**Kinetic model of methane explosion in a spheroidal explosion tank**

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**Abstract.** Methane explosion is studied with a 20-liter content, spheroidal explosion tank. Mixture of methane with oxygen at varied ratio is detonated in the enclosed tank while pressure inside the tank is real-time recorded during the experiments. Temperature and conversion rate of the methane inside the tank during the explosion is calculated according to actual gas equation of state and energy conservation equation. Kinetic equations of the explosion process are derived among conversion rate, species concentration and temperature. Kinetic parameters of the steadily accelerating phase of the explosion process, including the apparent equilibrium constant and enthalpy change, as well as activation energy and pre-exponential factor, are calculated based on kinetic equations and experimental data.

1. Introduction

Gas explosion is a major disasters in coal mine, which often causes a large number of casualties and huge economic losses. Kinetics of methane combustion and explosion under different conditions is the bases for prediction the power of explosion that harms the mining operation. [1,2]

Methane explosion is a free radical chain reaction process, which includes many steps of elementary reactions. Deng et al. [3] studied the detailed reaction process of methane with oxygen, and calculated the reaction enthalpy changes, Gibbs free energies as well as activation energies of each reaction step by using density functional method B3LYP / 6-31G. Liang et al. [4,5], Li et al. [6], and Jia et al. [7] also studied Gas explosion reaction dynamics by quantum mechanics simulation. Lawrence Livermore national laboratory in the United States has proposed a detailed reaction mechanism and kinetic model of natural gas combustion which includes 53 components and 325 elementary reactions [8]. The activation energy of each elementary reaction is relatively small, but once the chain reaction is initiated, the reaction velocity accelerates rapidly, which usually leads to an explosion. However, the detailed mechanism is too complicated to analyze a specific working condition or product. In order to better understand the process of methane oxidation and reduce the difficulty of calculation, some simplified mechanisms [9,10,11,12] have been proposed one after another, such as Peter et al. [12] The simplified mechanism contains 14 components and 18 reactions. Zhu et al. [13] based on a 58-step methane oxidation reaction process, analyzed the chemical kinetic parameters in the flame zone using the basic principle of chemical reaction, and concluded that the reaction process on both sides of a counter flow flame of gas explosion has obvious symmetrical characteristics.
The relation between the overall reaction rate and temperature, and concentration of each component of a complex chemical reaction process can be expressed by Arrhenius equation. The apparent activation energy and frequency factor of the whole reaction can reveal development of the whole process conveniently. This apparent kinetics greatly simplifying the explosion calculation and simulation. However, due to the complexity and fast development of methane explosion process, it is difficult to obtain the apparent activation energy and frequency factor of explosion as a whole through experiments.

Many researchers have carried out experimental study of methane explosion in spherical sealed containers. Lu et al. [14] used spherical sealed containers to study the methane explosion characteristics and estimated the maximum explosion pressure. Lv et al. [15] used Fluidyn-MP multi-physics numerical simulation software to establish a 20 L spherical explosive tank analysis model. By changing the initial temperature and initial pressure, the deflagration characteristics of methane-air mixture in the sewage space and their changes were simulated and studied. Wang et al. [16] used a 20 L spherical gas dust explosion test device to study the effect of ethylene on the explosion pressure characteristics and flame propagation of methane. Xi'an University of Science and Technology has established a spherical explosion test system, Wang et al. [17] with which has carried out a large number of experiments to study the law of gas explosion, but there is no report on kinetic parameters studied with the experiment. Li et al. [18] established the connection between the microscopic chemical reaction mechanism at the initial stage of methane explosion and the macroscopic explosion pressure using a 20 L standard spherical explosion test device.

In the present study, based on explosion experiments of methane-air mixture in a spheroidal tank, kinetic parameters of the explosion process is calculated, which provides solid theoretic basis for prediction and prevention of gas explosion.

2. Experimental study on methane explosion

In each experiment, mixtures of methane with air at given ratio is ignited in the spheroidal explosion tank attached to an detecting system, and pressure inside the tank is real-time recorded after ignition.

The explosion experiment system is composed of five parts: explosion tank, gas proportional mixing system, ignition system, explosive suppression material adding and spraying system and measuring system, measuring and controlling system, as shown in Fig. 1 [4].

![Diagram of experimental system](image)

1-20l explosion tank; 2- pressure sensor; 3- vacuum pump; 4- computer; 5- controller; 6- pressure gauge; 7- air compressor; 8- ignition electrode; 9- gas cylinders; 10- digital manometer; 11 - nozzle; 12- explosive suppression material adding port; 13- solenoid valve; 14- high pressure air storage tank; 15- compressed air bottle.

Fig. 1 near-spherical gas explosion experimental system.

The explosion tank is a spheroidal stainless steel container whose internal maximum diameter is 30
cm, maximum height is roughly 35 cm and whose volume is about 20 L. The gas proportional mixing system is mainly composed of a vacuum pump, an air compressor, a precision digital manometer, a methane cylinder, etc. Ignition of the explosion is controlled by a computer, who operate the ignition electrode in the center of the reaction tank with an ignition energy of about 1 J. The monitoring and controlling system is mainly composed of pressure monitor, controllers and computers. When explosion is initiated, pressure inside the tank is recorded synchronously. The response time of pressure is 1 ms, the maximum collection time is 500 ms, and the sampling frequency is 1000 times/s.

The ambient temperature for the experiment was 20 ℃, relative humidity was 74%, and the initial pressure before detonation was normal. Mixed gas of air and methane was prepared by partial pressure ratio method: the explosion tank was firstly vacuumized with the vacuum pump, and then infuse pure methane to a certain pressure, and finally the solenoid valve was opened to let outside air in to balance the pressure inside and outside the reaction tank. According to Wang [19], that it takes only 0.140 s for the homogenization of the mixed gas, and 30 s for immobilization of the perturbations due mixing of different species of gases. Thereby the ignition of the mixture was conducted 30 seconds after the balanced pressure was achieved.

Mixed gases whose volumetric concentrations of methane being 7%, 8%, 10% and 12% were prepared for explosion experiments. In each experiments, when the mixture of methane and air was ignited, the monitoring system recorded the pressure inside the tank varies with time automatically, as shown in Fig. 2.

![Fig. 2 curves of explosion pressure vary with time for mixture at different methane concentrations.](image)

Fig. 2 curves of explosion pressure vary with time for mixture at different methane concentrations.

It can be seen from each time-pressure curve that after ignition of the CH4-air mixture, the pressure in the experimental tank rises increasingly at the beginning, and keeps a stable growth after some time, then pressure rise starts to slow down as time goes on, and declines after the peak value.

As to experiments with different methane concentration, the pressure rise is the fastest and the peak pressure is the highest when initial CH4 is around 10%.

Since pressure sensor is near the wall while the explosion is ignited from the centre of the tank, the pressure fluctuation was roughly eliminated during explosion wave propagate to the sensor, the pressure curve obtained in this experiment is fairly smooth and with no sharp peak. Thereby the pressure monitored is generally the mean pressure inside the tank, [20,21]which facilitates homogeneous model for kinetic analysis.
2. Quantitative analysis of methane explosion process

2.1 Concentration of each component and specific heat capacity of mixed gas

Methane explosion is a very complex radical chain reaction process to produce the ultimate products of H2O and CO2. The amount of other intermediate products in this process is very small, so the mixed gas in the tank after explosion is mainly composed of CH4, O2, CO2 and H2O [22,23]. The overall reaction equation is:

\[ \text{CH}_4 + 2\text{O}_2 \xrightarrow{k_1} \text{CO}_2 + 2\text{H}_2\text{O} + 802.292\text{kJ} \] (1)

The initial gas species in the explosion experiment include O2, N2, CH4 and a small amount of water vapor. The initial pressure is normal and the temperature is 25 degree centigrade. As the reaction goes on, CO2 and H2O are produced, but the total number of molecules remain basically unchanged. When conversion rate of CH4 is \( \alpha \), the molar concentration of each component is:

\[
\begin{align*}
C_{\text{CH}_4} &= (1 - \alpha)C_{\text{CH}_4,0} \\
C_{\text{O}_2} &= C_{\text{O}_2,0} - 2\alpha C_{\text{CH}_4,0} \\
C_{\text{CO}_2} &= \alpha C_{\text{CH}_4,0} \\
C_{\text{H}_2\text{O}} &= C_{\text{H}_2\text{O},0} + 2\alpha C_{\text{CH}_4,0}
\end{align*}
\] (2)

Where, \( C_{\text{CH}_4,0} \), \( C_{\text{O}_2,0} \), \( C_{\text{O}_2,0} \) and \( C_{\text{H}_2\text{O},0} \) are initial concentration of CH4, O2 and N2 inside the tank respectively, mol m\(^{-3}\).

Neglecting the interaction between different components, the specific heat capacity of mixed gas is:

\[ C_{VT,i} = \sum C_{VT,i}X_i \] (3)

Where, \( C_{VT,i} \) is specific heat capacity of component i at temperature T, and Xi is the mass fraction of the component i.

2.2 State equation of mixed explosive gas

As H2O phase transition may occur in the system, the Peng-Robinson Equation [24,25,26], which accurately defines gas-liquid equilibrium and saturated vapor pressure, was chosen to characterize the P-V-T relation of the mixed gases in the explosion tank:

\[ P = \frac{RT}{V_m-b} - \frac{a(T)}{V_m+b} \] (4)

Where, \( a(T) \) and \( b \) can be represented as the following formula

\[ a(T) = 0.45724 \frac{RT^2}{P_c} \left[ 1 + k(1 - T_r^{0.5}) \right]^3 \] (5)

\[ b = 0.07780 \frac{RT}{P_c} \] (6)

\[ k = 0.3746 + 1.54226w - 0.26992w^2 \] (7)

Where, \( V_m \) is molar volume, \( T_c \) and \( P_c \) denote critical temperature and pressure respectively; \( w \) denotes the eccentricity factor; \( T_r \) is the relative temperature, with \( T_r = T/T_c \).

2.3 Variation of temperature and methane conversion rate during explosion

Suppose that temperature in the container is uniform during the experiment, and heat capacity of the explosion tank is so small and can be ignored, then heat dissipated through the wall of the explosion is proportional to the temperature difference between inside and outside the tank [27,28,29], that is
\[ q_E = \beta (T - T_E) \]  
(8)

Where, \( q_E \) is heat flux through the tank wall, J s\(^{-1}\); \( \beta \) is heat dissipation coefficient, with \( \beta = 58.185 \text{J/(s·K)} \); \( T_E \) is the ambient temperature, K.

During the gas explosion experiment, the heat balance equation in the experimental tank is:

\[
\rho V C_v \frac{dT}{dt} = V q C_{\text{CH}_4} \frac{d\alpha}{dt} - \beta (T - T_E)
\]  
(9)

Where \( \rho \) denotes density of the mixed gas g m\(^{-3}\); \( V \) is the volume of the experimental tank, m\(^3\); \( q \) is reaction heat of the explosion process, with \( q = 802.292 \text{kJ mol}^{-1} \).

Difference form of the differential equation (9) is as follows:

\[
\rho V C_v \{T_{j+1} - T_j\} = C_{\text{CH}_4,0} \cdot V \cdot q \{\alpha_{j+1} - \alpha_j\} - \beta (T_j - T_E) \{T_j - t_j\}
\]  
(10)

Where, \( T_j \) represents temperature inside the tank at time \( j \); \( \alpha_j \) is the conversion rate at time \( j \). The initial conditions are \( \alpha_0 = 0 \) and \( t_0 = 293 \text{K} \).

In equation (10), conversion rate \( \alpha \) is the function of temperature \( T \) and time \( t \), where \( T \) can be calculated with the state equation according to the P value monitored in the experiment; \( C_v \) can be calculated according to equation (3). Therefore, by substituting \((t, P)\) obtained from the experiment into equation (10) from starting time \( t_1 \), the temperature and conversion rate at different times is obtained by numerical iteration, as shown in Fig. 3 and Fig. 4.

Fig. 3 explosion temperature vs time.
Fig. 4 methane conversion rate vs time.

FIG.3 showed variation of temperature calculated with time according to experimental data. It can be seen that when the methane concentration is 6%, 7%, 8% and 9%, the maximum explosion temperature are 1638.8 k, 1905.4 k, 2107.3 k and 2180.0 k respectively. The peak pressure as well as the average increasing slope of the stable increasing segment of each curve increases as the original methane concentration increased from 7% to around 10%.

The CH4 conversion rate (as shown in Fig. 4) speeds up growing at the beginning, then slows down, and finally approaches a certain value. The maximum conversion rate tends to decrease with the increasement of initial methane concentration. When the initial methane concentration is about 10%, it takes the shortest time to reach the maximum conversion rate.

3. Kinetic parameters of explosion reaction

3.1. Kinetic model of explosion reaction

Methane explosion is a complex free radicals chain reaction, including initiation, transfer and branching of the chain, which accelerates spontaneously once triggered.[30] Each elementary process of the reaction is reversible since both the reactants and the products are in gaseous states in an airtight container. According to the mass action law, the reaction rate is proportional to the concentration of oxygen and methane, namely

$$\frac{dC_{CH_4}}{dt} = k_1 C_{CH_4} C_{O_2} - k_2 C_{CO_2} C_{H_2}O$$  \hspace{1cm} (11)$$

Where, $k_1$ and $k_2$ denote rate constants of positive and negative reactions respectively. $C_{CH_4}, C_{O_2}, C_{CO_2}$ and $C_{H_2}O$ represent concentration of O2, CO2 and CH4 respectively, mol/m3.

$$\frac{d(1-\alpha)C_{CH_4,0}}{dt} = k_1 [C_{CH_4,0} (1-\alpha) C_{O_2,0} - 2\alpha C_{CH_4,0} ] - k_2 (\alpha C_{CH_4,0}^2)$$ \hspace{1cm} (12)$$

Assume that methane explosion reaction conforms to Arrhenius law, then

$$\frac{d\alpha}{dt} = k_{1,0} (1 - \alpha) (C_{O_2,0} - 2\alpha C_{CH_4,0}) \exp\left(\frac{-E_{a,1}}{RT}\right) - 2k_{2,0}\alpha^2 C_{CH_4,0} \exp\left(\frac{-E_{a,2}}{RT}\right)$$ \hspace{1cm} (13)$$
Where, $k_{1,0}$, $k_{2,0}$ are pre-exponential factors, and $E_{a1}$, $E_{a2}$ are activation energies (kJ/mol) of positive and negative reactions respectively; $T$ denotes temperature, K.

### 3.2 Apparent reaction enthalpy change and reaction equilibrium constant

It can be derived from equation (13) that

$$\frac{d\alpha}{dt} = k_{1,0}\exp \left(\frac{E_{a1}}{RT}\right)W$$

$$W = \frac{(1 - \alpha)(C_{O_2, o} - 2\alpha C_{CH_4, o})}{2\alpha^2 C_{CH_4, o}K_0 \exp \left(\frac{-\Delta H}{RT}\right)}$$

Where, $W = k_{2,0}/k_{1,0}$, $\Delta H$ is apparent reaction enthalpy change with $\Delta H = E_{a2} - E_{a1}$.

At the beginning of the explosion experiment, rate of the forward reaction is much higher than that of the backward reaction. As the reaction goes on, concentrations of the reaction products rise while that of the reactants fall gradually, which leads to a rising backward reaction rate and a descending forward reaction rate, and the chemical equilibrium attained ultimately.

When equilibrium of the reaction attained, both conversion rate of methane and temperature reaches the highest, namely $\alpha_{max}$, and $T_{max}$ respectively, therefore

$$k_{1,0}(1 - \alpha_{max})C_{O_2, o} - 2\alpha_{max}C_{CH_4, o}\exp \left(\frac{-E_{a1}}{RT}\right) = 2k_{2,0}\alpha_{max}^2 C_{CH_4, o}\exp \left(\frac{-E_{a2}}{RT_{max}}\right)$$

$$\ln B = \ln K_0 = \frac{\Delta H}{RT_{max}}$$

$$k_0 = k_{2,0}/k_{1,0} = \frac{(1 - \alpha_{max})C_{O_2, o}/C_{CH_4, o} - 2\alpha_{max}}{2\alpha_{max}}\exp \left(\frac{-\Delta H}{RT_{max}}\right)$$

$$B = \frac{(1 - \alpha_{max})C_{O_2, o}/C_{CH_4, o} - 2\alpha_{max}}{2\alpha_{max}}\exp \left(\frac{-\Delta H}{RT_{max}}\right)$$

Where, $K_0 = \frac{k_{2,0}}{k_{1,0}}$.

It can be seen from equation (16) that $\ln B$ and $1/T_{max}$ follow linear correspondence. Slope of the $\ln B - 1/T_{max}$ curve is $-\Delta H/R$ and intercept of it is $\ln K_0$. Based on the values of $C_{O_2, o}$, $C_{CH_4, o}$ and $\alpha_{max}$ in each experiment, the linear fitting curve of $\ln B - 1/T_{max}$ can be obtained, and $\Delta H$ and $K_0$ is obtained accordingly.

According to the explosion experiments with initial methane volume fraction being 7%, 8%, 9%, 10%, 11% and 12% respectively, the $\alpha_{max}$, $1/T_{max}$ and B value of each experiment are calculated. The linear fitting curve of $\ln B - 1/T_{max}$ is shown in Fig. 5.

From Fig. 5 we know that the points $(1/T_{max}, \ln B)$ distributed evenly on both sides of the linear fitting curve, indicating that $\ln B - 1/T_{max}$ conforms to the linear law perfectly. The fitting curve equation is $\ln B = -6860/T_{max} + 0.8298$, so the apparent equilibrium constant is $K_0 = e^{0.8298} = 2.29$, and apparent reaction enthalpy change is $\Delta H = 6860 \times 8.31/1000 \approx 57.00$kJ mol$^{-1}$.
3.3 Activation energy and frequency factor of the explosion reaction

Taking the logarithm operations for both sides of equation (14), we have

\[ F = \frac{E_a}{RT} - \ln(k_{1,0}) \]

\[ F = \ln \left( \frac{W}{d\alpha/dt} \right) \ln \left[ 1 - \alpha (C_{0,0} - 2\alpha C_{CH} - 4.58572 \alpha^2 C_{CH} \exp(-57.04/RT)) \right] \]

(17)

Where,

\[ W = \frac{1}{\alpha^2} \ln \left( \frac{1}{RT C_{0,0}} \right) \]

According to equation (17), when the reaction conforms to Arrhenius law, \( F \sim 1/T \) follows a linear law. Where, \( W \) is the function of \( T \) and \( \alpha \), \( d\alpha/dt \) can be obtained from \( \alpha \) curve. Therefore, \( F \) values at different temperatures can be obtained according to experimental data. According to the explosion experimental results, \( F \) and \( 1/T \) value with initial concentration of methane being 7%, 8%, 10% and 12% are calculated respectively, as shown in Fig. 6.

As can be seen from Fig. 6 that the value of \( F \) decreases sharply (the right most part of the curves) in the initial stage of the explosion, and followed by a linear decrease, and then a sharp increase (the left most part of the \( F \sim 1/T \) curves).

The above phenomenon may indicate that concentration of the free radical chain increases radically at the beginning of ignition, then kept roughly stable as explosion propagate steadily, and decreases sharply when the explosion reaction is going to be ended.

Fig. 6 also shows that the reaction in the stable development stage of the explosion conforms to Arrhenius law.
Fig. 6 $F \sim 1/T$ curves of explosion experiments with different initial methane concentrations.

Linear fitting equation of $F \sim 1/T$ curves of the various explosion experiments in the range of temperature 310K~1800K is $F = 854.17/T - 1.837$. By comparing the fitting equation with equation (17), the activation energy and frequency factor of the reaction are derived, namely

$$E_{a1} = 854.17 \times 8.31447/1000 \approx 7.10 \text{ kJ mol}^{-1}$$

and

$$k_{10} = e^{1.837} \approx 6.277677 \text{ m}^{3} (\text{mol} \cdot \text{s})^{-1}$$

Activation energy and frequency factor of its reverse reaction are

$$E_{a2} = \Delta H + E_{a1} = 57.04 + 7.10 = 64.14 \text{ kJ mol}^{-1}$$

and

$$k_{20} = k_{10} \times K_0 = 6.277677 \times 2.29286 = 14.39383 \text{ m}^{3} (\text{mol} \cdot \text{s})^{-1}.$$  

4. Conclusions

(1) Through the sealed spheroidal explosion tank experiment, the change of pressure in the tank during methane explosion was tested, and the change rule of temperature and conversion rate was calculated. As to experiments with different methane concentration, the pressure rise is the fastest and the peak pressure is the highest when initial CH$_4$ is around 10%.

The peak pressure as well as the average increasing slope of the stable increasing segment of each curve increases as the original methane concentration increased from 7% to around 10%.

When the initial methane concentration is about 10%, it takes the shortest time to reach the maximum conversion rate.

(2) the explosion of methane in a sealed container is a reversible reaction, and the overall reaction during the stable development period conforms to Arrhenius law. According to the experimental results, the apparent equilibrium constant of the overall reaction in the stable development period of methane explosion is 2.29, the apparent reaction enthalpy change is 57.04, and the apparent activation energy of the positive reaction is 7.10 kJ, and the apparent frequency factor is 6.28. Activation energy of reverse reaction was 64.14, and frequency factor is 14.39.

(3) The apparent kinetic parameters of methane explosion reaction obtained from the experiment can reflect the law of explosion from a macroscopic point of view, so as to facilitate the quantitative prediction of gas explosion conveniently.
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