Explosion regions of propane, isopropanol, acetone, and methyl acetate/inert gas/air mixtures

Aksam Abdelkhalik [a, *], Enis Askar [b], Detlev Markus [c], Elisabeth Brandes [c], Ibrahim El-sayed [d], Mohamed Hassan [a], Mohamed Nour [a], Thomas Stolz [c]

[a] National Institute for Standards (NIS), P.O. Box 136, Giza, 12211, Egypt
[b] Bundesanstalt für Materialforschung und-prüfung (BAM), Unter den Eichen 87, 12205, Berlin, Germany
[c] Physikalisch Technische Bundesanstalt (PTB), Bundesallee 100, 38116, Braunschweig, Germany
[d] Menoufia University, Faculty of Science, Department of Chemistry, El-Menoufia, Egypt

ARTICLE INFO

ABSTRACT

The explosion regions for propane, isopropanol, acetone, and methyl acetate with air in the presence of nitrogen, argon, helium, and carbon dioxide were determined experimentally according to EN 14756/EN1839, method T. Except for propane, all the measurements were executed at 323 K and 1 bar. Propane experiments were carried out at 293 K and 1 bar. The results show that for the same type of inert gas, propane, isopropanol, and acetone have great closeness concerning the concentration of the inert gas at the apex of the explosion envelope in a ternary diagram with air as oxidizer. This leads to consistency in the limiting oxygen concentration (LOC) and minimum required amount of inert gas (MAI) values. Concerning methyl acetate, the apex was always reached at higher percentages of inert gases compared with the other fuels. This can be attributed to the presence of two oxygen atoms inside the chemical structure. Calculation of the explosion regions was carried out based on calculated adiabatic flame temperature (CAFT) method. The flame temperatures for the experimentally determined fuel/air/N_2 mixtures were calculated. Then, these temperatures were used to predict the explosion limits of similar mixtures with other inert gases than nitrogen. The modeling results show reasonable agreement with the experimental results.

1. Introduction

The explosion limits of chemical vapors and gases still attract researchers' attentions in the academic and industrial sectors. This is due to their importance for safe handling, and setting up of safe chemical processes. Explosion limits define the concentration range of combustible and oxidizer mixtures (explosion range), within which an explosion may occur. The upper explosion limit (UEL) is the upper limit of the explosion range and the lower explosion limit (LEL) is its lower limit (EN1839, 2010). The explosion limits are not part of the explosion range.

Propane, isopropanol, acetone, and methyl acetate are widely used in industrial processes. They have the same number of carbon atoms and are able to form explosive mixtures with air or other oxidizers. The UEL and LEL values for these substances in air are collected by Coward and Jones (1952), Zabetakis (1965), Kuchta (1985) and Brandes and Möller (2008). One means to avoid formation of explosive mixtures with an oxidizer in the gas phase of these substances is to add inert gas. These gases are added in such quantities that the concentration of oxygen in the mixture is reduced to below the respective limiting oxygen concentration (LOC) (Planas-Cuchi et al., 1999; Crowl and Louvar, 2002; Molnárné et al., 2008; Chen et al., 2009). Molnárné et al. (2008) discussed the effect of adding different inert gases to propane/air mixtures on the respective explosion ranges. They also stated the effect of temperature and calculated other safety characteristic data which are of interest for inerting processes like maximum permissible flammable gas concentration (MXC), minimum inert gas/air ratio (IAR), LOC, MAI, and minimum inert gas/flammable gas ratio (ICR). The complete definitions and the way of calculating these terms are

presented by Molnárné et al. (2008), and Molnárné and Schröder (2011). Furthermore, Zabetakis (1965) recorded the explosion limits of propane, acetone and methyl acetate in the presence of N_2 and CO_2 as inert gases. Coward and Jones (1952) presented the effect of CO_2 and N_2 addition on acetone-oxygen and propane/air mixtures.

The data about the inerting for propane/air mixtures is much discussed in literature. However, the information about acetone, isopropanol, and methyl acetate which have the same number of carbon atoms as propane is still limited. Moreover, the comparison of the explosion limits and explosion regions in the presence of different inert gases of these substances will help in identifying the effect of oxygen content inside the chemical structure on the inerting process.

Calculated adiabatic flame temperature (CAFT) is a well known method for predicting explosion limits for many combustible materials. The CAFT values for UEL and LEL can be used to predict mixture compositions along the explosion region boundary curve in a ternary system of fuel, inert gas and oxidizer. The CAFT method was suggested by White (1925) and then used by many authors to predict UEL, LEL and the whole explosion region boundary curve for different fuel/oxidizer/inert gas mixtures (Hertzberg et al., 1985; Hansel et al., 1992; Melhem, 1997; Mashuga and Crowl, 1999; Brooks and Crowl, 2007a,b; Askar et al., 2008; Du et al., 2015).

The aims of our work are, first, to study the explosion region of isopropanol, acetone, and methyl acetate/air mixtures in the presence of N_2, CO_2, He, and Ar; second, to compare the results with the data available for propane/air/inert gas mixtures and, moreover, to present the effect of oxygen content inside the chemical structures on the explosion regions; and third, to calculate explosion limits with the CAFT method.

2. Experimental set up and procedure

2.1. Materials

The purity of all gases (propane, CO_2, N_2, He, and Ar) was at least 99.9%. The compressed air (O_2 (20.95%), N_2 (78.09%), CO_2 (0.03%), Ar (0.933%)) used for the measurements was dried by silica gel. Acetone, isopropanol, and methyl acetate had a purity of 99%. The liquids were used without any further purification process.

2.2. Glass tube apparatus

The explosion limit measurements were carried out according to EN1839 method T (EN1839, 2010). Fig. 1 shows the schematic diagram for the device used. A glass test tube having a diameter of 80 ± 2 mm and a length of 500 mm was used. The tube was closed at the bottom end and vented from the upper end. The test vessel was placed vertically in a heating chamber. The temperature of the heating chamber can be adjusted in the range from 293 K to 573 K with an uncertainty of 1 K. The ignition source was a series of induction sparks between two electrodes. The electrodes were positioned at 60 ± 1 mm from the bottom of the test tube. The distance between the electrodes was 5 ± 1 mm. The spark discharge time was 0.2 s. The induction sparks were generated by a high voltage transformer (15 kV). Caloric and electric measurements showed that such an ignition source generates sparks in air having a power of 10 W. The criterion for an explosion was the detachment and upward movement of a flame for at least 100 mm from the ignition source. The mixture was considered to be non-flammable when the flame detachment from the ignition source and propagation over 100 mm failed in five tests. The mixture was prepared using calibrated mass flow controllers. Before each ignition trial the vessel was purged with the test mixture by ten times its volume.

The purging mixture and combustion products left the test tube through the lid. After the test mixture became quiescent inside the test tube, ignition attempts were started. For each fuel concentration, the inert gas concentration was selected and changed by $\pm$ 0.5 mol percent until the mixture was just non-flammable. All measurements of propane were carried out at 293 K and 1 bar; isopropanol, acetone, and methyl acetate measurements were performed at 323 K and 1 bar due to the vapor pressures of the substances. The uncertainty of measurements according to EN1839 standard test method is $\pm 10\%$ relative for the molar percentage of the test substance $\leq 2\%$, and $\pm 0.2\%$ absolute for the molar percentage of the test substance $>2\%$. Fig. 1 describes the device setup used with liquid substances. However, in the case of propane mixtures the evaporator tube was removed and the pump was replaced by a filled propane cylinder and mass flow controller.

3. Results and discussion

3.1. LEL and UEL values for the selected combustibles

The data in Table 1 show the experimental results of this work and some literature values for UEL and LEL of the selected fuels in air. They are given in detail to judge possible differences between the literature data and ours, which are determined using the new standard EN 1839. Propane results at 293 K are 1.8% for LEL and 10.7% for UEL. These values agree with the values obtained by Molnárné et al. (2008). The LEL of isopropanol 2.1% shows satisfactory agreement with the literature data. In contrast, the UEL value 12.8% shows a certain difference from the literature data. Acetone findings for LEL and UEL are 2.2%, and 14.0%. These results show certain differences with regard to the literature data. Methyl acetate results are 2.6% and 16.0% for LEL and UEL. The LEL result is different from the literature. However, there is good agreement between the experimentally determined UEL value and the data recorded by Zabetakis (1965) and Brandes and Möller (2008). For better comparison the literature data if determined at temperatures other than 323 K were calculated for 323 K using to the following equation

$$\chi_{FG}(T) = \chi_{FG}(T_0)[1 \pm K(T - T_0)], \tag{1}$$

where $\chi_{FG}(T)$ is the mass or volume fraction from flammable gas at the explosion limit for a temperature T, and $\chi_{FG}(T_0)$ is the fraction at reference temperature T_0. The positive sign is used in calculating the UEL, and the negative sign at the LEL. The values for the factor K is taken from Hirsch and Brandes (2015). According to Equation (1), the difference in temperature by 303 K doesn't have a significant effect on the explosion limits of the selected combustibles in air.

The variation between the experimental and literature results is due to the differences in the test tube dimension, criteria of the explosion, and the test procedure applied. The evidence for this assumption is the experimental results for the LEL and the UEL of propane which are obtained at 293 K. It shows good agreement with data which are obtained under the same conditions and with the same standard test method (Molnárné et al., 2008). The difference in results is $\pm 0.1\%$ for the LEL and UEL. This value, $\pm 0.1\%$, lies within the permissible uncertainty in measurements according to the EN1839 standard test method.

3.2. Effect of inert gases

At first the effect of the different inert gases CO_2, He, N_2, and Ar on the explosion region is discussed in detail taking acetone as an example of the combustible materials. Following the effect of each inert gas on the four combustible materials are compared.