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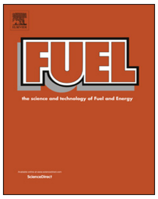

Full Length Article

Minimum ignition energy for the $CH_4/CO_2/O_2$ system at low initial temperature

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ABSTRACT

In this paper, the minimum ignition energy (MIE) for methane in an atmosphere of CO_2/O_2 is measured at 0.1–0.7 MPa and 183–273 K using a gas explosion experimental device that is able to withstand a temperature as low as 113 K. Effects of initial temperature (T_0) and initial pressure (P_0) on MIE are studied via experiment and a simple theoretical analysis. Results indicate that in our experimental setup, the sensitive conditions of MIE are an equivalence ratio of 1 and an electrode gap of 1 mm. Under low initial temperature, the trends in MIE with respect to initial pressure and temperature in an atmosphere of CO_2/O_2 are similar to those in an atmosphere of N_2/O_2. With an increase in initial pressure and temperature, MIE gradually decreases. MIE has a linear correlation with the reciprocal of the square of the initial pressure and the reciprocal of the initial temperature. At low initial temperature, P_0 has a large impact on MIE, whereas at low initial pressure, MIE is more sensitive to initial temperature. At the same initial temperature and pressure, MIE in an atmosphere of CO_2/O_2 is about 1.2 times larger than that in an atmosphere of N_2/O_2 atmosphere. CO_2 is more dilute than N_2 in accordance with large heat capacities and small thermal conductivities.

1. Introduction

With worldwide increasing energy demands and attention to environmental protection, as well as with an increasing shortage of oil and other traditional energy sources, developing and using liquefied natural gas (LNG) and oxygen-bearing coal-bed methane (CBM) has important economic and social benefits [1–6]. However, when LNG leaks occur, flammable mixed gas containing an LNG vapor cloud combines with air, which can cause fire and explosion hazards in the event of static sparks, mechanical sparks and fire. In addition, in the process of deoxidation of oxygen-bearing CBM at low temperatures, the methane concentration in the gas phase is within the explosion limit at the top of the distillation column, where the temperature is below 133 K and the pressure is in the range of 0.1–0.3 MPa [7,8]. If there is an energy greater than the minimum ignition energy (MIE), there is a risk of explosion in the distillation column, which is a huge safety hazard for the liquefaction of oxygen-bearing CBM. Therefore, taking necessary measures to inhibit methane explosion under low initial temperatures is significantly important in terms of safety.

There are many ways to suppress methane explosions to prevent fire and explosion hazards, such as application of an ultra-fine water mist of ultra-fine magnesium hydroxide powder and using an inert atmosphere of N_2 or CO_2 gas [9–12]. Of these approaches, application of inert gas such as N_2 or CO_2 is cost-effective and productive. In addition, MIE is a very important parameter for reflecting the flammable and explosive characteristics of combustible gases [13]. If external energy is lower than MIE, an explosion will not occur. Therefore, studying MIE under an atmosphere of an inert gas (N_2 or CO_2) is an important issue. MIE in atmosphere of air has been studied extensively via experiments, simulations, and theoretical calculations. Experimentally, Zhang et al. [14] studied effects of various initial temperatures on ignition characteristics for CH_4/air mixtures and found that the MIE of CH_4/air mixtures decreased with an increase in initial temperature. Coronel et al. [15] studied the MIE of lean H_2-N_2O mixtures using a short duration spark discharge. Effects of N_2 dilution and initial pressure on MIE have been determined. Zhang et al. [16] studied effects of nitromethane concentration on ignition energy of nitromethane/air mixtures, obtained accurate ignition energy values using their experimental setup, and determined the effects of nitromethane concentration on ignition characteristics. Cui et al. [13] experimentally studied the MIE of CH_4/air mixtures at elevated temperatures and pressures and determined the effects of initial temperature and pressure on MIE. Using simulations,

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Han et al. [17] studied spark ignition characteristics of a methane/air mixture using detailed chemical reaction mechanisms. The main parameters that affected the MIE that they studied are as follows: size of the electrodes, gap distance between the two electrodes, equivalence ratio, and duration of the spark. They also studied ignition behavior of a hydrogen/air mixture using the same numerical model in which heat loss was considered [18]. Effects of the energy supply procedure, radius of the spark channel, electrode size, and electrode gap distance on MIE were also determined. Using theoretical calculations, Kondo et al. [19] calculated MIE of premixed gases using two theoretical expressions. Calculation results were then compared with experimental data, and they found that the error in the calculation results of the two theoretical models was large. Coronel et al. [20] used a cylindrical kernel model to predict the ignition energy density of H_2-N_2O mixtures at various initial pressures. They found that there are 50%, 10%, and 14% differences between the cylindrical kernel model and the experimental data at initial pressures of 15, 20, and 25 kPa, respectively. Zhang et al. [21] used computational methods to study effects of diluents on the ignition of H_2/air mixtures. Effects of different diluents (He, Ar, N_2, and CO_2) on the ignition process of various H_2 fractions were determined. In summary, whether experiment, simulation, or theoretical calculation is used, the current research on MIE has mainly focused on an atmosphere of air with N_2 as an inert gas. Study of MIE in an atmosphere of CO_2 has rarely been reported.

Additionally, regardless of whether combustion or explosion is caused by LNG leaks or by liquefaction production of oxygen-bearing CBM, there is a common factor: the initial temperature is cryogenic. Under low temperatures, combustion properties differ from those at room temperature. Study on combustion characteristics of flammable gas under low initial temperatures has been carried out at home and abroad. Karim et al. [22] used a cylindrical stainless steel tube (5 cm diameter, 1 m long) to test the lower explosion limit of methane, hydrogen, and carbon monoxide at initial temperatures of 143–298 K and at atmospheric pressure. Wierzba et al. [23] used the same experimental setup and method to study the upper explosion limit of methane, ethane, propane, and mixed gas at temperatures of 213–298 K. Wierzba et al. [24] experimentally measured the explosion limits of hydrogen and some other fuels using an initial temperature range of 213–298 K for the upper explosion limit, and of 173–298 K for the lower explosion limit. Li et al. [25] constructed an explosion vessel with an inner diameter of 100 mm and length of 200 mm and experimentally measured explosion limits with different methane/nitrogen concentrations at an initial temperature of 150–300 K. In recent years, our research team built an explosion test setup that was able to withstand a temperature as low as 113 K and conducted a methane explosion experiment at different low initial temperatures of 123–273 K, different initial pressure of 0.1–0.9 MPa, and different equivalence ratios. The explosion limit and MIE of methane at low initial temperatures were obtained [26,27]. From the above studies, we can conclude that the existing explosion measurement at initial low temperature is based on air conditions (i.e., N_2/O_2 atmosphere). Study of MIE in an atmosphere of CO_2 at low initial temperature has not previously been conducted.

Therefore, in this paper, MIE of methane in an atmosphere of CO_2/O_2 is measured at 0.1–0.7 MPa and 183–273 K using the experimental gas explosion device in our laboratory. Then, effects of initial pressure and temperature on MIE are studied and discussed, and our results are compared with the existing experimental results and results of theoretical calculation.

2. Experimental

The experimental setup used in this paper is shown in Fig. 1. The experimental setup is identical to that used in [26,27] and is not described in detail here, although the general structure of the experimental setup is introduced. The experimental setup consisted of gas sources, an explosion vessel, refrigeration system, vacuum system, ignition system, and data acquisition system. The data acquisition system was set up for temperature acquisition, pressure acquisition, and MIE acquisition. The explosion vessel was a cylindrical stainless steel container with a volume of 2.35 L and pressure rating of 45 MPa. The refrigeration system was a refrigeration box, and the controlled temperature range was 103–313 K. The initial temperature was measured using a fast response thermocouple with a temperature range of 88–643 K and a response time of 20 μs. The explosion pressure was measured using a charge-type high-frequency pressure sensor with a pressure range of 0–34.5 MPa and a response time of 2 μs. The fast response thermocouple and pressure sensor maintained good performance at 113 K. The initial pressure was measured using a precision pressure gauge, and the pressure range was 0–1.6 MPa with an accuracy level of 0.02.

The MIE acquisition system is shown in Fig. 2; it mainly consisted of an ignition energy generator, ignition electrodes, oscilloscope, voltage probe, and current probe. Energy was generated using an ignition energy generator and released at the gap of the ignition electrodes. The ignition electrodes were two stainless steel electrodes, and the gap between them was continuously adjustable. During the spark release process, the voltage probe recorded variations in voltage between the two ignition electrodes with respect to time, and simultaneously, the current probe measured the current through the ignition electrodes. The voltage–time and current-time curves were recorded and displayed using an oscilloscope (Fig. 3). The actual MIE can be obtained by integrating the product of current and voltage with time (Eq. (1)). Refs. [13,27] show that the MIE calculated using this method is the actual spark energy, which is only about 10% of the stored energy in the ignition energy generator. With certain ignition energy and for 25 successive ignition tests, the combustible gas is considered to be successfully ignited during the MIE measurement even if the combustible gas explodes only once.

$$E = \int_0^t u(t)i(t)\mathrm{d}t \tag{1}$$

Here, E is MIE; $u(t)$ is the variation in voltage with respect to time; $i(t)$ is the variation in current with respect to time; t is the spark duration.

The gas preparation, accuracy test of gas concentration, ignition process, and measurement of MIE during the experiment are identical to those used in the literature [27]. The only difference is that the inert gas used in this experiment is CO_2 instead of N_2. The uncertainty of MIE in this study is consistent with that reported in the previous study (2%). The explosion vessel has already been validated, and repeating the work is not necessary here. It is worth noting that the boiling point of CO_2 is much higher than N_2. At low temperature and high pressure, CO_2 liquefies more easily. Therefore, in this study, the maximum initial pressure was 0.7 MPa, and the minimum initial temperature was 183 K.

3. Results

3.1. Sensitive conditions

In this experiment, there are many parameters that affect MIE, such as initial temperature, initial pressure, methane concentration, ignition voltage, capacitance, and electrode gap. Of the possible combinations of these factors, there is a set of conditions for which MIE is a minimum, and these conditions are called the sensitive conditions. Because initial temperature and pressure are variables, measurement of MIE is the actual spark energy, and thus, the only remaining sensitive conditions to determine are the equivalence ratio and electrode gap.

Variations of MIE for methane in an atmosphere of CO_2/O_2 with different equivalence ratios are shown in Fig. 4. The initial pressure is 0.1 MPa, the initial temperature is 213 K, and the electrode gap (d) is 1 mm. In Fig. 4, each data point is the average value of three parallel experimental results, and the error bars on the data points represent the