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Experimental study and theoretical calculation of flammability limits of methane/air mixture at elevated temperatures and pressures

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ABSTRACT

The flammability limits of methane/air mixture were experimentally studied at a temperature range of 30–150 °C and a pressure range of 0.1–0.9 MPa. The lower flammability limit (LFL) was calculated using a limiting flame temperature concept (White's role) and the temperature dependence of geometric mean G was also defined to predict the upper flammability limit (UFL). The results show that in our experiment, the influence of temperature and pressure on flammability limits show accordance with previous studies. The calculated LFL results using limiting flame temperature concept shows good agreement with the experimental values. The temperature dependence of geometric mean G remains unchanged at a certain pressure no matter what the temperature is. Within the temperature range examined in this paper, the UFL results can be predicted very accurately using the temperature dependence of geometric mean G. However, it is worth noting that with the increase of the initial temperature, the difference between experimental UFL and calculated UFL using geometric mean G becomes larger.

1. Introduction

Under normal conditions, the combustible gas and air mixtures which are in the flammability limit range have explosive properties. The upper flammability limit and low flammability limit are the maximum and minimum concentrations which can cause an explosion, respectively (Bolk and Westerterp, 1999). There are so many factors which have influence on the flammability limits (Takahashi et al., 1998, 2003; Kondo et al., 1999; Schoor et al., 2006; Li et al., 2011; Pekalski et al., 2005; Zhang et al., 2014; Schoorl et al., 2008): shape and size of the explosion vessel, type and location of the ignition electrode, ignition energy density, initial temperature and pressure, criterion of explosion, inert gases, and so on. The combustible gases are widely used in the chemical process. Therefore, in order to evaluate the explosion risk and guarantee safety production, it is necessary to study the flammability limits of the combustible gases under the work conditions (especially pressure and temperature) (Vanderstraeten et al., 1997).

As everybody knows, under normal pressure and temperature, the flammability limit range of methane is about 5–16% (volume fraction). Additionally, with the increase of temperature or pressure, the flammability limit range gets wider, i.e. the upper flammability limit increases and the lower flammability limit decreases. Many researchers have studied the flammability limits at various temperatures and pressures. White had proposed that the limit flame temperature remained unchanged no matter what the temperatures was (White, 1925). If it was the case, the lower flammability limits would be linear relationship with the temperature (Zabetakis et al., 1959; Zabetakis, 1965; Britton and Frurip, 2003). Zabetakis (1965) studied the flammability limits of saturated hydrocarbons. He found that the lower flammability limit had linear relationship with the temperature. Kondo et al. (2011) built up a 12 L explosion vessel and studied the flammability limits of several compounds at various temperatures and normal pressures. They found that both upper flammability limit and lower flammability limit had linear relationship with temperature. Schoor et al. (2008) simulated the upper flammability limit of methane/air mixtures using CHEM 1D. The initial temperature

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range is from 0.1 to 1 MPa, and the temperature range is from 25 to 200 °C. They found that at normal pressure, the simulation results were good enough. However, at elevated pressures, the simulated results were not satisfied. More important, up to now, the flammability limits of combustible gases calculated by most people are based on the empirical formula. Therefore, human factors dominate the calculated accuracy.

Based on the studies above, it is of particular interest to study the flammability limits of combustible gases at elevated pressures and temperatures. Additionally, it is necessary to propose an accurate calculation method to predict the upper flammability limits at elevated pressures. Therefore, in this paper, firstly, the flammability limits of methane/air mixtures at elevated pressures and temperatures are studied experimentally. The temperature range is from 30 to 150 °C and pressure range is from 0.1 to 0.9 MPa. Then, a calculation method of the flammability limits at elevated pressures and temperatures is proposed.

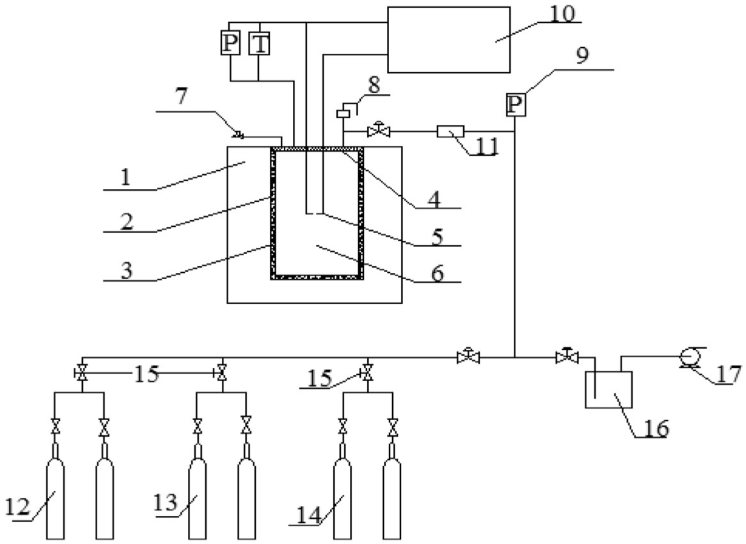
2. Experimental

The schematic diagram of the experimental system is shown in Fig. 1. It includes the gas cylinders, the explosion vessel, the heating system, the ignition system, the vacuum-pumping system and the data acquisition system. The explosion vessel is a vertical stainless steel cylinder which has a height of 400 mm, an inner diameter of 140 mm and a wall thickness of 18 mm. The pressure rating of the vessel is 15 MPa which is much higher than the explosion pressure at all temperature and pressure conditions in this experiment. The explosion vessel is placed in the heating box which has automatic temperature control function. The initial temperature of the combustible gas can be tested by a fast response thermocouple (Nanmac E Graduation S) fixed near the centre of the vessel.

The combustible gas is made by partial pressure method. Each component is added into the explosion vessel in series by regulating the needle valve. In this experiment, the methane is first added into the vessel until it reaches the required partial pressure, and then the nitrogen and oxygen are added subsequently. For the purpose of acquiring the accurate concentration of the prepared combustible mixture, a gas chromatography is used for analysis before ignition. The initial pressure is recorded by a precision pressure gauge that has an accuracy grade of 0.02. A spark is generated by two stainless steel electrodes mounted at the centre of the explosion vessel. The two electrodes are electrically insulated from the explosion vessel and have a gap distance of 5 mm. Then, spark energy of 10 J is generated, which is much higher than the minimum ignition energy of methane/air mixtures and thus, the influence of ignition energy on flammability limits can be avoided.

The explosion pressure and temperature curve are recorded by a piezo-electric pressure transducer (Dytran 2300V5) at a sample rate of 500 kHz and a fast response thermocouple (Nanmac E Graduation S), respectively. For the pressure transducer, the testing range is 0–34.5 MPa and the response time is 2 μs which is fast enough to record the pressure-time curve in the whole combustion process. For the thermocouple, the testing range is 0–1768 °C and the response time can be as high as 20 μs. In this study, the explosion is considered successful when the pressure increases higher than 5% of the initial pressure according to the BS-EN-1839-2003 standard. Once an experiment is finished and before the next combustion, firstly, the explosion vessel is flushed with nitrogen 3 times of the vessel volume and then, vacuumized by the vacuum pump. Therefore, the influence of the gaseous product can be avoided. In order to make sure that the combustible gases are homogeneous and motionless, a residence time of 5 min is needed before ignition. Additionally, in order to explain the experimental repeatability, three sets of parallel experiments must be done to measure the flammability limit at each condition (Tang et al., 2014).

In this experiment, the initial temperatures are chosen from 30



1-Heating box, 2-Inner wall of the heating box, 3-Aluminium powder, 4-Insulating layer, 5-Ignition electrode, 6-Explosion vessel, 7-Sampling valve, 8-Safety valve, 9-Precision pressure gauge, 10-Data acquisition system, 11-Flame arrester, 12-Methane gas cylinders, 13-Oxygen gas cylinders, 14-Nitrogen gas cylinders, 15-Needle valve, 16-Vacuum vessel, 17-Vacuum pump

Fig. 1. Experimental setup.