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The Inhibition Effect of Gas–Solid Two-Phase Inhibitors on Methane Explosion

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Received: 11 December 2018; Accepted: 21 January 2019; Published: 27 January 2019



Abstract: In order to study the inhibition effect of gas–solid two-phase inhibitors on a methane explosion, the influence of these parameters was investigated and compared with that of single-phase inhibitors. The results show that the inhibition effect of gas–solid two-phase inhibitors on a methane explosion is better than the added effect of two single-phase inhibitors, indicating that a synergistic effect can be obtained by gas–solid two-phase inhibitors. The two-phase inhibitors which are composed of NaHCO₃ (BC) powders and inert gas have a better suppressing property than those composed of NH₄H₂PO₄ (ABC) powders and inert gas. The two-phase inhibitors composed of CO₂ and powders have a better suppressing property than those composed of N₂ and powders. The 9.5% premixed methane–air mixture can be completely inhibited by 0.10 g/L BC powders mixed with 8% CO₂. The suppression mechanisms of the gas–solid two-phase inhibitors on the methane explosion were discussed.

Keywords: methane explosion; explosion suppression; gas–solid two-phase inhibitors; cooperative effect

1. Introduction

Methane is the main constituent of natural gas and mine gas [1,2]. It is also an important industrial raw material. However, it is easily ignited and causes explosion accidents, such as gas explosions in mines [3,4]. Explosion accidents caused by methane have caused great environmental damage and economic losses. In order to reduce the damage caused by methane explosions, inert gases and some chemical powders have been used to suppress them.

Inert gases, such as N_2 and CO_2 , can dilute the concentration of combustible gas and oxygen. Their suppression effects on methane explosions have been studied by many researchers. Liang [5] studied the effect of N_2 on methane explosions and found that the flame stability and the maximum explosion pressure decreased distinctly with the increase of the nitrogen content in the mixture. Benedetto [6] stated that the maximum explosion pressure and pressure rise rate of a hydrogen–methane mixture increased with decreasing CO_2 content or increasing O_2 concentration. In addition, research indicated that CO_2 had a better suppression effect than N_2 on the methane explosion, and the explosion was completely inhibited by 22% CO_2 or 32% N_2 [7–9]. Li [10] compared the suppression effect of He, Ar, N_2 and CO_2 on the hydrogen cloud explosion, and found that the suppression effect of CO_2 is better than He, Ar and N_2 . Chemical powders, such as carbonate, phosphate, halide etc., have widely been used as extinguishing agents on methane explosions and dust explosions, due to their physical and chemical inhibition effects. The inhibition effects of various chemical powders (such as SiO₂, CaCO₃, ABC, BC, zeolite, red mud, Al(OH)₃, composite powders, etc.) on methane explosions have been studied [11–26]. The experimental results presented that different powders showed different suppression performances on methane explosions.

Recently, more attentions have been paid to gas–solid two-phase inhibitors [27–29]. Deng [30] studied the influence of a two-phase inhibitor $Mg(OH)_2/CO_2$ on methane explosion, and observed that the actual superposition effect was inferior to the theoretical superposition effect. The research by Luo [31] showed that the ABC/CO₂ two-phase inhibitor had a cooperative synergism, which gave a good suppression performance on methane explosion. Jiang [32] researched the suppression effect of ultrafine ABC powders and N_2 mixture on methane explosions. The results showed that the maximum decreasing amplitudes of the overpressure and the flame speed were 76.8% and 100%, respectively. According to the research, it is clear that the suppression effect of gas–solid two-phase inhibitors was better than the single-phase gases or powders. However, there are few studies comparing the actual suppression effect of gas–solid two-phase inhibitors with the theoretical addition effect suppressed by single-phase gas and powders, respectively. It is not clear whether there is a cooperative effect between inert gas and powders.

In this paper, the influence of ABC powders (NH₄H₂PO₄), BC powders (NaHCO₃), N₂, CO₂ and gas–solid two-phase inhibitors on the 9.5% premixed methane-air explosion parameters, including the max-pressure, the time to max-pressure and the maximum rate of pressure rise, was experimentally researched using a 20 L spherical vessel. The actual suppression effect of gas–solid two-phase inhibitors was compared with the theoretical addition effect suppressed by single-phase gas and powders respectively, aiming to clarify whether there was a cooperative effect between inert gas and powders. The suppression mechanisms of the gas–solid two-phase inhibitors on methane explosions were discussed.

2. Materials and Methods

2.1. Explosion Test System

Experiments were performed in a 20 L spherical explosion test system. The system mainly includes an explosion vessel, a gas allocation system, a control and data acquisition system and a powders injection system, as shown in Figure 1.



Figure 1. The illustration of 20 L spherical explosion test system. 1—Circulating water export; 2—Ignition electrode; 3—Powder storage tank; 4—Control box; 5,12—Compressed air; 6—Gas-powder two-phase valve; 7—Methane; 8—Solenoid valve; 9—Vacuum gauge; 10—Pressure sensor; 11—Vacuum pump; 13—Inert gas.

The test procedure is as follows. Firstly, a certain amount of powders was placed into the 0.6 L powder storage tank, and then the compressed air was filled into it to 2 MPa. Secondly, a part of air in the explosion vessel was pumped out to negative pressure. Using the partial pressure method, the 9.5% premixed methane–air mixture and a certain amount of inert gases were injected into the explosion vessel to -0.06 MPa. Then, the solenoid valve between the powder storage tank and the test chamber was triggered within 10 ms when the ignition button was pressed, and the high-pressure air and the powders were dispersed into the chamber to make sure the explosion proceeded under atmospheric pressure. The injection time was 50 ms and the ignition delay time was 60 ms. A chemical igniter with the ignition energy of 100 J was used in this study and the explosion pressure measured in gauge. The data acquisition card had a resolution of 0.021 kpa, and the data measured by the pressure sensor were accurate to four significant digits. The experimental data were collected and recorded by the data acquisition system. In the experiment, the tests were repeated at least 3 times under the same conditions until the test results tended to be stable.

According to the experimental criterion made by the ASTM (American Society for Testing and Materials), it can be considered that the explosion occurs when the pressure increases by 7% or more [31].

2.2. Materials

The powders used in the experiment are ABC and BC powders. As explosion suppression powders, BC and ABC powders possess physical and chemical inhibition mechanisms against explosion. The main components of ABC powders are $NH_4H_2PO_4$ (>99%), and the average particle size used in the experiment was 75.8 µm. The main components of BC powders are $NaHCO_3$ (>99.5%), and the average particle size used in the experiment was 43.9 µm. The concentrations of ABC/BC powders used in the experiments were 0.06 g/L, 0.08 g/L, 0.10 g/L, and 0.12 g/L, respectively. The inert gases used in the experiment are N₂ and CO₂. As explosion suppression materials, N₂ and CO₂ have the characteristics of economic and environmental protection. The volumetric fractions of CO₂ and N₂ (99.99%) are 2%, 4%, 6% and 8%, respectively.

3. Results and Discussion

3.1. Explosion Suppression Effects of Single-Phase Inhibitors

The suppression effects of single-phase inhibitors on methane explosion were tested first. The explosion pressure–time curves with different concentrations (0.06 g/L and 0.10 g/L) of ABC or BC powders and with different volume fractions (2% and 8%) of CO₂ or N₂ are presented in Figure 2a,b, respectively. It was shown that the chemical powders and inert gases displayed different degrees of inhibition effects on the methane explosion. The suppression effect increased with the increase of the inhibitors' concentration or volume fraction. BC powder showed better inhibition performance than ABC powder with the same concentration. CO₂ presented better inhibition performance than N₂ with the same volume fraction.

The explosion characteristic parameters of methane under different concentrations of single-phase inhibitors are listed in Table 1. As can be seen from Table 1, for the powder inhibitors, the max-pressure of methane decreased from 0.70 MPa to 0.52 MPa after the addition of 0.10 g/L BC which decreased by 25.7%. The time to max-pressure extended from 0.12 s to 0.37 s, extending by about 2.08 times. The maximum rate of pressure rise decreased by 91.2%. As the same concentration of ABC powders was added, the max-pressure decreased by 15.2%, the time to max-pressure extended by 0.92 times, and the maximum rate of pressure rise decreased by 72.8%. For inert gas inhibitors, the max-pressure decreased from 0.70 MPa to 0.58 MPa after the addition of 8% CO₂, which decreased by 17.1%. The time to max-pressure extended from 0.12 s to 0.20 s, which extended by 0.67 times. The maximum rate of pressure rise decreased by 0.67 times.

decreased only by 10.7%, the time to max-pressure extended by 0.42 times, and the maximum rate of pressure rise decreased by 49.0%.



Figure 2. Pressure–time curves of methane explosion under single-phase inhibitors: (a) NaHCO₃ (BC)/NH₄H₂PO₄ (ABC) powders; (b) N_2/CO_2 .

Sample	Max-Pressure (MPa)	Decline Rate of Max-Pressure (%)	The Time to Max-Pressure (s)	Multiple of the Time to Max-Pressure Extension (s)	The Maximum Rate of Pressure Rise (MPa·s ⁻¹)	Decline Rate of the Maximum Rate of Pressure Rise (%)
No Suppressants	0.70	0	0.12	0	29.7	0
0.06 g/L BC	0.58	17.1	0.28	1.33	4.35	85.4
0.10 g/L BC	0.52	25.7	0.37	2.08	2.62	91.2
0.06 g/L ABC	0.62	11.4	0.19	0.58	10.15	64.1
0.10 g/L ABC	0.59	15.2	0.23	0.92	8.07	72.8
2% CO ₂	0.61	12.9	0.17	0.42	15.59	47.5
8% CO2	0.58	17.1	0.20	0.67	10.7	64.0
2% N ₂	0.67	4.3	0.15	0.25	22.21	25.2
8% N2	0.62	11.4	0.17	0.42	15.14	49.0

Table 1. The explosion characteristic parameters of methane under a single-phase inhibitor.

3.2. Effects of Powders in Gas-Solid Two-Phase Inhibitors on Methane Explosion

In this part, we studied the effects of two different powders in gas–solid two-phase inhibitors on the methane explosion. The suppression effects of the two-phase inhibitors of ABC or BC powders mixed with two different inert gases are presented in Figure 3. Figure 3a,b exhibits the max-pressure and the time to max-pressure of the methane explosion inhibited by 0.10 g/L ABC or BC powders mixed with CO_2 (volumetric fractions of 2%, 4%, 6%, 8%) and N_2 (volumetric fractions of 2%, 4%, 6%, 8%), respectively.

From Figure 3, it can be seen that the max-pressure decreased and the time to max-pressure extended gradually with the increase of the volumetric fraction of CO_2 or N_2 . Whether mixed with CO_2 or N_2 , two-phase inhibitors of BC/inert gas revealed more effective suppression than ABC/inert gas. As the volumetric fraction of N_2 increased from 2% to 8%, the max-pressure of methane explosion decreased from 0.60 MPa to 0.55 MPa and the time to max-pressure increased from 0.21 s to 0.28 s by ABC/N₂. Under the same condition, the max-pressure decreased from 0.54 MPa to 0.44 MPa and the time to max-pressure increased from 0.26 s to 0.57 s by BC/N₂. When using CO_2 with the gas–solid two-phase inhibitors and a volumetric fraction of 8%, the max-pressure of methane explosion was decreased to 0.47 MPa by ABC/CO₂. When the BC/CO₂ two-phase inhibitor with the same concentration was added, the 9.5% premixed methane–air explosion was suppressed completely. That may be because BC powders, which are composed of NaHCO₃, can release CO₂ in the decomposition process, but ABC powders cannot.



Figure 3. The explosion characteristic parameters of methane inhibited by different powders with inert gas: (a) different powders/ CO_2 ; (b) different powders/ N_2 .

3.3. Effects of Inert Gases in Gas-Solid Two-Phase Inhibitors on Methane Explosion

On the other hand, we studied the effects of different inert gases in gas–solid two-phase inhibitors on the methane explosion. Figure 4 shows the explosion characteristic parameters of methane inhibited by N₂ or CO₂ mixed with given powders. Figure 4a,b exhibited the max-pressure and the time to max-pressure of methane inhibited by two-phase inhibitors of N₂ or CO₂/ABC, respectively. Figure 4c,d presented those with N₂ or CO₂ mixed with BC powders. The volume fraction of N₂ or CO₂ was 8%, and the concentration of the ABC or BC powders varied from 0.06 g/L to 0.10 g/L. It was shown that mixing with the same concentration of powders, two-phase inhibitors of CO₂/powders exhibited more effective suppression than N₂/powders. The max-pressures decreased and the times to max-pressure extended gradually with the increase of ABC or BC concentration. It should be noted here that the methane explosion could be completely inhibited by 8% CO₂/0.10 g/L BC powders, but could not be completely inhibited by 8% N₂/0.10 g/L BC powders. This may be because CO₂ could participate in the chain reaction H + CO₂ \leftrightarrows OH + CO to consume H free radicals [33] and can react with the explosion free radicals more actively than N₂.



Figure 4. The explosion characteristic parameters of methane inhibited by different inert gases mixed with powders: (**a**) the max-pressure of different inert gases/ABC; (**b**) the time to max-pressure by different inert gases/ABC; (**c**) the max-pressure of different inert gases/BC; (**d**) the time to max-pressure of different inert gases/BC; (**d**) the time to max-pressure of different inert gases/BC.

In order to compare the suppression performance of gas–solid two-phase inhibitors with different ratios on methane explosion more intuitively, the max-pressure, the time to max-pressure and the maximum rate of pressure rise with single-phase inhibitor and gas–solid two-phase inhibitors are presented in Figure 5a–c, respectively. The corresponding explosion parameter values are shown in Table 2.



Figure 5. The suppression effects of different inhibitors: (a) the max-pressure; (b) the time to max-pressure; (c) the maximum rate of pressure rise.

Table 2. The methane explosion parameters with different inhibitors.

Sample	Max-Pressure (MPa)	The Time to Max-Pressure (s)	The Maximum Rate of Pressure Rise (MPa·s ⁻¹)
No Suppressants	0.70	0.12	29.70
8% N ₂	0.62	0.17	15.14
8% CO ₂	0.58	0.20	10.70
0.10 g/L BC	0.52	0.37	2.62
0.10 g/L ABC	0.59	0.23	8.07
8% N ₂ /0.10 g/L BC	0.44	0.57	1.09
8% N ₂ /0.10 g/L ABC	0.55	0.28	5.25
8% CO ₂ /0.10 g/L BC	No Explosion	-	-
8% CO ₂ /0.10 g/L ABC	0.47	0.39	2.27

It can be seen from Table 2 that, among all the inhibitors, only the 8% CO₂/0.10 g/L BC mixture could suppress the methane explosion completely. The pure inert gases or powders and the gas–solid two-phase inhibitors of other ratios could not suppress the explosion completely. It can be deduced that the gas–solid two-phase inhibitors possess the cooperative suppression effect on the methane explosion.

3.5. Comparison of Actual Suppression Effect and Theoretical Addition Effect

In order to explore the cooperative effect of the gas-solid two-phase inhibitors on the methane explosion, the actual suppression effect of CO_2/BC two-phase inhibitor and the theoretical addition effect (theoretical superposition of suppression effects of two single-phase inhibitors) of the two single-phase inhibitors are compared in Figure 6. Figure 6a shows the differences between the actual max-pressure decreased by CO_2/BC and the theoretical addition of CO_2 and BC. Figure 6b displays the differences of the time delay to max-pressure. It can be seen from this figure that the differences between the actual suppression effect and the theoretical addition effect changed with the concentration of BC powder and the volume fraction of CO_2 . For the max-pressure, when CO_2/BC with a CO_2 volume fraction of 8% was added, the actual max-pressure decrease was greater than the theoretical addition. For the time delay to max-pressure, the actual time delay was longer than the theoretical addition when the volume fraction of CO_2 was more than 6%. Furthermore, when the volume fraction of CO_2 was below 6%, the actual suppression effect of the CO_2/BC two-phase inhibitor was worse than the theoretical addition effect of the two single-phase inhibitors, which indicated that inert gas played a more significant role in the cooperative suppression effect on methane explosion. With the increase of inert gas in the gas-solid two-phase inhibitors, cooperative synergism exists between the inert gas and powders, which makes the effect of inert asphyxiation and dilution of methane more significant.



Figure 6. The difference between the actual effect and the theoretical addition effect of BC/CO_2 : (**a**) the difference of the max-pressure; (**b**) the difference of the time to max-pressure.

4. Suppression Mechanism of Gas-Solid Two-Phase Inhibitors

The experimental results showed that the gas–solid two-phase inhibitors have a significant suppression effect on methane explosions. The suppression mechanism can be explained as follows.

The inhibition effect of chemical powders: Firstly, the BC or ABC powders can absorb the amount of heat generated by the methane explosion reaction. Secondly, the ions decomposed from chemical powders can react with the explosion free radicals, which leads to a chemical suppression effect. That aside, the inert gas released from the decomposition of BC powders can dilute the concentration of oxygen and CH₄. It is why BC powders show better suppression performance than ABC powders.

The inhibition effect of inert gas: Firstly, inert gases in the two-phase inhibitors can dilute the concentration of CH_4 and O_2 . Secondly, inert gases can absorb part of the heat generated during the reaction process. In addition, the collision between inert gas molecular and the explosion free radicals results in the interruption of the methane chain reactions. In particular, CO_2 can participate in the chain reaction $H + CO_2 \rightleftharpoons OH + CO$, and consume H free radicals, which leads to a better suppression effect than N_2 on the methane explosion [34].

The cooperative inhibition effect of gas–solid two-phase inhibitors: The inhibition mechanism diagrammatic sketch is illustrated in Figure 7. When the gas–solid two-phase inhibitors are injected into the explosion space, two aspects of inhibition effects work simultaneously. The inert gases dilute the concentration of CH_4 and O_2 and the powders react with free radicals to interrupt the explosion

chain's development. Both the inert gases and chemical powders play a critical role in the whole explosion process. According to the test results, the two-phase inhibitors exhibited a more obvious suppression effect than the single-phase inhibitor.



Figure 7. The inhibition mechanism diagrammatic sketch of the gas-solid two-phase inhibitors.

5. Conclusions

The suppression effects of the gas–solid two-phase inhibitors on the methane explosion were investigated using a 20 L spherical explosion test system. Inert gases of N_2 and CO_2 , and chemical powders of ABC and BC were chosen in this study. Some qualitative conclusions can be drawn as follows:

The explosion suppression test results indicate that CO_2 presented a better inhibition effect than N₂, and BC powder showed better inhibitory performance than ABC powder. Compared with the single-phase inhibitor, the gas–solid two-phase inhibitors presented a better suppression effect. When the gas–solid two-phase inhibitors were injected into the explosion space, inert gas and chemical powders inhibited the explosion simultaneously. For gas–solid two-phase inhibitors, two-phase inhibitors of BC/inert gas exhibited more effective suppression than ABC/inert gas, and two-phase inhibitors of CO_2 /powders showed more effective suppression than N₂/powders. The 8% $CO_2/0.10$ g/L BC powders could suppress the methane explosion completely.

By analyzing the differences of the actual suppression effect of CO_2/BC powder and the theoretical addition effect of the two pure phase inhibitors, when the volume fraction of CO_2 was more than 6%, the actual suppression effect of CO_2/BC was better than the theoretical addition effect, indicating that the gas–solid two-phase inhibitors possessed a cooperative suppression effect on methane explosion. The cooperative suppression effect increases with the increase of CO_2 . Based on these experimental results, gas–solid two-phase inhibitors could be applied to the active explosion suppression device for gas explosion suppression. In future research, we will complete the simulation analysis about fluid dynamics and molecular dynamics to further explore the explosion suppression mechanism of gas–solid two-phase inhibitors.

Author Contributions: Y.W. conceived and designed the experiments; X.M., B.P., C.L., H.F., and L.Z. performed the experiments and analyzed the data; W.J. managed all the experimental and writing process as the corresponding authors; all authors discussed the results and commented on the manuscript.

Funding: This work was supported by the National Natural Science Foundation of China (51874120, 51674104, 51604095), Program for Science & Technology Innovation Talents in Universities of Henan Province (19HASTIT042), the Research Foundation for Youth Scholars of Higher Education of Henan Province (2017GGJS053), the Fundamental Research Funds for the Universities of Henan Province (NSFRF1606), Program for Innovative Research Team in University of Ministry of Education of China (IRT_16R22), Program for Innovative Research Team of Henan Polytechnic University (T2018-2), Foundation for Distinguished Young Scientists of Henan Polytechnic University (J2017-3) and the State Key Laboratory Cultivation Base for Gas Geology and Gas Control (Henan Polytechnic University) (WS2017A03).

Conflicts of Interest: The authors declare no conflicts of interest.

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