

INFLUENCE OF VARIABLE HEAT TRANSFER COEFFICIENT OF FIREWORKS AND CRACKERS ON THERMAL EXPLOSION CRITICAL AMBIENT TEMPERATURE AND TIME TO IGNITION

by

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Aim of this study was to investigate influence of heat transfer coefficient of fireworks and crackers on thermal explosion, critical ambient temperature and ignition time. It is proposed that heat transfer coefficient is power function of temperature, mathematical model to describe thermal explosion, steady-state and unsteady-state of finite cylindrical fireworks and crackers with complex shell structures are established. Models have been based on two-dimensional steady-state thermal explosion theory. The influence of variable heat transfer coefficient on thermal explosion critical ambient temperature and time to ignition are analyzed. When heat transfer coefficient is changing with temperature and in the condition of natural convection heat transfer, critical ambient temperature lessen, thermal explosion time to ignition shorten. If ambient temperature is close to critical ambient temperature, the influence of variable heat transfer coefficient on ignition time will be large. For firework with inner barrel, the critical ambient temperature of propellant is 463.88 K and ignition time is 4054.9 s. At ambient temperature 466 K, and with constant heat transfer coefficient critical temperature is 0.26 K lower and time to ignition 450.8 s less smaller. The calculation results show that the influence of variable heat transfer coefficient on thermal explosion time to ignition is greater in this example. Therefore, the effect of variable heat transfer coefficient should be considered into thermal safety evaluation of fireworks to reduce potential safety hazard.

Key words: cylindrical fireworks and crackers, variable heat transfer coefficient, critical ambient temperature, thermal explosion time to ignition

Introduction

Fireworks and crackers are ethnic characteristics and export products of China, enjoying high reputation in the international market. The thermal safety of fireworks and crackers in the production, storage, transport, and discharge has attracted considerable attention. The parent of modern theory of thermal explosion Semenov [1] introduced mathematical methods to establish a critical condition of thermal explosion for the first time. Frank-Kamenetskii [2] established the thermal explosion model with the spatial distribution of temperature in 1939. Thomas [3] put forward a new boundary condition, and a more universal theory was established comprising Semenov and Frank-Kamenetskii theory in 1958. In the early 1980s, Feng studied and summarized the domestic and foreign research results of thermal explosion. The theory of

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thermal explosion was established [4]. The theory of thermal explosion was applied to study the thermal safety of fireworks and firecrackers by Zhou [5]. There is much study about steady-state theory of fireworks and crackers in recent years, such as criterion of thermal explosion, critical ambient temperature, and so on [6]. However, the amount of the study of unsteady-state theory such as temperature process, thermal explosion time to ignition is less, especially the study of Non “class” A geometries.

As we know, it is very important that we should prevent the thermal accidents from happening in the process of transportation and storage of fireworks and crackers, which need scientific and reliable thermal explosion critical data to monitor the ambient temperature. Heat transfer coefficient is one of the most important boundary parameters in thermal explosion model, the accuracy of this parameter has an obvious effect on the simulation results. However, it is common that chemical reaction in system will be treated as zero order reaction when study the thermal safety of fireworks and crackers in previous work. In the energy conservation equation temperature affect nothing but the reaction rate. In fact, when the fireworks has a larger temperature difference between the chemical reaction system and the environment or a longer spontaneous combustion time (such as a few hours to a few days), variable heat transfer coefficient changes as the temperature changes observably, which enlarging the error of results. Thus, the effects of variable heat transfer coefficient should be considered in the study of thermal safety of fireworks and crackers. El-Sayed [7] examined how the criterion for criticality for a system with uniform temperature (Semenov case) is affected when the heat transfer coefficient is not constant, but depends on temperature. Wang and Du [8] considering the heat transfer coefficient as the function of temperature, studied the influence of variable heat transfer coefficient on critical ambient temperature of Semenov system and analyzed the thermal safety margin. A new method for theoretical analysis of the self-heating kinetics for monomolecular exothermic reactions has been proposed by Filimonov [9] and the detailed phase trajectories analysis has been performed for the case of first-order reactions. But no temperature process and thermal explosion time to ignition involved. The relationship between heat transfer coefficient and temperature is analyzed in this paper. According to two-dimensional thermal explosion theory, Mathematical thermal explosion steady-state and unsteady-state model of finite cylindrical fireworks and crackers with complex shell structures and variable heat transfer coefficient were established, simulating and analyzing the influence of variable heat transfer coefficient on critical ambient temperature and thermal explosion time to ignition.

The relationship between heat transfer coefficient and temperature

In literature [5] boundary heat transfer mode of the fireworks and crackers have been studied and clarified. Considering that the shell is exposed to air and that there is no forced convection in the production, storage and transport process, the external heat transfer coefficient is natural convection heat transfer coefficient. The heat transfer coefficient is the function of temperature [4]:

$$\chi = \chi_0 h(\theta) \quad (1)$$

where, χ_0 is the heat transfer coefficient with constant, $\theta = (T - T_a)/(RT_a^2/E)$ – the dimensionless temperature, and $h(\theta)$ stands for the function of heat transfer coefficient changing with temperature. The value of $h(\theta)$ in convection heat transfer is $h(\theta) = \theta^n$ [10]. Thus:

$$\chi = \chi_0 \theta^n \quad (2)$$

When $n = 0$, it means that heat transfer coefficient does not change with temperature. $n \neq 0$ represents for heat transfer coefficient changing with temperature. Here χ_0 is the heat transfer coefficient of the surface of shell at $\theta = 1 \cdot (T = RT_a^2/E + T_a)$. When $n > 0$, the situation of heat transfer coefficient increases with temperature can be simulated. Conversely, heat transfer coefficient decreases with temperature when $n < 0$. Particularly, when $n = 0.25$, it is the natural convection heat transfer for most materials [11].

Thermal explosion model and boundary condition treatment

Take the cylindrical fireworks as subject and the physical finite cylinder model can be seen in fig. 1.

According to literature [12], mathematical thermal explosion steady-state and unsteady-state model of finite cylindrical fireworks and crackers with complex shell structures are established based on energy conservation law, which consider the change of heat transfer coefficient with a_0 radius and H length diameter ratio (Model hypothesis is not mentioned here).

The conservation equation of thermal explosion steady-state model:

$$k \left(\frac{\partial^2 T}{\partial R_1^2} + \frac{1}{R_1} \frac{\partial T}{\partial R_1} + \frac{\partial^2 T}{\partial R_2^2} \right) + QA \exp \left(-\frac{E}{RT} \right) = 0 \quad (3)$$

The conservation equation of thermal explosion unsteady-state model:

$$\sigma c_v \frac{\partial T}{\partial t} = k \left(\frac{\partial^2 T}{\partial R_1^2} + \frac{1}{R_1} \frac{\partial T}{\partial R_1} + \frac{\partial^2 T}{\partial R_2^2} \right) + QA \exp \left(-\frac{E}{RT} \right) \quad (4)$$

The dimensionless parameters were introduced [4]. The dimensionless conservation equation of thermal explosion steady-state model was obtained:

$$\frac{\partial^2 \theta}{\partial \rho_1^2} + \frac{1}{\rho_1} \frac{\partial \theta}{\partial \rho_1} + \frac{1}{H^2} \frac{\partial^2 \theta}{\partial \rho_2^2} + \delta \exp \left(\frac{\theta}{1 + \varepsilon \theta} \right) = 0 \quad (5)$$

The dimensionless conservation equation of thermal explosion unsteady-state model:

$$\delta \frac{\partial \theta}{\partial \tau} = \frac{\partial^2 \theta}{\partial \rho_1^2} + \frac{1}{\rho_1} \frac{\partial \theta}{\partial \rho_1} + \frac{1}{H^2} \frac{\partial^2 \theta}{\partial \rho_2^2} + \delta \exp \left(\frac{\theta}{1 + \varepsilon \theta} \right) \quad (6)$$

Considering the relationship between heat transfer coefficient and temperature, effective Biot number can be written:

$$Bi = \frac{a_0}{k} \chi_0 \theta^n = Bi_0 \theta^n \quad (7)$$

Ignition point is unknown because of different cooling conditions of the upper and lower surface. In order to solve the equation, assuming ignition point location for x , the cylinder is divided into two zones by ignition point. The dimensionless boundary conditions of zone 1:

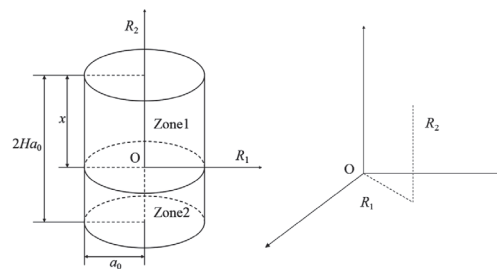


Figure 1. Physical finite cylinder model

$$\rho_1 = 0, \quad \frac{\partial \theta}{\partial \rho_1} = 0; \quad (0 \leq \rho_2 \leq x) \quad (8)$$

$$\rho_2 = 0, \quad \frac{\partial \theta}{\partial \rho_2} = 0; \quad (0 \leq \rho_1 \leq 1) \quad (9)$$

$$\rho_1 = 1, \quad \frac{\partial \theta}{\partial \rho_1} + \text{Bi}_r \theta^{1+n} = 0; \quad (0 \leq \rho_2 \leq x) \quad (10)$$

$$\rho_2 = x, \quad \frac{\partial \theta}{\partial \rho_2} + H \text{Bi}_{z1} \theta^{1+n} = 0; \quad (0 \leq \rho_1 \leq 1) \quad (11)$$

The dimensionless boundary conditions of zone 2:

$$\rho_1 = 0, \quad \frac{\partial \theta}{\partial \rho_1} = 0; \quad (x-2 \leq \rho_2 \leq 0) \quad (12)$$

$$\rho_2 = 0, \quad \frac{\partial \theta}{\partial \rho_2} = 0; \quad (0 \leq \rho_1 \leq 1) \quad (13)$$

$$\rho_1 = 1, \quad \frac{\partial \theta}{\partial \rho_1} + \text{Bi}_r \theta^{1+n} = 0; \quad (x-2 \leq \rho_2 \leq 0) \quad (14)$$

$$\rho_2 = x, \quad \frac{\partial \theta}{\partial \rho_2} + H \text{Bi}_{z2} \theta^{1+n} = 0; \quad (0 \leq \rho_1 \leq 1) \quad (15)$$

The initial conditions of thermal explosion unsteady-state model:

$$\tau = 0, \quad \theta = \theta_i \quad (16)$$

The numerical solution and influence analysis

The numerical solution

The mathematical model thermal explosion steady-state is described by equations (5), (8)-(15) and unsteady-state model by equations (6), (8)-(16). Five point difference method was used to discretize the model by equal step length. The model was solved by programming based on Matlab with Newton-homotopy method [13]. For the comprehensive and concise, the differential equation is not mentioned here. The critical parameter of zone 1 and zone 2 can be calculated. If the value of zone 1 is equal to zone 2, solution is obtained; If not, the calculation should be repeated by moving x by one step length until the two results are equal.

Influence of variable heat transfer coefficient on critical ambient temperature (Thomas system)

The boundary conditions of thermal explosion model vary from characteristic exponent n . When n determined, critical ambient temperature and thermal explosion time to ignition of fireworks and crackers can be obtained by solving non-linear equations. Using exponential approximation $\varepsilon = 0$, $\text{Bi}_{z1} = \text{Bi}_r = \text{Bi}_{z2}$ and the length diameter ratio $H = 1$. In order to analyze the influence of variable heat transfer coefficient on critical ambient temperature accurately,

the range of Biot number and length diameter ratio H taken for study is same to the literature (similarly hereinafter). The calculation results are listed in tab. 1.

Table 1. The influence of Biot number and n on δ_{cr} , $\theta_{0,cr}$

n	$Bi = 10^{-2}$		$Bi = 10^{-1}$		$Bi = 1$		$Bi = 10$		$Bi = 10^2$		$Bi \rightarrow \infty$	
	δ_{cr}	$\theta_{0,cr}$	δ_{cr}	$\theta_{0,cr}$	δ_{cr}	$\theta_{0,cr}$	δ_{cr}	$\theta_{0,cr}$	δ_{cr}	$\theta_{0,cr}$	δ_{cr}	$\theta_{0,cr}$
-0.25	0.011	0.756	0.112	0.793	0.908	1.093	2.474	1.570	2.730	1.600	2.760	1.613
0	0.011	1.000	0.107	1.041	0.841	1.311	2.293	1.601	2.706	1.613	2.760	1.613
0*	0.011	0.999	0.107	1.041	0.841	1.311	2.298	1.603	2.712	1.615	2.764	1.612
0.25	0.011	1.253	0.106	1.294	0.810	1.537	2.135	1.641	2.636	1.617	2.760	1.613

When $Bi_{z1} = Bi_r = Bi_{z2} = 10$, the calculation results are shown in tab. 2.

The results show that the error between the numerical solution ($n = 0$) and literature solution [14] ($n = 0^*$) is less than 10^{-2} . This proves that the numerical method used in this paper is accurate and reasonable

Seen from tab. 1 and fig. 2, criterion of thermal explosion δ_{cr} , temperature rise of system center $\delta_{0,cr}$ increase and tend to constant as Biot number increases. The effect of variable heat transfer coefficient on δ_{cr} first becomes larger then decreases with the increase of Biot number. When $10^0 < Bi < 10^2$, Biot number has greater effect on δ_{cr} . That is because the larger Biot number, the better heat dissipation, the higher critical temperature rise of system. The influence of heat transfer coefficient decreases with the increase of Biot number. Because heat dissipation is good enough and the influence of variable heat transfer coefficient can be ignored.

From tab. 2 that the smaller the length diameter ratio, the greater the influence of variable heat transfer coefficient on δ_{cr} . This is because the smaller length diameter ratio, the bigger the specific surface area of cylinder when feature size a_0 is certain. Thus, surface heat transfer has great effect on system.

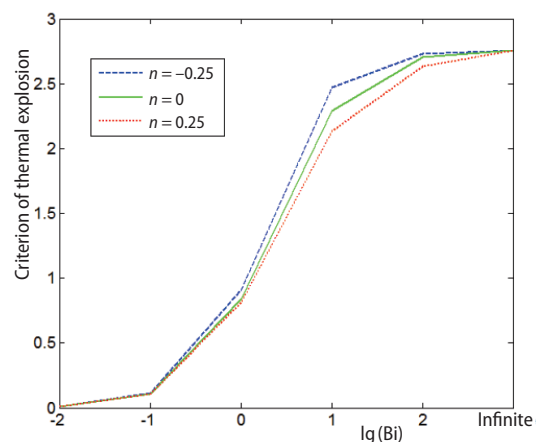


Figure 2. The influence of Biot number and n on δ_{cr}

Table 2. The influence of H and n on δ_{cr} , $\theta_{0,cr}$

n	$H = 0.2$		$H = 0.5$		$H = 1$		$H = 5$		$H \rightarrow \infty$	
	δ_{cr}	$\theta_{0,cr}$	δ_{cr}	$\theta_{0,cr}$	δ_{cr}	$\theta_{0,cr}$	δ_{cr}	$\theta_{0,cr}$	δ_{cr}	$\theta_{0,cr}$
-0.25	12.940	1.296	4.330	1.523	2.474	1.570	1.797	1.420	1.785	1.365
0	11.742	1.448	3.968	1.575	2.298	1.603	1.667	1.453	1.654	1.378
0*	11.743	1.448	3.968	1.575	2.298	1.603	1.667	1.453	1.654	1.378
0.25	10.963	1.570	3.673	1.641	2.136	1.647	1.545	1.512	1.532	1.420

According to tabs. 1 and 2, in practical application, the natural convection heat transfer condition ($n = 0.25$), considering heat transfer coefficient changing with temperature, the criterion of thermal explosion (critical ambient temperature) is lower.

Influence of variable heat transfer coefficient on thermal explosion time to ignition (Thomas system)

Using exponential approximation $\varepsilon = 0$, $Bi_{z1} = Bi_r = Bi_{z2} = 10$ and $H = 1$. Now ambient temperature is the major influence factor on thermal explosion time to ignition. Its dimensionless form can be expressed as: $\delta = a_0^2 Q E \sigma A \exp(-E/RT_a) / \lambda R T_a^2$. Study the effect of n on thermal explosion time to ignition with selecting different values of δ . The data in tab. 3 is dimensionless thermal explosion time to ignition τ_{ign} .

Table 3. The influence of δ and n on τ_{ign}

n	δ					
	2.5	2.8	3.0	4.0	5.0	10.0
-0.25	13.2666	3.1004	2.4091	1.4822	1.2510	1.0449
0	3.7816	2.3492	1.9958	1.3834	1.2039	1.0291
0*	3.7817	2.3493	1.9959	1.3835	1.2040	1.0250
0.25	2.6413	1.9655	1.7461	1.3085	1.1769	1.0231

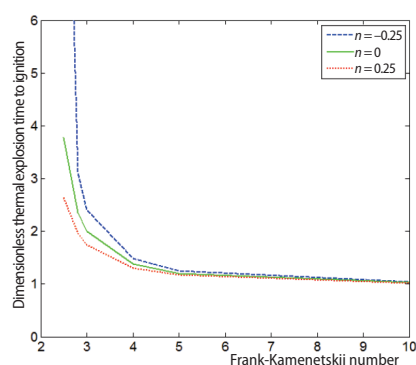


Figure 3. The influence of δ and n on τ_{ign}

delay of thermal explosion time to ignition.

Example analysis

Take “Dragones Voladores”, a kind of fireworks with inner barrel, as example. The propellant is black powder and the effector is fulminating powder. The height of the tube is 290 mm, the inside diameter is 30 mm, and the thickness of the side shell is 3 mm. The thickness of the mud shell is 25 mm. The mass of black powder is 3.7 g, the density of the charge is 0.4 g/cm³ and the height of the charge is 13 mm. The height of the effector is 120 mm, the inside diameter is 22 mm, and the thickness of the effector is 3 mm. The thicknesses of the upper, and down mud shell of effector are both 20 mm. The mass of fulminating powder is 15 g, the density of the charge is 0.33 g/cm³ and the height of the charge is 80 mm.

It can be seen from tab. 3 and fig. 3, thermal explosion time to ignition τ_{ign} decreases with the increases of δ . It means that the higher the ambient temperature, the more prone to thermal explosion. When the value of δ is small but greater than δ_{cr} , the influence of variable heat transfer coefficient on thermal explosion time to ignition is large.

Comparing case when $n = 0$ with case $n \neq 0$, $n > 0$, heat transfer coefficient increases when temperature increases, which can enhance the heat dissipation capacity. However, thermal explosion time to ignition shorten. The reason is that thermal explosion time to ignition starts from $\theta = 0$, and the temperature of chemical reaction system rises with the accumulation of heat. Before θ reaches 1, χ is always less than χ_0 which results in the rapid accumulation of heat and short of thermal explosion time to ignition. Conversely, when $n < 0$, heat transfer coefficient decreases as temperature increases. Before θ reaches 1, χ is always greater than χ_0 which results in

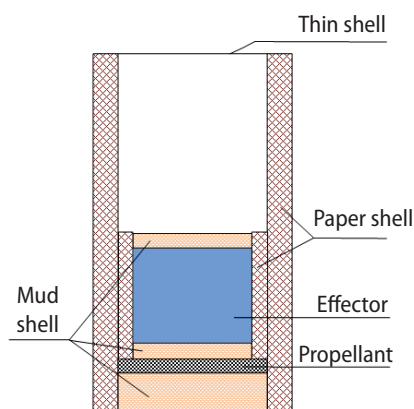


Figure 4. Simplified structure of the fireworks “Dragones Voladores” with inner barrel

Table 4. Physical and chemical parameters of various materials

Reactant	Activation energy E [kJmol ⁻¹]	Pre-exponential factor A [s ⁻¹]	Reaction heat Q [kJkg ⁻¹]	Charging density σ [kgm ⁻³]	Thermal conductivity coefficient [Wm ⁻¹ K ⁻¹]
Propellant/black powder	$1.91 \cdot 10^5$	$1.40 \cdot 10^{16}$	1526.26	748	0.0847
Effector/fulminating powder	$2.83 \cdot 10^5$	$8.09 \cdot 10^{15}$	593.25	876	0.1073

The thermodynamics and thermophysical parameters are listed in tab. 4. Activation energy, exponential factor and reaction heat are obtained by using TGA-DSC Analysis. The thermal conductivity are obtained by the DRY Thermal Conductivity Coefficient Tester. The thermal conductivity coefficient of paper shell, sealing powder and mud shell are 0.0357, 0.95, 0.95 W/mK, respectively.

The value of air natural convection heat transfer coefficient is usually between 1~10 W/m²K. The value of χ_0 is 5 in literature [15]. As a result, the effective Biot numbers of the propellant are $Bi'_r = 1.04$, $Bi'_{z1} = 0.08$, $Bi'_{z2} = 0.36$, respectively, and the effector's are $Bi'_r = 0.53$, $Bi'_{z1} = 1.69$, $Bi'_{z2} = 1.13$. The critical ambient temperature of propellant and effector obtained by solving the steady-state equation are 464.14 K and 696.92 K, respectively. If considering the change of the heat transfer coefficient, the critical ambient temperature of propellant and effector change to 463.88 K and 696.65 K, respectively. Take the lowest critical ambient temperature 463.88 K as assessment criteria of thermal safety evaluation.

In the study of thermal explosion time to ignition, only propellant need to be considered in this case, so the choice of ambient temperature should be higher than the critical ambient temperature. Because the shells of fireworks and crackers are made up of kraft paper and straw board, characteristic exponent n is 0.25 under the condition of natural convection heat transfer. Thermal explosion time to ignition is calculated as followed with several different ambient temperature.

From tab. 5 it can be seen that when the environment temperature is much higher than the critical ambient temperature (such as 490 K, 500 K), the difference in time to ignition is less than 10^{-1} , between the case when heat transfer coefficient change with temperature and case with constant heat transfer coefficient. It can be ignored in thermal safety evaluation of fireworks and crackers.

However, when the environment temperature is close to the critical temperature, variable heat transfer coefficient has a great influence on thermal explosion time to ignition. For example, when the ambient temperature is 466 K, considering heat transfer coefficient changing with temperature, thermal explosion time to ignition is 450.8 s less. Moreover as the environment temperature is closer to the critical temperature, the difference is greater.

It can be seen from fig. 5, at 470 K, the heating rate of the center is faster when considering the change of the heat transfer coefficient. This is because lower temperature rise lead to smaller heat transfer coefficient, more heat will be rapidly accumulated. As a result, for the complicated environment, it's

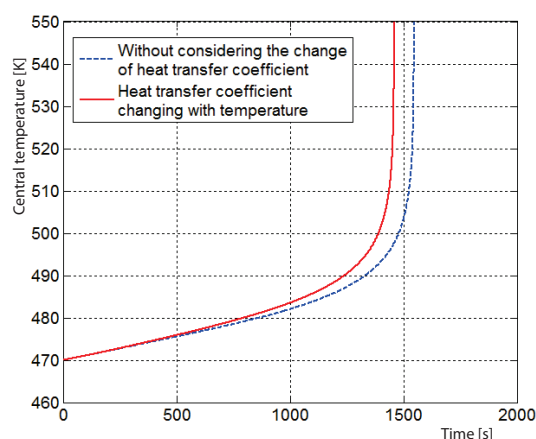


Figure 5. Temperature rise of system center of propellant at 470 K

Table 5. The results of thermal explosion time to ignition of propellant

n	T_a [K]						
	466	467	470	475	480	490	500
0	4054.9s	2955.7s	1545.0s	730.9s	403.2s	145.3s	58.7s
0.25	3604.1s	2701.0s	1466.4s	712.3s	398.1s	145.0s	58.7s

more reasonable to consider the influence of variable heat transfer coefficient when calculating the thermal explosion time to ignition.

Conclusions

- The influence of variable heat transfer coefficient on the boundary condition of heat dissipation of fireworks and crackers are analyzed. The mathematical thermal explosion steady-state and unsteady-state model of finite cylindrical fireworks and crackers which considering the heat transfer coefficient as the power function of temperature are established for the first time. When $n = 0$, the error between numerical solution and literature solution is less than 10^{-2} . Therefore the model is correct and the calculation procedure meets the practical computational requirements.
- When heat transfer coefficient is the power function of temperature and in the condition of the natural convection heat transfer, for Thomas system, there are several conclusions:
 - Firstly, the critical ambient temperature of cylindrical fireworks and crackers lessen. Meanwhile, Biot number has great influence on the critical ambient temperature within the range of 10^0 to 10^2 and the smaller the length diameter ratio, the greater the influence. In example analysis, the critical ambient temperature of propellant is 463.88 K, 0.26 K less than without considering the change of heat transfer coefficient.
 - Secondly, the thermal explosion time to ignition of cylindrical fireworks and crackers shorten. For firework with inner barrel, the thermal explosion time to ignition is 78.6 s less at 470 K. However, the thermal explosion time to ignition is 450.8 s less at 466 K. Thus, when ambient temperature is slightly higher than the critical ambient temperature, variable heat transfer coefficient has greater effect on the thermal explosion time to ignition. The calculation results also show that the influence of variable heat transfer coefficient on thermal explosion time to ignition is greater than the influence on the critical ambient temperature in this example.

In practice, this paper has a great application value for the thermal safety design and evaluation of fireworks and crackers. It provide basic data and theory guidance for safe storage and use of pyrotechnic products. The thermal explosion critical ambient temperature and time to ignition of any cylindrical fireworks can be obtained by solutions used in this paper. The ambient temperature can be monitored by the temperature sensor, *etc.* Once the ambient temperature higher than the critical ambient temperature, relevant and effective measures can be taken based on time to thermal explosion. Obviously, thermal explosion critical data will be scientific and reliable with considering the effect of variable heat transfer coefficient. Therefore, the effect of variable heat transfer coefficient should be considered into thermal safety evaluation of fireworks to reduce potential safety hazard.

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Nomenclature

A	– pre-exponential factor, [s ⁻¹]
Bi_r	– Biot number of the lateral surface, [–]
Bi_{z1}	– Biot number of the upper surface, [–]
Bi_{z2}	– Biot number of the lower surface, [–]
c_v	– pyrotechnic heat capacity, [JK ⁻¹]
E	– activation energy, [kJmol ⁻¹]
H	– length diameter ratio, [–]
k	– thermal conductivity of pyrotechnic, [Wm ⁻¹ K ⁻¹]
n	– characteristic exponent, [–]
Q	– reaction heat, [kJkg ⁻¹]
R	– universal gas constant, [Jmol ⁻¹ K ⁻¹]
T	– temperature of system, [K]
T_a	– ambient temperature, [K]
x	– dimensionless value of the distance from the ignition point to the upper surface ranging from 0 to 2, [–]

Greek symbols

δ	– Frank-Kamenetskii number, [–]
δ_{cr}	– criterion of thermal explosion, [–]
ε	– dimensionless activation energy, [–]
θ	– dimensionless temperature rise, [–]
$\theta_{0,cr}$	– dimensionless temperature rise of system center, [–]
ρ_1	– dimensionless co-ordinate variables in the r direction, [–]
ρ_2	– dimensionless co-ordinate variables in the z direction, [–]
σ	– pyrotechnic density, [kgm ⁻³]
τ	– dimensionless time, [–]
τ_{ign}	– thermal explosion time to ignition, [s]
χ	– heat transfer coefficient, [Wm ⁻² K ⁻¹]
χ_0	– heat transfer coefficient at $\theta = 1$, [Wm ⁻² K ⁻¹]

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