disordered heat releases reproduces the experimental burn rates and experimental combustion limit.

**Subjects:** Computational Physics; Physical Sciences; Physics; Statistical Physics; Theoretical Physics

**Keywords:** heterogeneous combustion; heat release; ignition temperature; burn rate; normal distribution

1. Introduction

Combustion of powdered mixtures (Beckstead & McCarty, 1982; Bharath, Rashkovski, Tewari, & Gundawar, 2013; Denisyuk, shabalina, & Shepelev, 1998; Dvoryankin, Strunina, & Merzhanov, 1985; Kubota, 1978, 2002; Kubota & Okuhara, 1989; Kulkarni & Sharma, 1998; Rashkovskii, 2005; Rashkovskiy, Kumar, & Tewari, 2010; Rogachev & Baras, 2009) and burning of solid propellants, liquid droplet combustion, spray combustion, combustion of coal, and engines are the examples of heterogeneous combustion. CMDB (Kulkarni & Sharma, 1998) propellants (DNC + CL, DNC + CL + AP), also known as composite modified double base, are heterogeneous mixtures of solid components and small particulate oxidizers held together with a rubbery material referred to as binders (AP). Such systems with addition of binding agent (AP) become high-performing propellants and are realized with minimum release of gasses. The combustion efficiency increases with an increase in the binding agents. Powdered systems with gasless combustion are as well realized as the simplest form of heterogeneous system. Heterogeneous combustion of the mixtures such as Ti + xSi (Rogachev & Baras, 2009), Ni + xAl commonly involves in the formation of gasless combustion products. Chemical reaction, ignition, and burning away of such mixtures occur in solid state, and result in solid combustion products. Furthermore, combustion of thermite systems such as Fe₂O₃ + Al, Fe₂O₃ + Ti, Cr₂O₃ + Zr (Dvoryankin et al., 1985) are apparently linked with the formation of gaseous combustion products. Fortunately, for the above-mentioned thermite systems, a gasless combustion can still be obtained by diluting the respective thermite mixtures with their respective end product dilutent (Al₂O₃, TiO₂, ZrO₂). Self-sustained combustion for such systems is realized within the limits of quantity of inert dilutent. Self-sustained high-temperature synthesis which is used for the synthesis of materials is an important class of heterogeneous combustion.

The behavior of combustion front in such heterogeneous mixtures, with and without addition of binding agents, and at different stoichiometric mixtures of reactants, also at limits of combustion for different reactants, is of fundamental interest in explaining the combustion process through most accurate model. Such models can account for the effects of practical synthesis of materials, percentage of binding agent, stoichiometries, and inert dilutent on the combustion process. Despite extensive investigations (Bharath et al., 2013; Rashkovskii, 2005; Rashkovskiy et al., 2010), different combustion limits for the thermite mixtures (Fe₂O₃ + Al, Fe₂O₃ + Ti, ...) and modes of combustion front in heterogeneous mixtures are unexplained because of the internal microstructure of actual heterogeneous mixtures is apriori unknown and absence of accurate models in agreement with experimental data. The role of internal microstructure and consumption of reactant (distribution of heat release) in combustion process became apparent after numerical (Bharath et al., 2013; Rashkovskiy et al., 2010) and experimental studies using high-speed micro video camera. Such studies have shown that the self-propagating combustion front becomes complex waves at macro scales. The HSMV camera revealed (Willcox, Brewster, Tong, & Stewart, 2007; Favier, 2004; Gardner, Romme, & Turner, 1999; Hwang, Mukasyan, & Varma, 1998; Kerstein, 1987; Mukasyan & Rogachev, 2008; Rashkovskii, 1999; Rogachev, 2003; Rogachev & Baras, 2007; Viegas, 1998) the mechanism modes of combustion which is either quasi-homogeneous or relay race. Heterogeneous combustion is complicated by microstructure and the consumption of reaction cells of the system even at stationary mode of combustion. Such complications results in micro-oscillations of combustion wave and leads to limit of combustion front propagation. Combustion limit is a situation where the combustion front doesn’t propagate further. Combustion limit is different for different reactants and the analysis for combustion limits is less
concentrated in earlier literatures (Bharath et al., 2013; Rashkovskii, 1999, 2005; Rashkovskiy et al., 2010). Such an analysis for extreme reaction condition during gasless combustion process requires new methods for tailoring the internal microstructure of the system (Candel, 2002; Goroshin, Lee, & Shoshin, 1998; Gross, 2010; Humphrey, 1971; Varma & Mukasyan, 2001). Actual heterogeneous mixtures are of three dimensional ones, the internal microstructure is automatically formed while mixing and packing and the particles in them have definite size depending on concentration of diluents. As demonstrated in Bharath et al. (2013), Rashkovskii (2005), and Rashkovskiy et al. (2010), the one-dimensional study of heterogeneous combustion process can still explain the role of different physical parameters (internal microstructure, heat release, burn rate, disordereness) if the particles are considered of point size. In Bharath et al. (2013), we have not only established and validated the relevance of one-dimensional model but also have shown that the Arrhenius macrokinetics observed during actual combustion process can be related with ignition temperature ($T_{\text{ign}}$). The one-dimensional model has purely thermal nature. However, the work shown in Bharath et al. (2013) did not concentrate on different aspects such as the role of different random distributions and randomness of heat release of reactive cells.

In this study, we concentrate on diagnosis of combustion with significance to two factors that are different and extensions to our previous work (Bharath et al., 2013) (a) different random distributions employed for positioning neighboring reaction cells (b) Randomizing the heat release by considering a distribution of heat release among all the unburnt cells. The methodology, resembles to Monte Carlo simulation, enable us to perceive the affect of minute changes in microstructure on dynamic combustion parameters such as stability of combustion front, ignition delay times, and average burn rate. Such minute aspects are controlled by our model and are illustrated with significant change in parameters such as scale and shaping parameter of distribution. The affect of randomizing heat releases on combustion process is given very little importance and almost times neglected in earlier literatures. Such situations commonly arise from improper mixing of heterogeneous mixtures, changing the diluter and percentage of diluter. Two different cases of reactant consumption are considered, in present work, by identical distribution of heat release or by randomizing the heat release. The above problem is addressed by describing the system in terms of the other random distributions (normal, beta and uniform). Particularly, the normal distribution allows for positioning the heat sources with minimum clusterization and symmetric around the mean. The novel approach shall optimistically list the priority of distributions in modeling a discrete combustion process.

Section 3 reports the numerical results obtained for developed system. The burn rate of the system including the dynamical behavior of reaction waves in such systems is reported. The Possible limits of a combustion process are studied for normal distribution. Sections 3.1–3.5 discusses the numerical results obtained for identical heat release. The heat release is carried through thermal bridge and its physical significance is explained in modeling section. The developed model is validated and compared with experimental results performed on combustion of CMDB propellants, Ti–Si system, and different thermit systems. Here we establish the most accurate distribution of neighboring reaction cells and its physical significance. Most works on combustion modeling have reported that the effect of random heat release in combustion process (Bharath et al., 2013; Rashkovskiy et al., 2010) is negligible and considered the heat release to be identical for all the cells. Section 3.6 studies the systematic effect of heat consumption of reaction cells (Gardner et al., 1999; Hwang et al., 1998; Mukasyan & Rogachev, 2008; Rogachev, 2003) on combustion process.

2. Modeling

During combustion process, the exothermic reaction is restricted to a thin zone that propagates throughout the combustible system. The vicinity of the reaction zone is referred as combustion (reaction) front. The heat transfer takes place across the entire combustor volume. Traditional explanation of gasless combustion process is based on diffusion process of reaction cell that undergoes exothermic reaction. During practical situations, sample reacting generally has cylindrical geometry. Temperature gradient along transversal direction is very little under many circumstances. Neglecting heat loss upon such situations the one-dimensional model, within the macroscopic framework, can be used for describing the
temperature profiles. One-dimensional discrete combustion model, similar to our previous works (Bharath et al., 2013; Rashkovskiy et al., 2010), comprising chain of reaction cells (point particles) distributed either in periodic or random trend is considered here. Nevertheless, in the present methodology, we not only introduce different random distributions (uniform, beta, normal) for spacing neighboring reaction cells but also randomize the consumption of reaction cells. The instabilities of heterogeneous combustion process can be predicted in the framework of macroscopic studies. The statistical study of instabilities, significant to different minute aspects of random process, allows us to identify the situation at macroscopic levels. Combustion process, at higher ignition temperatures, is of complicated in nature. Detailed work in this direction leads us with the possibility of finding the accurate model of heterogeneous combustion process at higher ignition temperatures. Propagation of combustion front is modeled by distributing immobile reaction cells on one-dimensional chain and taking into consideration of spontaneous combustion of reaction cell with heat release ($Q_i$) as soon as temperature of the active cell reaches a pre-determined ignition temperature ($T_{ign}$). Experimental data (Dvoryankin et al., 1985; Kulkarni & Sharma, 1998; Rogachev & Baras, 2009) illustrate that the chemical reaction of the mixtures begins after the melting of reactants. Ignition and burning time for active cells in such systems are always less when compared to characteristic time for heating of reaction cells. These observations during the experiments permit us to regard the combustion process of active cells in the system as spontaneous. The moment at which the threshold temperature ($T_{ign}$) is reached (melting of reactants begin) is considered as ignition time of the reaction cells. The chains of reaction cells described in the model are interconnected using a thermal bridge. Thermal bridge is described by thermal conductivity $k$, linear mass density $\rho$, and specific heat $c$. Heat transfer between reaction cells is carried through the thermal bridge and it is assumed that there are no heat losses i.e. total heat energy released by combustion of preceding particle is utilized completely for combustion of succeeding unburnt particles. The model under consideration simplifies the reaction kinetics of actual heterogeneous combustion process. The parameters of developed model such as ignition temperature and heat release (consumption of reactant) signify the characteristic features of the reactants. If we regard this model to actual combustion of heterogeneous mixtures (Dvoryankin et al., 1985; Kulkarni & Sharma, 1998; Rogachev & Baras, 2009), the sense of consumption of reactant (heat release) and ignition temperature becomes apparent with respect to combustible limits and quantity of inert diluent. The gasless combustion of heterogeneous mixtures, chemically inert by itself, undergoes self-sustained combustion after releasing a large amount of heat. Such process of combustion is achieved only after initial melting of reactants. In the present model, sufficient amount of initial thermal energy is given to the system by initiating a fraction of cells at time ($t = 0$). The problem in consideration is explained by a one-dimensional equation of thermal conductivity of point cells as (see Rashkovskiy et al., 2010):

$$T(t,x) = T_{in} + \sum_{i=1}^{k-1} \Delta T_i (t - t_i, x - x_i),$$

where $T_{in}$ is the initial temperature of the system; $t_i$ is the instant of ignition of the $i$th cell, located at $x_i$. The above equation has a solution given by Equation (1). Governing equation for a one-dimensional combustion process involving dimensionless parameters (Bharath et al., 2013; Rashkovskiy et al., 2010) is as follows:

$$2 \sqrt{\pi \varepsilon} = \sum_{k=0}^{k-1} \frac{q_j}{\sqrt{s_k - s_j}} \exp \left( -\frac{(\xi_j - \bar{x})^2}{4(s_k - s_j)} \right) \tag{1}$$

In the above, the non-dimensional parameters are: ignition time for burning cell $s = t(k/T_i^2)$, distance of unburnt particle $\xi = x/l_o$, ignition temperature $\varepsilon = \frac{T_{ign} - T_{in}}{T_{in} - T_{ad}}$ and heat release in combustion $q_o = Q/Q_o$ (here in this paper, we consider two cases of $q_o$ one with identical value at all heat sources and other with randomized), where $T_{ad} = T_{in} + \frac{Q_o}{\rho c l_o}$ is mean adiabatic temperature of the thermal bridge, $Q_o = \lim_{N \to \infty} \frac{1}{N} \sum_{i=1}^{N} Q_i$ is a mean energy release in combustion of one heat source and $l_o = \lim_{N \to \infty} \left( (x_N - x_o)/N \right)$ is a mean distance between neighbor point heat source of the system.
Equation (1) is utilized for the numerical calculation of the ignition times of all particles in the developed model (system). The model developed here is described by a single parameter \( \varepsilon \) (non-dimensional ignition temperature of active cells). Spacing of unburnt reaction cells in periodic system is done by regular spacing at a distance of unit. Random system that involves distribution of positioning unburnt cells is achieved by probability density function of various random distributions, namely uniform, normal, and beta with a mean of unity. The probability density function for different distributions is listed in Table 1. A gamma distribution with shaping parameter \( a = 0.7 \) (Bharath et al., 2013) is also considered for comparison. Total reaction cells in the model are considered as concatenation of 1,000 burnt and 2,500 unburnt cells. Figure 1 shows the probability density functions for spacing unburnt cells significant to shape and scale. The average spacing of particles is always maintained to be unity to facilitate a direct comparison of the results with a periodic system (Rashkovskii, 2005).

The combustion process for such systems (disordered spacing of reaction cells) takes place non-uniformly in oscillating mode (see Bharath et al., 2013; Rashkovskiy et al., 2010); hence average burn rate is considered. The average burn rate of combustion front in the system between cells \( i \) and \( k > i \) is equal to:

\[
\omega_{ik} = (\xi_k - \xi_i)/(s_k - s_i)
\]  

The numerical simulation of the modeled system requires high-computing speed. C language, with the facility of MPI, for simulation of combustion process is employed for executing the calculation.

| Table 1 Probability density function for different random distributions |
|-----------------------------|------------------|------------------|
| S. no. | Distribution | Probability density function | Parameters | Mean |
|-------|-------------|-------------------------------|------------|------|
| 1.    | Normal      | \( p(L) = \frac{1}{\sigma \sqrt{2\pi}} e^{-\frac{(L-\mu)^2}{2\sigma^2}} \) | \( \sigma, \mu \) | \( \mu \) |
| 2.    | Uniform     | \( p(L) = \frac{1}{b-a} \) | \( a, b \) | \( \frac{a+b}{2} \) |
| 3.    | Beta        | \( p(L) = \frac{1}{B(\alpha, \beta)} L^{\alpha-1} (1-L)^{\beta-1} \) | \( \alpha, \beta \) | \( \frac{\alpha}{\alpha+\beta} \) |

Figure 1. Probability density function for different distributions.
3. Numerical results

3.1. Identical heat release

Ignition time profiles are obtained using Equation (1) for different distributions of positioning neighboring reaction cells at different ignition temperatures τ. Note; here we consider identical distribution of heat release at all reaction cells. Figure 2 shows the ignition time profiles at different ignition temperatures of a combustible system described by different probability density functions employed for positioning neighboring reaction cells. σ = 0.05 for normal distribution and α = 0.7 for gamma distribution are considered for comparison. Ignition time profiles depend both on the ignition temperatures and minute aspects of distributions of neighboring reaction cells. The randomness (Bharath et al., 2013; Rashkovskii, 2005; Rashkovskiy et al., 2010) in microstructure of a system also has critical influence on the ignition time profiles (Rogachev & Baras, 2009). The ignition time profiles are linear for both normal distribution and beta distribution of neighboring reaction cells and are identical to the periodic system. This can be inferred from Figure 1, where it can be seen for a beta distribution about ~70 percent of the cells spacing’s are in the range from 0.8 to 1.2 and ~100 percent for the normal distribution. The particles in spite of their random spacing bear a close resemblance to a periodic system with unit spacing and hence a linear relation. Such an analysis determines the effect of internal microstructure (packing of particles) on combustion process. However, the combustion front, for gamma distribution of neighboring reaction cells with shaping parameter α = 0.7, is noticeable with more stops as the ignition temperature increases. Also a similar affect can be seen for a normal distributed system, which will be shown in next Section. Instability in combustion or reaction front stops (Bharath et al., 2013; Gardner et al., 1999; Hwang et al., 1998; Mukasyan & Rogachev, 2008; Rashkovskii, 2005; Rashkovskiy et al., 2010; Rogachev, 2003; Rogachev & Baras, 2009) is more prominent at higher ignition temperatures and higher degree of randomness in microstructure of the system. At higher ignition temperatures, though the quantity of heat energy obtained from the burnt cells is same as compared to a corresponding periodic system, the combustion front sees stops in the propagation as a result of the randomness of neighboring reaction cells. With an increase in the degree of randomness of positioning neighboring reaction cells, the obtained heat energy has to overcome the disordered microstructure of system, particularly those points on the chain where the neighbor-cell spacing is very large (Gardner et al., 1999; Hwang et al., 1998; Mukasyan & Rogachev, 2008; Rogachev, 2003). This can be quantified by observing the maximum ignition delay times of a system. The stops in combustion front are result of increased ignition delay times. Delays in ignition times are due to increase in induction periods between active cells and ignition temperature of a combustible system.

Figure 3(a) shows plot between ignition temperature and maximum ignition delay times of a system. These maximum ignition delay times are obtained from the correlation analysis between adjacent reaction cells and their ignition delay times.
From the correlation analysis, it is observed that the variance of the ignition delay times increases with an increase in the degree of randomness in microstructure and ignition temperature. From Figure 3(a), it is observed that maximum delay in ignition times of a system is proportional to degree of randomness in microstructure and ignition temperature of the system. The maximum delay in ignition times at particular ignition temperature shows the trend periodic < normal (σ = 0.05) < beta < uniform < gamma (α = 0.7). The maximum delay in ignition times also plays the critical role in effecting the stability of combustion front and thus burn rates. Burn rates of the systems are obtained using Equation (2) and are normalized to burn rates of a periodic system for different ignition temperatures as shown in Figure 3(b). Figure 3(b) shows that burn rates of a Periodic system can be reproduced approximately by random systems described with normal (σ = 0.05) distribution. Again this is a consequence of closeness of these systems to a periodic system and is explained in the theoretical analysis section. By changing the parameters of a distribution, the inter cell spacing can be altered, which is reflected in the probability density function. For beta and uniform distributions, we observe no change in shape of distribution and consequently there is no effect on burn rates. However, for normal distribution, the burn rates can be varied by changing the parameter (σ). The normalized burn rate obtained with disordered system described by normal distribution with minimum variance is observed to be ~98% at each ignition temperature but for system’s microstructure described by uniform, beta distributions the percentage of normalized burn rates decreases with an increase in the ignition temperature. As the nature of distribution of reaction cells in actual systems is apriori unknown and hence it is of academic interest to find out the accurate model. Compared to gamma distribution (Bharath et al., 2013) of neighboring reaction cells, the normal distribution of reaction cells also has an advantage of parameter known as standard deviation. Hence the numerical calculations for model developed in the view of normal distribution of neighboring cells have to be carried out to establish accurate model and also explore the limits of different models. Normal distribution is employed for modeling in this paper as it offers several advantages—its fixed shape and positioning adjacent cells with controlled deviation from mean, around 65 percentage of adjacent reaction cells are positioned within one standard deviation, describe the microstructure of high energy material (Kulkarni & Sharma, 1998) mixtures. The flexibility of using normal distribution is due to fact that curve may be centered over any number of real lines and it may be flat or peaked to correspond to amount of dispersion in the values of random variable. The variance of normal distribution corresponds to concentration of point source particles of system when compared to that of periodic system. Different cases of normal distributed systems characterized with different standard deviations (~0.05, 0.19, 0.28, 0.40, ...) are considered for modeling and simulation. The combustion stops are more prominent for higher values of variance and higher ignition temperatures. The induction periods for higher ignition temperatures and higher variance of adjacent reaction cells increase. Symbols in Figure 4 show the normalized reaction burn rates calculated for different normal distributions. This detail provides us the possibility of tailoring burn rates of a system by suitable normal distribution. The normalized burn rates of a normal distribution are in the wide range (0.3–1). It can be seen that ratio = 1 resembles to a periodic system and 0.3 is closer to a high degree of disordered system. As variance of positioning adjacent cells in developed system increases, the concentration of sample density (Rogachev & Baras, 2009) decreases and its reaction front propagation corresponds to relay race homogeneous mechanism where as the reaction front propagation for the system with low
variance corresponds to quasi homogeneous mechanism (Gardner et al., 1999; Hwang et al., 1998; Mukasyan & Rogachev, 2008; Rogachev, 2003; Rogachev & Baras, 2009). Our numerical simulations suggest that apart from gamma distribution of reaction cells, the normal distribution of reaction cells also play a crucial role in describing the wide range of the combustion of actual heterogeneous systems. This shall be made evident by comparing the developed model (utilizing normal distribution of neighboring reaction cells) with available experimental data. In due process of comparison, we also try to establish scopes of the developed model.

3.2. Theoretical analysis for burn rates

The present section illustrates the theoretical analysis for dependence of burn rates of a disordered system, normalized to burn rates of periodic system, on variance of the distribution of neighboring reaction cells. Thus obtained theoretical expression is also compared with numerical results. Burn rates of combustible system (developed model) under consideration are determined by sequential calculation of ignition times of reaction cells. In the view of academic interest, it is essential to perform the correlation analysis between delay in ignition times and neighboring reaction cells. The correlation analysis is performed similar to the description as shown in theoretical section (Rashkovskiy et al., 2010). The non-dimensional steady-state burn rate of a periodic system can be written as:

\[ \tau = \omega_p^{-1} L \]  

(3)

where \( \tau \) is the delay in ignition times for neighboring reaction cells; \( L \) is the non-dimensional distance between neighboring reaction cells; \( \omega_p(\varepsilon) \) is the non-dimensional burn rate of periodic system where neighboring reaction cells are positioned with unit distances. The correlations (m), between \( \tau \) and \( L \), are obtained for a disordered system, described by a normal-distribution of neighboring reaction cells (\( L \)), as a functional dependence on variance (\( \sigma^2 \)) of distribution of neighboring reaction cells (\( L \)) and ignition temperature (\( \varepsilon \)). As \( \sigma^2 \rightarrow 0 \), the disordered system described by normal distribution of neighboring reaction cells (\( L \)) is similar to periodic system. Correlation analysis is performed on numerical calculations of Equation (1) for a disordered system over a broad range of parameters \( \sigma^2 \in [0.4 \ldots 0.05] \) and \( \varepsilon \leq 0.48 \). The correlation factor (m) thus obtained in terms of power functional dependence of \( m(\varepsilon, \sigma^2) \) is as follows:

\[ m = 2 + 0.2 \exp \left( 3.18 \varepsilon^{0.66} + (0.1 + 0.029 \ln \varepsilon)\sigma^2 - (0.0012 + 0.003\varepsilon)\sigma^4 \right) \]  

(4)

As \( \sigma^2 \rightarrow 0 \) for a disordered system, the correlation limit is \( m \rightarrow 2 \) at \( \varepsilon \rightarrow 0 \). The limiting range considered for normal distribution of neighboring reaction cells (\( L \)) is \( \sigma^2 \in [0 \ldots 0.4 \ldots 0.05] \) hence Equation (4) can be further modified to straightforward expression as:

Figure 4. Comparison plot between normalized burn rate and nondimensional ignition temperature \( \varepsilon \) for different values of \( \sigma \). Markers are obtained using Eqs. (1) and (2); solid lines are obtained by the theoretical Eqs. (5), (6) with \( \phi = 1 \).
The expectation value for $L^m$ with $L$ described by normal distribution in accordance with periodic system Equation (3) is given as:

$$\langle L^m \rangle = \int_0^\infty L^m P(L) dL$$

The above equation converges for $m$ being integer and the solution obtained is given below as:

$$\omega_r/\omega_p = 1/\phi \cdot (1/\sigma^2)^m \cdot \frac{\Gamma(1/\sigma^2)}{\Gamma((1/\sigma^2)+m)}$$

In the limit $\sigma^2 \rightarrow 0$, the system becomes periodic; this shows that $\omega_r \rightarrow \omega_p(\epsilon)$, in accordance to Equation (3) $m \rightarrow 2$ in this limit. Subsequently, we obtain $\lim_{\sigma^2 \rightarrow 0} \frac{1}{\phi} \cdot (1/\sigma^2)^m \cdot \frac{\Gamma(1/\sigma^2)}{\Gamma((1/\sigma^2)+m)} = 1$; thus one can conclude that $\phi \rightarrow 1$ for this limit. Figure 4 shows the comparison of burn rates, obtained by direct numerical solution of Equations (1) and (2), represented by markers, and the theoretical dependency, calculated using Equations (5) and (6), represented by solid lines, with $\phi = 1$.

It is evident that the theoretical dependence Equations (5) and (6) with $\phi = 1$ properly explains the results calculated by numerical simulations using Equations (1) and (2) for the whole range of parameters $\sigma$. Generalized expression for the theoretical dependence of normalized burn rates on variance is obtained and now can be used for direct calculation of burn rate if $\sigma$ is known.

### 3.3. CMDB Propellants

In this section, we compare experimental data for CMDB class of propellants (Kulkarni & Sharma, 1998) with the model developed. Systems modeled with normal or gamma distribution of neighboring reaction cells are compared with experimental data (Kulkarni & Sharma, 1998) to establish accurate model and also account the affects for dynamical combustion properties. In the experiments of Kulkarni and Sharma (1998), the samples were comprised of selected components such as DNC (Dinitrocarbonilide) and CL mixed in exact ratio with a binding agent AP (Amonium perchlorate) or without binding agent. The burning temperatures and burning rate of such propellants can change over broad range either by adding binding agent (AP) or by changing the compositions of components; the mechanism of heat release during combustion process of such propellant is not altered. In such a system, the group of active cells play the role of reaction cells, while the components are capable of reacting chemically. $T_{\text{ign}}$ is the melting point at which the cells commence reacting. The values of burning temperature and burning rate of CMDB propellants with binding agent or without binding agent are collected from the work (Kulkarni & Sharma, 1998). Ignoring the heat losses, calculated burning temperature of propellant mixtures is recognized as adiabatic temperature ($T_a$) with reference to developed system. Figure 5(a) includes the data from the work (Kulkarni & Sharma, 1998), with burn rate and burning temperature on the axis, for two propellants. The burn rate for CMDB propellants varies with percentage of components and also by adding binding agents. The first propellant comprises 60% of DNC and 40% of CL without the binding agent and other propellant comprises 50% of DNC and 40% of CL with AP as binding agent. It is observed that burn rate decreases with addition of binding agent, particularly at 690 k.

A similar treatment, using Equations (27–29) as shown in Bharath et al. (2013), is performed in the coordinates of $\epsilon-\omega$ for experimental data (Kulkarni & Sharma, 1998). In doing so it is considered that critical temperature, noticed in experiments, corresponds to critical values of the theoretical parameters such as $\epsilon_c$ and $\omega_{c\omega}$. In view of the developed model, the critical ignition temperature is established at 0.48; we consider these parameters as critical parameters for the disordered system. Assuming that minimum burn rate and burning temperatures, determined in the experiments,
correspond to the critical regime of combustion for real system. The values $\omega_{cr} = 5$ and $\omega_{cr} = 5.6$, respectively, were used for the mixtures DNC + CL and DNC + CL + AP in these calculations.

We assume that the change of burning rate with change in burning temperature for combustion of propellants is associated with change of parameters either $\sigma$ in case of normal distribution or $a$ for gamma distribution which describes system internal microstructure considering uniform identical reactant consumption. The data (Kulkarni & Sharma, 1998) are now processed in variables $\varepsilon - \omega$. The numerical results calculated for appropriate parameters of $\sigma$, $a$ at different ignition temperatures $\varepsilon$, is superimposed in Figure 5(b). Solid lines 1–4 are the theoretical dependences $\omega(\varepsilon)$, calculated using expressions Equations (6), (5) taking into account the matched dependence $\sigma(\varepsilon)$. Line 1 and 4 is obtained by utilizing the gamma distribution of neighboring reaction cells with parameter $a = 12$ and $a = 6$, respectively. Line 2 and 3 is obtained by utilizing the normal distribution of neighboring reaction cells with parameter $\sigma = 0.2886$ and 0.408, respectively.

Combustion of CMDB propellants releases high amount of energy, which propagates throughout the system, even at low burning temperatures. Thermal reactions below 670 k are like streak of lightening and are complex to analyze; however for the burning temperatures above 670 k, the developed model (Line 2 and 3) shows close agreement when compared to that of with Line 1 and 4. The minute aspect of controlling internal micro structure (Candel, 2002; Dvoryankin et al., 1985; Rogachev & Baras, 2009), by normal distribution, allowed for accurate explanation of data (Kulkarni & Sharma, 1998). The data (Kulkarni & Sharma, 1998), processed in variables $\varepsilon - \omega$ in Figure 5(b), show that burn rates for CMDB propellants change with ignition temperature either by the addition or absence of binding agent (AP). This binding agent corresponds to standard deviation parameter, of our developed model, employed for describing system’s internal microstructure; this is evident from Figure 5(b). It is apparent that the present model developed with normal distribution of neighboring reaction cells is accurate for describing the combustion process of CMDB type of heterogeneous mixtures.

### 3.4. Ti–Si system

Here we correlate the standard deviation, parameter of developed model, with the stoichiometric coefficient of actual heterogeneous mixtures. The work (Rogachev & Baras, 2009) for combustion of Ti–Si system is considered, for comparison in the view of the developed model, since the amount of gas released is relatively small. The combustion process of Ti–Si mixture is represented as $\text{Ti} + x\text{Si}$, where $x$ is referred as a stoichiometric coefficient. The plot for measured mean burn rate $r$ and stoichiometric coefficient $x$ is shown in Figure 3 from work (Rogachev & Baras, 2009), along with measured adiabatic temperature $T_a$. It is investigated that heterogeneous combustion of $\text{Ti} + x\text{Si}$ mixtures occurs in range of $x = [0.3, 1.5]$ as shown in work (Rogachev & Baras, 2009). Burning rate changes with change in stoichiometric coefficient. The maximum burning rate (38 mm/s) of mixture
coincides at highest value of burn temperature $T_B$. Burn rate is maximum at $x = 0.6$ which corresponds to synthesis $5\text{Ti} + 3\text{Si} \rightarrow \text{Si}_3\text{Ti}_5$.

Behavior of burn rate in range of $x = [1, 1.4]$ varies by previous trend: rate of burning decreases in the specified range even for the constant burning temperature. Such situation commonly arises in the combustion process of heterogeneous mixtures of $\text{Ti} + x \text{Si}$, where combustion process is associated with phase transformations and complex microstructural properties. Currently, no such alternate heterogeneous combustion models exist, for quantitative description of change in burn rate of thermit mixtures at constant burn temperature. Developed model, in view of normal distribution of neighboring reaction cells, has an additional scale of choice for tailoring neighboring reaction cells with variation of standard deviation parameter. Change of standard deviation parameter, for describing microstructure of system, results in change of burn rate, even at constant non-dimensional ignition temperature $\varepsilon$. Such a model explains change of burn rate for combustion of $\text{Ti} + x \text{Si}$ mixtures in the range of $x = [1, 1.4]$ where its burning temperature is constant as shown in Figure 3 from work (Rogachev & Baras, 2009). The correlation between standard deviation parameter and stoichiometry coefficient $x$ is performed at burning temperature 1,840 k. From the developed model, it is observed that the burning rate decreases with an increase in the standard deviation parameter. It is sufficient to presume that standard deviation parameter depends on stoichiometry coefficient $x$. As described in earlier section, in the view of the developed model for combustion process it is not possible to systematize a stable combustion process for $\varepsilon > 0.42$ under several primary circumstances in the entire range of standard deviation parameter. $\varepsilon = 0.48$ is interpreted as natural combustion limit for the developed combustion model. Equivalent of 1,840 k is obtained, in nondimensional ignition temperature by Equation (27) from (Bharath et al., 2013), as $\varepsilon = 0.44$. The experimental burn rates at different stoichiometric mixtures for constant burning temperature are treated by:

$$\frac{\sigma}{\sigma_c} = \frac{x}{x_c}(l_0/l_{0c})$$

Figure 6(a) shows for Burn rate vs. stoichiometric coefficient and Burn rate vs. standard deviation $\sigma$ shows the correlated values of stoichiometric coefficient and standard deviation. The solid line shows the numerical burn rates for different standard deviation parameters at $\varepsilon = 0.44$. Symbols represent experimental data. We obtain matched dependence of stoichiometric coefficient $x$ and standard deviation $\sigma$ for common value of burn rate. Now the dependence of standard deviation $\sigma(x)$ on stoichiometry coefficient $x$ is obtained by performing best fit on experimental data of burn rate. For certainty, it is assumed $\sigma = 1$ for the lower combustion limits. Matched dependence $\sigma(x)$ is shown in the Figure 6(b). Thus, variations in standard deviation of neighboring reaction cells completely correlate dependence of burn rate on stoichiometry coefficient $x$ even at constant burning temperatures. It is evident from Figure 6(b), analysis for the dependence of $\sigma(x)$, that the standard deviation parameter $\sigma$ reaches a minimum value $\sigma \approx 0.01$ at $x = 0.6$. 

Figure 6. (a) Left shows comparison of experimental and theoretical nondimensional burn rates on stoichiometry $x$ (Rogachev & Baras, 2009) and parameter $\sigma$. (b) Correlation between parameter $\sigma$ and stoichiometry coefficient $x$; dashed lines represent inflammability limits.
As shown above, the less $\sigma \ll 1$ the more ordered is the system. Obtained dependence $\sigma(x)$ (Figure 6) shows that the Ti + xSi mixture becomes more disordered as it moves away from $x = 0.6$. Such an effect of microstructure of the system on combustion process is associated with binding of Ti and Si particles, mixing at different concentrations, over the combustor volume.

3.4.1. Modes of combustion front

The mechanism for propagation of combustion front, is of fundamental importance, and is analyzed with the role of internal microstructure. The developed model explains these combustible limits in the view of ignition time profiles (in the range $[0.42, 0.48]$), if we assume, that $\varepsilon > 0.48$ is not in the range of $x = [0.3, 1.5]$). Hence, our numerical calculations consider that $\varepsilon = 0.48$ for both lower combustible limits and this determined the value of $T_{\text{ign}} = 990$ K. The existence of ignition temperature in delay, for combustion of heterogeneous mixtures, initiates a limit to reaction front propagation even in the absence of heat losses and with uniform heat release. The instant at which the heat release of the active cell is adequate to increase the temperature of the particles to the predetermined ignition temperature is defined as combustion limit. We observe in our numerical calculations, as time is incremented the temperature of active cell reaches critical temperature which is still lower than the predetermined ignition temperature. This instant is determined as numerical combustion limit (Bharath et al., 2013; Dvoryankin et al., 1985; Rashkovskii, 2005; Rashkovskiy et al., 2010; Rogachev & Baras, 2009) of developed model. The numerical combustion limit for a periodic system is 0.5459 (Rashkovskii, 2005; Rashkovskiy et al., 2010), however for disordered system the combustion limit still further reduces and is observed to be 0.48. The combustion front cannot propagate through the system above the Combustion limit (Bharath et al., 2013; Dvoryankin et al., 1985; Rashkovskii, 2005; Rashkovskiy et al., 2010; Rogachev & Baras, 2009). Figure 7 shows the ignition time profiles of different normal disordered systems ($\sigma = 0.05$ and $\sigma = 0.4$) for higher ignition temperatures ranging from 0.42 to 0.48. The periodic system and disordered system with $\sigma = 0.05$ has similar characteristics for lower values of ignition temperature < 0.42, however as ignition temperature increases the microstructure of system plays a dominant role in deciding the combustion limit at higher values of ignition temperature. The combustion front propagates in quasi-homogeneous mode when the thickness of the adjacent particles is much smaller than the vicinity of reaction front.

The combustion front propagates in relay race mode when inter particle distance of the mixture is of the order of width of the vicinity of combustion front.

3.5. Thermite systems at higher combustion limits

The present section illustrates the comparison of experimental data for combustion of thermite systems with the theoretical model developed in earlier section. In due process, we try to establish the effect of distribution of heat release on combustion limit of thermite mixtures. The work of authors (Dvoryankin et al., 1985) investigate that the burning rate changes with burning
temperature for a broad range by altering the inert diluter and percentage of inert diluter; in doing so the phenomenon of heat release in combustion process is not altered.

As established in above sections, by the developed model, the noticeable oscillations in those system commence at $ɛ_{\text{cr(developed)}} = 0.4$; we classify thermite mixtures from the work of the authors (Dvoryankin et al., 1985) based on their inflammability limits. Thus, we refer thermite systems from the work (Dvoryankin et al., 1985) into two: $ɛ_{\text{cr(Work)}} < ɛ_{\text{cr(developed)}}$, $ɛ_{\text{cr(developed)}} < ɛ_{\text{cr(Work)}}$. Here we consider thermite systems with $ɛ_{\text{cr(developed)}} < ɛ_{\text{cr(Work)}}$. Figure 10(a) comprise the data of work (Dvoryankin et al., 1985) (2Fe$_2$O$_3$ + 3Zr + n*ZrO$_2$, 2Cr$_2$O$_3$ + 3Zr + n*ZrO$_2$, Cr$_2$O$_3$ + 2Al + n*Al$_2$O$_3$), processed in co-ordinates of burn rate and burning temperature. Neglecting heat losses and considering uniform heat release of reaction cells, the calculated burning temperature of the mixtures is identified as adiabatic temperature ($T_{ad}$) of the heterogeneous mixture. The average burn rate, during entire time of combustion process of different heterogeneous mixtures, determined in the work (Dvoryankin et al., 1985); corresponds to theoretical average burning rate Equation (6) (Figure 4). The arrows in Figure 8(a) correspond to commencement of oscillating mode for combustion process of different thermite mixtures. Treatment of experimental data (Dvoryankin et al., 1985) has been performed in non-dimensional co-ordinates $ɛ - ω$. Hence theoretical values of the critical parameters such as $ɛ_{\text{cr}}$ and $ω_{\text{cr}}$ are assumed as commencement of oscillating modes of combustion, practical in experiments. As established above, by the developed model, the noticeable oscillations in those system commence at $ɛ_{\text{cr}} = 0.42$; and is considered as critical parameters of developed model. Assuming the critical values of rate of burning ($r_{\text{cr}}$) and burning temperature ($T_{ad,\text{cr}}$), measured in experiments, corresponds to commencement of oscillating modes of combustion process for an actual system. Then Equation (27), from (Bharath et al., 2013), in accordance with Figure 4, critical non-dimensional burning rate $ω_{\text{cr}} = ω(ɛ_{\text{cr}})$ depends on the standard deviation of neighboring reaction cells; this implies that $ω_{\text{cr}}$ depends on the system microstructure.

Experimental data (Dvoryankin et al., 1985) are treated using expressions (27)–(29) from Bharath et al. (2013), similar to method shown in Bharath et al. (2013), are shown in Figure 8(b). Line 2 (Figure 8(b)) shows the theoretical dependence for different values $α = $ Const. Line 1 shows the theoretical burn rates obtained for gamma distribution ($α = 0.7$) of neighboring reaction cells.

We assume that the change of non dimensional ignition temperature ($ɛ$) in diluting the system mixture is associated with change of the standard deviation parameter $α$, which describe the system microstructure considering uniform heat release at all cells. Furthermore, we consider that combustion process of thermite mixtures from work Dvoryankin et al. (1985) can be explained by the respective dependence $σ(ɛ)$. The dependence $σ(ɛ)$ is coordinated for the condition of concurrence of theoretical dependence $ω(ɛ)$ calculated by developed model with the experimental data (Dvoryankin et al., 1985) (Figure 8(b)). Such dependence $σ(ɛ)$ is shown in Figure 8(b). Theoretical dependence $ω(ɛ)$ is calculated using expressions Equations (6), (5) in the view of matched dependence $σ(ɛ)$, is
shown in Figure 8(b) (line 2); it describes experimental data (Dvoryankin et al., 1985) accurately for broad class of thermite mixtures. The value $\omega_{cr} = 0.25$ was used in these calculations. Solid line 2 is the result of disordered system described with normal distribution of reaction cells and line 1 is from gamma distribution ($\alpha = 0.7$). The Chi square at higher ignition temperatures ($\varepsilon > 0.32$) has been calculated for lines 1 and 2 with respect to experimental data; and we achieve a Chi square value of 0.054 for line 2 (normal distribution) and 0.523 for line 1 (gamma distribution $\alpha = 0.7$). The data (Dvoryankin et al., 1985), processed in variables $\varepsilon - \omega$, show that combustion becomes impossible at $\varepsilon = 0.49 \ldots 0.5$, this reality correlates well with the theoretical results obtained by the developed model for uniform heat release at all reaction cells.

3.6. Disordered heat release

In the work (Dvoryankin et al., 1985), for different thermite mixtures of Fe$_2$O$_3$, it is observed that two thermite mixtures (Fe$_2$O$_3$ + 2Al + n*Al$_2$O$_3$, 2Fe$_2$O$_3$ + 3Ti + n*TiO$_2$) have the inflammability limit less than $\varepsilon_{(work)} < 0.4$, and the steady-state mode of combustion is seen for further lower values of $\varepsilon$. The model developed in earlier sections with an assumption of uniform heat release ($q_i$) at all reaction cells could not explain for the thermite systems that have lower combustion limits. Currently, no such combustion models exist that can quantitatively explain the change of combustion limit for thermite mixtures. Unlike the above model, where the heat release is considered identical for all cells, the present section analyzes the effect of randomizing the heat release at all cells on combustion limit. It is established from the above developed model with normal distribution of neighboring reaction cells is more accurate with experimental data considered, here we consider normal distribution for positioning neighboring reaction cells with randomized heat release at each cell. We note that the system modeled with gamma distribution of neighboring reaction cells and randomizing the heat losses, the combustion front does not propagate even at lower ignition temperature ($\varepsilon < 0.05$). Hence the gamma distribution cannot be used in robust modeling of combustion process that considers randomizing heat release. Note the average heat release at each cells is maintained as unit. Heat release in the developed model is also viewed as the consumption of reactant. Randomizing the heat release not only allows us to study the combustion limit but also reveals the nature of distribution of heat releases of neighboring reaction cells.

An identical heat release at all point sources was assumed for the above numerical calculations. However, there can be a possibility of unequal heat release or unequal consumption of reactant (Gardner et al., 1999; Hwang et al., 1998; Mukasyan & Rogachev, 2008; Rogachev, 2003) at different cells. The heat release $q_i$ is randomized and its effects on the combustion are studied in the present section. The heat release distribution, with two extreme cases has been considered. The system with small spread in the distribution of heat release is described by a normal distribution ($q_i \in [0.7–1.2]$) and the more spread by a normal distribution ($q_i \in [0–7]$), however the average heat release is maintained at one. The modeled systems are calculated numerically and analyzed for their effect on combustion process. We observe that the small spread of distribution of heat release does not show considerable effect on combustion process and the combustion process is same as with uniform heat release. However as the degree of randomizing the distribution of heat release increases we observe considerable change in the process of combustion of such systems. Figure 9 shows the ignition time profile for the microstructure of system described by $\sigma = 0.05$ and $\sigma = 0.4$ and with high disordered heat release.

It can be seen that the combustion process is affected by the disordered heat release. With the inclusion of the random heat release, the burn front moves slower and combustion stops are observed. The effects are more pronounced at higher ignition temperatures and higher disorder of the heat release. Figure 10 shows the comparison of the burn rate obtained for a system with high disorder in the heat release with identical heat release. As the randomness in heat release increases, the burn rates decreases with an increase in the ignition temperature. It can be observed that at higher ignition temperatures, the decrease in the burn rate is more when compared to an identical heat release. At any given ignition temperature, a system described by a low degree of randomness in the position of its cells and higher degree of disorderness in nature of heat releases shows a higher
The combustion limit for the present disordered system with randomizing heat release is still further reduced to 0.40 from 0.48. And we detect oscillations of ignition time profiles at $\varepsilon = 0.35$. Our numerical calculations show that the combustion process of a system is affected by randomizing the heat release that leads to slow heating of system. Thus, the distribution of heat release in the system affects the combustion process (Hwang et al., 1998; Mukasyan & Rogachev, 2008; Rogachev, 2003) and hence cannot be neglected while modeling the combustible system.

As the clusterization of heat release increases then the combustion process differs to a great extent and this increases with an increase in the ignition temperature. Heat release study would be helpful for robust modeling of combustible system which not only accounts for microstructure of system but also the different possibilities of heat releases.

### 3.6.1. Thermite systems with inert diluents

Present section compares the experimental data for thermite mixtures (Dvoryankin et al., 1985), that consist of powder components ($\text{Fe}_2\text{O}_3 + 2\text{Al} + n\text{Al}_2\text{O}_3$, $2\text{Fe}_2\text{O}_3 + 3\text{Ti} + n\text{TiO}_2$) mixed with inert diluter, with theoretical model developed in section 3.2 where the heat release is disordered. Powdered
Figure 11. (a) Plot between burn rate and burn temperature for different thermite systems from work (Dvoryankin et al., 1985), having lower combustion limits, based on data of work (Dvoryankin et al., 1985). (b) Comparison plot (enlarged scale) between theoretical and experimental dependencies $\omega(\varepsilon)$. Symbols are the converted experimental data (Dvoryankin et al., 1985).

Notes: In (a) the arrow shows critical points at which combustion begins in oscillating modes. In (b) solid lines are the theoretical dependence for randomized heat release and normal distribution with microstructure described by line 1 $\sigma = 0.05$ and by line 2 $\sigma = 0.40$. 
components are capable of exothermic transformation. The temperature for burning the heterogeneous mixtures and their rate of burning can be altered for a broad range by changing the diluter and also by the amount of inert diluter; in doing so the lower inflammability limit in combustion is changed. The values of burning rate and burning temperature of heterogeneous mixture that contain different inert diluters and different percentages of inert diluter are collected from the work (Dvoryankin et al., 1985). Neglecting heat losses and considering randomizing distribution of heat release, the burning temperature of the heterogeneous mixtures is identified as adiabatic temperature ($T_{ad}$) of the mixture. It is established above, from the developed model (considering disordered heat releases), that the detectable oscillations in such mixtures begin at $\omega = 0.35$; we classify the thermite systems from the work of the authors (Dvoryankin et al., 1985) based on their combustion limits. Present section refers to thermite mixtures from the work (Dvoryankin et al., 1985) for which $\omega_{crit(work)} < \omega_{crit(developed)}$. Figure 11(a) contains the data of work (Dvoryankin et al., 1985) (Fe$_2$O$_3$ + 2Al + n*Al$_2$O$_3$, 2Fe$_2$O$_3$ + 3Ti + n*TiO$_2$), performed in the co-ordinates of burn rate and burning temperature. The arrows in Figure 11(a) correspond to the beginning of oscillating (critical) modes of combustion of thermite mixtures. Treatment of experimental data (Dvoryankin et al., 1985) is performed, similar to method in Bharath et al. (2013), in non dimensional co-ordinates of $\varepsilon - \omega$. Hence it is considered that commencement of oscillating mode of combustion process, noticeable during experiments, is associated with critical values of theoretical parameters such as $\varepsilon_{cr}$ and $\omega_{cr}$. As established above the noticeable oscillations of such heterogeneous mixture commence at $\omega = 0.35$, this parameter is considered as critical value of inflammability limit of the developed model. Assuming critical values of rate of burning $r_s$ and burning temperature $T_{ad}$, noticeable during experiments, corresponds to commencement of oscillating modes of combustion in an actual system. Experimental data (Dvoryankin et al., 1985) now converted using Equations (27–29) from (Bharath et al., 2013) are shown in Figure 11(b). Lines 1–2 (Figure 11(b)) show for different values $\sigma = const$ and randomized $q_i$.

We speculate the changing of nondimensional ignition temperature ($\varepsilon$) in dilution of thermite mixtures is associated with change of standard deviation ($\sigma$), which describe the system internal structure. And also assume change of combustion limit is associated with randomizing of distribution of heat release. Moreover, we consider the thermite systems and their combustion limit can be described by the dependence $q_i$ and $\sigma(\varepsilon)$. The value of parameter $q_i$ and $\sigma(\varepsilon)$ is matched from the concurrence of numerical (developed model) burn rates $\omega(\varepsilon)$ with the experimental data (Dvoryankin et al., 1985) (Figure 11(b)). Such dependence of $\sigma(\varepsilon)$ and $q_i$ is shown in Figure 11(b) (line 2). Theoretical dependence $\omega(\varepsilon)$, in view of the matched dependence of parameters $\sigma(\varepsilon)$ and $q_i$, is shown in Figure 11(b) (line 2); it appropriately explains the experimental data (Dvoryankin et al., 1985) of thermite mixtures having lower inflammability limits. The value $\omega_{cr} = 0.26$ was used for above calculations. Experimental data (Dvoryankin et al., 1985) allow estimating an inflammability limit of ignition temperature ($\varepsilon$) for each mixture, beyond which a stable combustion is not possible. The experimental data (Dvoryankin et al., 1985), now converted in the coordinates of $\varepsilon - \omega$, show that stable combustion is not possible at values $\varepsilon = 0.35 ... 0.4$; this reality agrees well with numerical results of the developed model obtained by randomizing distribution of heat release.

4. Conclusions
Gasless heterogeneous combustion system is modeled as a one-dimensional chain of combustible particles. The positions of adjacent reaction cells with significance to classification of distribution are described by normal, uniform, and beta. The reactions at higher ignition temperatures become complex and in such situations minute changes in internal microstructure can play a crucial role in affecting combustion process. In such situations, normal distribution of neighboring reaction cells has an additional scale of choice characterized by the parameter known as standard deviation. Relay race condition of combustion reaction front is observed for higher values of ignition temperature and higher degree of randomness in microstructure of system where as quasi homogeneous condition of combustion reaction front is observed for lower values of ignition temperature. Burn rates of a normal distributed disordered system are observed to be in the range of burn rates for periodic to
Randomizing the nature of distribution of heat release explains combustion process for thermite systems with lower inflammability limits. The theoretical combustible limit for disordered heat release in system is observed to be 0.38 and correlates with experimental inflammability limit, which is much less than that of identical heat release with normal distribution of active adjacent cells. We understand that a one-dimensional description of disperse system cannot reveal the flame structure; however, detailed numerical study on effect of microstructure, with significance to classification of random distributions, on combustion process shall list the priorities of distribution. The information from this work would be referred for multi-dimensional modeling of heterogeneous combustion process.

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