Kinetic Analysis on Spontaneous Combustion of Pressurized Hydrogen in Tubes

Xiaofang Zhuo and Xiaolong Gou*

ABSTRACT: Highly pressurized hydrogen storage is considered as one of the best methods currently due to its economic performance. However, the highly pressurized storage technology is facing the threat of spontaneous combustion of high-pressure leakage, and there is still a lack of research on the kinetics of chemical reactions in the spontaneous combustion process, which greatly restricts the development of safe and efficient hydrogen-storage technology. Therefore, in this study, a three-dimensional simulation using the open-source packages OpenFOAM with a detailed kinetic model is proposed to analyze the hydrogen spontaneous combustion process in tubes. Subsequently, the effects and mechanisms of release pressures and tube geometry parameters are studied by means of kinetic simulation. The results show that the magnitude of the release pressure and tube diameter and length directly affects the spontaneous ignition and the location. In order to get more deep insights into the pressurized hydrogen release, reaction path analysis is performed. Three different hydrogen-consumed channels are found by reaction path analysis. The special performances found in spontaneous ignition with different release pressures and tube geometry parameters are caused by the competition between the chain-terminating channel and chain-branching channel. This work provides novel insights to understand the hydrogen spontaneous combustion process and enhances the theoretical basis for seeking safe hydrogen-storage means.

1. INTRODUCTION

With global energy shortage and the increasingly serious problem of the environment, hydrogen is widely concerned due to its high efficiency and nonpollution. A safe and economical storage and transport technology is the key bottleneck that restricts the widespread adoption of hydrogen. Highly pressurized hydrogen storage is considered as the best method currently due to its economic performance. However, the high-pressure hydrogen storage has to face the threat of spontaneous ignition, and there have been multiple auto-ignition incidents of high-pressure hydrogen leakage in the last century, which seriously threaten the safety of people and devices. Since that, to promote the use of this technology, it is urgent to avoid the autoignition accident.

Many investigations on high-pressure hydrogen-release process have been carried out, which were mostly based on the length of a downstream tube after hydrogen breaking through a diaphragm. In some early research works, the influence of tube length and release pressure on spontaneous ignition incidents of high-pressure hydrogen leakage in the last century, which seriously threaten the safety of people and devices. Since that, to promote the use of this technology, it is urgent to avoid the autoignition accident.

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affect the flow characteristics in the pipe, which is closely related to the fire phenomenon.

On the basis of previous studies, it is concluded that a large amount of sufficient mixing of shock-heated hydrogen and air is the major factor for autoignition and is closely related to the shock interaction and flow pattern inside the tube, which are influenced directly by burst pressure and downstream geometries. However, the deep understanding on spontaneous ignition mechanism is still lacking universality because experimental results were largely dependent on the various experimental facilities and limited by measuring instruments, while numerical results were limited by computation ability. For instance, some works that have a good performance in numerical simulation were limited to two-dimensional simulations.19,23

For more than 10 years, researchers have been searching for the mechanism of high-pressure hydrogen leakage and spontaneous combustion. However, so far, there is still a lack of research on the kinetics of chemical reactions so that the understanding of its microscopic mechanism is not deep enough, which greatly restricts the development of a safe and efficient hydrogen-storage technology. In this study, the three-dimensional simulation using the open-source packages OpenFOAM with a detailed kinetic model is proposed to analyze the spontaneous ignition process in pressurized hydrogen release and its kinetic characteristics. Besides, in order to save the problem of limiting of computation ability, the parallel computing is adopted.

2. Numerical Setups

2.1. Numerical Method. As one of the open-source CFD softwares, OpenFOAM provides the basic functions for the combustion-reacting flow simulation, and its powerful scalability and openness make it possible to implement different special tasks. In this study, the parallel computation is adopted to reduce the works and times of calculation. Besides, the reacting flow solver reactingFoam with a function of performing both flow calculations and the reaction calculations in OpenFOAM 4.0 Ubuntu is selected as the basic solver. The general governing equations are as follows:24

\[
\frac{\partial \rho}{\partial t} = - \nabla \cdot (\rho U) \tag{1}
\]

Mass equation

\[
\frac{\partial \rho U}{\partial t} = - \nabla \cdot (\rho U \otimes U) + \nabla \cdot \tau - \nabla p + \rho g \tag{2}
\]

Momentum equation

\[
\frac{\partial \rho Y_i}{\partial t} + \nabla \cdot (\rho U Y_i) = \nabla \cdot (D_i \nabla Y_i) + S_i \tag{3}
\]

Transport equation

\[
\frac{\partial (\rho h)}{\partial t} + \nabla \cdot (\rho U h) + \frac{\partial \rho K}{\partial t} + \nabla \cdot (\rho U K) = -\nabla \cdot q + \frac{\partial p}{\partial t} \tag{4}
\]

Energy equation

where \( \rho, U, p, h, \tau, Y_i, D_i, S_i, K_i \), and \( q \) are the density, velocity, pressure, mixture enthalpy per unit mass, viscous stress tensor, mass fraction of species \( i \), diffusion coefficient of species \( i \), chemical reaction rate, kinetic energy per unit mass, and heat flux, respectively.

A lot of studies have shown that the Lewis number (Le) of hydrogen in laminar or turbulent flames is significantly less than 1. However, in the process of high-pressure hydrogen-leakage spontaneous combustion, the formation and development of the flame is dominated by shock wave intensity. Also, the location and time of hydrogen spontaneous combustion process greatly depend on the interaction of multidimensional shock. Therefore, the Lewis number may have a certain influence on the diffusion flame structure in the mixing layer but has little influence on the probability and position of the spontaneous combustion. Considering the calculation cost and the robustness of numerical calculation, the hypothesis that \( Le = 1 \) is adopted in the model.

The governing equations are three-dimensional Navier–Stokes equations with the conservation equation of every chemical species for a reactive multicomponent mixture of perfect gases. The left of momentum equation, energy equation, and transport equation represent the unsteady and convective terms, respectively, and the right represent the diffusive and generalized source terms. The corresponding discretized schemes are as follows: the implicit Euler method is used for the unsteady term, a total variation diminishing flux limiter, the van Leer scheme is applied for the convection term, and the central difference format is applied for the diffusion term. There exist many different reaction mechanisms for hydrogen that can be used under the normal condition. However, for high-pressure conditions, predictions using the Burke mechanism adequately reproduce previous validation targets and show substantially improved agreement against recent high-pressure flame speed and shock tube speciation measurements.25 Therefore, the hydrogen mechanism of Burke et al.25 consisting of 13 components and 19 elementary reactions is selected in this paper.

2.2. System Modeling. In order to mimic the spontaneous ignition process in pressurized hydrogen release, a long circular tube is adopted. Figure 1 shows a schematic diagram of typical object of study, in which the overall pipe length is 300 mm, and \( D \) represents the inner diameter of the tube. There is a diaphragm in the tube, and the lengths of the upstream and the downstream are represented by \( L_1 \) and \( L_2 \), respectively. The right end is opening to the atmosphere. The transmissive boundary condition was used for the left and right ends, and the adiabatic nonslip boundary condition was set for the tube wall. The site of diaphragm is assumed as coordinate origin. The diaphragm eliminated at time zero is assumed because the time of bursting is instantaneous. The initial temperature of hydrogen and air was 300 K, and the initial air pressure was 1 atm. A nonuniform hexahedral grid was used in simulations, and the total number of the cells is 27,000.

In order to find the effects and mechanisms of release pressures and parameters, a great number of cases with different release pressures and downstream tube geometry parameters are calculated using this physical model and a detailed kinetic mechanism of hydrogen.25 Besides, reaction
PATH ANALYSIS IS PERFORMED UNDER SEVERAL TYPICAL CONDITIONS TO GET MORE DEEP INSIGHTS INTO THE PRESSURIZED HYDROGEN RELEASE.

3. RESULTS AND DISCUSSION

3.1. Model Validation. The whole process of autoignition of hydrogen at various release pressures is studied numerically, focusing on the ignition process and the formation of fire points, cross-sectional flames, and downstream propagation. Because the hydrogen spontaneous ignition is largely dependent on the downstream tube geometry, the bursting shape of the diaphragm, and release pressures, in this work, the parameter settings in each simulation case were consistent with the corresponding experiment such as the length and inner diameter of upstream and downstream pipe, burst pressures, and other boundary conditions. The simulation results are shown in Table 1, compared with the data of the previous related literature studies.

It can be seen from Table 1 that the simulated results of this work are in good agreement with the experimental results of Mogi and Lee under different burst pressures. Compared with the numerical results of Lee, they are close at very high pressures, and when the release pressure is 10 MPa, the simulated result of this paper is closer to the experimental ones. The availability and accuracy of the present computational model and algorithm have been validated by comparing the model predictions against experimental data and predictions of other modes. Consequently, it is reasonable to study the process of pressurized hydrogen release using this computational model.

3.2. Effect of Release Pressure on Ignition. 3.2.1. Flow and Heat-Transfer Process Analysis. Three cases were calculated to investigate the effect of burst pressure. Except for the release pressures of 10, 23.5, and 40 MPa, the other parameters were all fixed for all the three cases. Table 2 shows the ignition time and location of hydrogen at different release pressures in the configuration shown in Figure 1. As can be seen from Table 2, with the increase in burst pressure, the ignition time ($t = 0$ is the time of diaphragm breaking) becomes shorter and the distance of the ignition position from the diaphragm decreases. This is because the shock wave becomes stronger as release pressure increases, increasing the temperature of air in front of the hydrogen jet sharply and increasing the disturbance to promote the mixing of hydrogen and air, subsequently leading to ignition. Figure 2 shows the pressure and velocity distribution along the central axis of the pipe at various burst pressures, in which the vertical section near the right end shows the shock wave surface. As can be seen from Figure 2, with the increase in burst pressure, the pressure and flow rate changes caused by the shock wave in the pipe are aggravated, making the disturbance in the pipe more severe, promoting the mixture of hydrogen and air to ignition. The study also found that with the increasing pressure, the ignition time and location are less advanced (Table 2), which is similar to the experimental phenomenon as well.

As shown in Figure 3, the flame spreads to the right end after forming a flame in the cross section of the pipe. Due to the formation of the combustion wave interacting with the shock
wave, the disturbance in the pipe is more frequent and intense than before the ignition. Figure 4 shows the heat distribution at 20 μs from the formation of the flame through the entire cross section at various burst pressures. Obviously, as the release pressure is higher, the combustion is more intense and more heat is released.

3.2.2. Kinetic Analysis of Release Pressure Influence on Hydrogen Ignition. The literature studies\textsuperscript{20–22} have obtained the pressure limit curves for spontaneous ignition of some different experimental devices, but there is no uniform regularity, and in real applications, the hydrogen-storage pressure is about 35–75 MPa,\textsuperscript{26–28} far above the ignition limit pressure. Therefore, there is an urgent need to suppress ignition in the event of a leak. In order to seek new technologies to suppress the ignition, it is necessary to deeply understand the kinetic mechanism of high-pressure leakage spontaneous combustion. For kinetic analysis, the most critical is to get the reaction path flux of ignition. Based on the open convenience of OpenFOAM, this work replaced the implicit solution of the ODE toolkit with explicit discrete format, to obtain the intermediate parameters needed to solve the path flux.

The OH radical is the most important intermediate reactant in the self-ignition process. Therefore, the reaction paths of the OH radical at burst pressures of 10 and 40 MPa are shown in Figure 5. Table 3. Elementary Reactions

| number | reactions |
|--------|-----------|
| (1)    | H + O₂ → O + OH |
| (2)    | O + H₂ → H + OH |
| (3)    | H₂ + OH → H₂O + H |
| (9)    | H + O₂(+M) → HO₂(+M) |
| (11)   | HO₂ + H → OH + OH |
| (12)   | HO₂ + O → OH + O₂ |
| (14)   | HO₂ + HO₂ → H₂O₂ + O₂ |
| (15)   | HO₂₂(+M) → OH + O₂(+M) |
| (16)   | H₂O₂ + H → H₂O + OH |
| (17)   | HO₂ + H₂ → H₂O₂ + H |

Figure 4. Thermal contours at 20 μs from the formation of a flame through the entire section under different pressures (10.0, 23.5, and 40.0 MPa).

Figure 5. Reaction path analysis under two pressures (10 and 40 MPa). (a) $P_0 = 10$ MPa and (b) $P_0 = 40$ MPa.

Figure 6. Distribution of pressure at the ignition position under two release pressures.
of H₂ is initialized via H-atom abstraction from the fuel by O (O + H₂ = H + OH, reaction 2) and OH radicals (H₂ + OH = H₂O + H, reaction 3). Abundant H radicals are produced by the abstraction of a H atom. Then, H radicals can be consumed by the reaction 1, H + O₂ = O + OH. This reaction process can be summarized by the channel: H₂ → H → OH.

Figure 7. Path flux analysis under two pressures (7.7 and 7.8 MPa). (a) P₀ = 7.8 MPa (ignition) and (b) P₀ = 7.7 MPa (nonignition).

Figure 8. Distribution of temperature and velocity in the pipe at different times of different pipe diameters (t = 10, 20, 40, 60, and 65 μs). (a) P₀ = 10 MPa and d = 10 mm (before ignition t = 10, 20, 40, 60, and 65 μs), (b) P₀ = 10 MPa and d = 8 mm (before ignition t = 10, 20, 40, 60, and 65 μs), and (c) P₀ = 10 MPa and d = 3 mm (nonignition t = 10, 20, 40, 60, and 65 μs).
Besides, H radicals also can be consumed by the reaction with O₂ to form two HO₂ radicals [H + O₂(+M) = HO₂(+M), reaction 9]. Subsequently, the HO₂ reacts with H radicals to form two HO₂ radicals (HO₂ + H = OH + OH₂, reaction 11). This reaction process can be summarized by the oxidation of the \( \text{H}_2\text{O}_2 \) decomposed into two HO₂ radicals \( \text{HO}_2 \) and O₂ (HO₂ + H₂O₂ = H₂O₂ + O₂, reaction 14); then, the H₂O₂ decomposed into two OH radicals \( \text{H}_2\text{O}_2 \rightarrow \text{HO}_2 \rightarrow \text{OH} \). Moreover, the HO₂ can also be reacted to form H₂O₂ and O₂ (HO₂ + HO₂ = H₂O₂ + O₂, reaction 14); then, the H₂O₂ decomposed into two OH radicals \( \text{H}_2\text{O}_2 \rightarrow \text{HO}_2 \rightarrow \text{OH} \). Under a high-pressure condition (10 and 40 atm), two chain branching channels of 𝜋 and γ are the dominant reaction channels which lead to the phenomenon of ignition. Compared to the simulation of burst pressure at 10 MPa, both of the 𝜋 and γ channels are all enhanced in the positive direction when the burst pressure was 40 MPa. The greater the burst pressure, the higher the proportion of hydrogen is consumed and more radical OH is produced.

As shown in Figure 6, in the period before ignition, when the release pressure is 40 MPa, the pressure at the ignition position is 3–4 MPa, which is higher than the ignition pressure of 2 MPa at a release pressure of 10 MPa. It resulted in a positive promotion of the 𝜋 and 𝛾 channels. Therefore, with the increase in release pressure, the ignition time is decreased observably. A similar view that HO₂ pathways of hydrogen oxidize are sensitive at intermediate pressures was indicated by Burke et al.²⁵

In addition, the critical pressure of self-ignition was obtained by continuous simulation with reducing burst pressure, and the path fluxes of ignition and nonignition situations were compared to find the essential cause of ignition when high-pressure hydrogen leaks. In this study, the critical pressure is found to be 7.8 MPa. Therefore, it is impossible to ignite when the pressure is under critical pressure. When analyzing the simulation results without ignition, we selected the position corresponding to the highest temperature that occurred during the entire simulation as the study object, and the duration of the selection was 0.2 μs, as the same as the simulation of successful ignition.

A path flux analysis under two pressures (7.7 and 7.8 MPa) is shown in Figure 7. From Figure 7a, we can see that at a pressure of 7.8 MPa, about half of the H radicals are consumed by the 𝜋 channel, while the other H radicals are consumed by the 𝛾 channel. Hence, a large number of OH radicals are produced, which leads to the occurrence of ignition. However, in Figure 7b, the primary H radicals are consumed by the 𝜋 channel. A large number of intermediate species H₂O₂ are produced by this reaction path. However, the H₂O₂ is an unreactive species that seldom gets converted into OH radicals at low pressures and low temperature conditions. Therefore, under critical pressure, the 𝜋 channel becomes a chain-terminating pathway which leads to the phenomenon of nonignition.

### 3.3. Effect of the Tube Diameter on Ignition

#### 3.3.1. Flow and Heat-Transfer Process Analysis

Three cases were calculated to investigate the effect of the tube diameter on ignition. Except for the tube diameter of 10, 8, and 3 mm, the other parameters were all fixed for all the three cases. The distribution of temperature and velocity in pipe at different times of different pipe diameters is shown in Figure 8. As the pipe diameter decreases, the shock wave in the pipe develops faster and the shock intensity increases. The high-speed flow region represented by red in the figure continuously forms a Mach wave in front and superimposes with the foremost dominant wave so that the shock wave is not dissipated due to wall reflection. However, for the case of a pipe diameter of 3 mm, the high-speed flow region is still developing at 20 μs, but at 40 μs, the velocity was attenuated sharply. Also, the vortex was observed at 40–65 μs, which led to the change in the flow pattern for 3 mm. When the pipe diameter is 3 mm, a strong vortex appears quickly at the front end of the jet flow so that the shock wave no longer develops and then dissipates rapidly. The overall flow velocity decreases, and the mixed gas in the pipe can no longer be heated by the compression.

In order to observe the changes in the flow pattern when the tube diameter was 3 mm, the simulation calculation time was extended to the time of the mixed gas flow rushed out of the right end outlet. The distribution of velocity, vorticity, temperature, and mass fraction of the OH radical in the pipe at ignition time \( t = 113.8 \mu s \) with a diameter of 3 mm is...
shown in Figure 9. It turned out that in the later stage, the ignition happened in the center area behind the vortex, instead of appearing in the near-wall area, as shown in Figure 9. It means that the vortex makes the shock wave to be dissipated and then leads to a low temperature which makes the ignition fail in the near-wall region. Then, the vortex makes the gases to mix intensively and the temperature in the central region of the pipe behind the vortex to increase gradually.

For different pipe diameters, there are two ignition ways: ignition near the wall and ignition at the center. In order to find the ignition mechanism of different pipe diameters in more detail, the ignition times and positions with different tube diameters under the same release pressure were calculated and are shown in Figure 10. When the pipe diameter was decreased from 10 to 5.4 mm, the ignition time was reduced a little and then gradually increased. With the decrease in the pipe diameter, the shock wave intensified, which reduces the ignition time. However, it will also enhance the eddy current near the wall and make the heat in the near-wall region to be more easily taken away by the vortex. Therefore, the ignition

![Diagram](https://example.com/diagram.png)
increased gradually. When the pipe diameter is in the range of 5.3 to 4.5 mm, excessive heat is brought from the near-wall area to the center of the pipe, which causes the near-wall area to fail to ignite. Finally, a high-temperature zone in the center of the tube are formed and then ignited during the subsequent flow. The smaller the pipe diameter, the shorter the ignition time.

As can be shown in Figure 10, once the pipe diameter is less than 4.5 mm, the ignition positions change again. When the diameter is between 4.4 and 3.6 mm, ignition occurs in the near-wall region. When the pipe diameter is between 3.5 and 3 mm, ignition happens in the central region of the pipe. In order to find out the reasons, the cloud map distributions of the typical cases of tube diameters of 4.5 mm (ignition at the center), 4 mm (ignition near the wall), and 3 mm (ignition at the center) at 70 and 75 µs (approaching ignition time) were simulated and are shown in Figure 11. As described above, the vortex formed by the near-wall region takes away too much heat, which makes ignition in the near-wall area fail (Figure 11a). When the tube diameter is less than 4.5 mm, ignition near the wall reappeared. It can be illustrated by the fact that a high-speed vortex zone is formed at the front end of the airflow, which is much faster than the vortex of taking the heat away near the wall. It can compensate for the heat loss in the near-wall region, which makes the wall ignite successfully, as shown in Figure 11b. As can be seen from Figure 11, when the pipe diameter is in the range of 4.4 to 3.6 mm, the ignition time change curve is not unidirectional but decreases first and then increases as the pipe diameter decreases. This is because the high-speed vortex generated at the forefront of the airflow slows down the heat loss in the near-wall region. However, it also causes the shock wave to be dissipated faster. Therefore, as the pipe diameter decreases, the front-end vortex is generated earlier and the faster the shock is dissipated. Therefore, the inflection point occurs in the regularity of the ignition time. When the pipe diameter is 3 mm, the vortex near the wall increases, which makes the heat move away from the near-wall area. Also, the vortex at the front end is formed prematurely, which causes the flow pattern in the tube to change so that the shock wave is dissipated too quickly, which makes the heat loss in the near-wall area unable to be compensated and it cannot be ignited. Finally, in the later flow, there is a fire in the central area, as shown in Figure 11c.

3.3.2. Kinetic Analysis of Tube Diameter Influence on Hydrogen Ignition. In order to explore the influence of the downstream tube diameter, the path flux analysis by calculating several examples of the same release pressure of 10 MPa and different pipe diameters were obtained and are shown in Figure 12. Compared with a 10 mm pipe, the promotion of the channel is observed for an 8 mm pipe. Therefore, the ignition time was decreased for an 8 mm pipe diameter. However, when the pipe diameter decreased to 3 mm, the consumed channel of was replaced by the channel. The path flux analysis of a 3 mm pipe diameter shows that a large number of stable H2O2 was produced by the aforementioned chain-terminating pathway, which makes the ignition in the tube fail.
The reaction path of changing the diameter is consistent with the analysis in Section 3.2.

4. CONCLUSIONS

The open-source software package OpenFOAM with detailed kinetic models was used to conduct a three-dimensional simulation to analyze the spontaneous combustion process and its kinetic characteristics in the release of pressurized hydrogen. First, the influence of different leakage pressure conditions on the spontaneous combustion of hydrogen leakage is analyzed. The results of this study confirm that high pressure causes a stronger shock wave, which causes higher temperatures, makes ignition easier, and burns more intensely. For a specific structure and ambient temperature, there is a minimum leakage pressure that causes spontaneous combustion.

Then, the influence of the pipe diameter on ignition is studied. Different downstream pipe diameters may cause two different types of fire: fire near the wall and fire at the center. This is because the frequency of the interaction between the shock wave and the wall differs with the pipe diameter. The strength of the vortex near the wall and the formation time and strength of the high-speed vortex at the front end are also different. The numerical results show that as the diameter of the pipe decreases, the possibility of center fire increases. There may be two types of fire modes in a pipeline with a smaller diameter, which greatly increases the possibility of spontaneous combustion.

Subsequently, the chemical reaction path flux of the reaction flow under each typical working condition is analyzed to find out the key chemical species and reactions in the process of hydrogen spontaneous combustion. Three hydrogen-consumed channels, namely, $\text{H}_2 \rightarrow H \rightarrow OH$, $\text{H}_2 + H \rightarrow \text{HO}_2 \rightarrow OH$, and $\text{H}_2 + H \rightarrow \text{HO}_2 \rightarrow \text{H}_2 \text{O}_2 \rightarrow OH$, are found by reaction path analysis. The special performances found in spontaneous ignition with different release pressures and tube geometry parameters are caused by the competition between the chain-terminating channel and chain-branched channel. These simulation and analyses provide novel insights to understand the trends of the hydrogen spontaneous combustion process.

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