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Flammability limits of binary mixtures of dimethyl ether with five diluent gases

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ABSTRACT

Flammability limits of binary mixtures of dimethyl ether with five kinds of diluent gases were measured by ASHRAE method at room temperature. The five diluent gases are nitrogen, carbon dioxide, chlorodifluoromethane (HCFC-22), 1,1,1,2-tetrafluoroethane (HFC-134a) and 1,1,1,2,3,3,3-heptafluoropropane (HFC-227ea). The experimental results were correlated with the extended Le Chatelier's formula. It was found that the experimental results were well reproduced by the formula. In addition, flammability limits of binary mixtures of dimethyl ether with nitrogen and carbon dioxide were compared with the estimated values based on the adiabatic flame temperature method. The experimental results were found to be in satisfactory agreement with the estimated values.

1. Introduction

Dimethyl ether (DME) is an excellent industrial chemical. It is often used as propellant gas, assist solvent, blowing agent, fuel additive, liquefied petroleum gas substitute etc. In addition, dimethyl ether is an ideal alternative refrigerant because it is extraordinary environment friendly owing to a zero ozone depression potential (ODP) and negligible global warming potential (GWP). However, dimethyl ether is extremely flammable like hydrocarbons. Security issues contribute a prominent constraint to the application of dimethyl ether. Thus, dimethyl ether was generally used together with nonflammable refrigerants. The mixture of 1,1,1,2-tetrafluoroethane (HFC-134a) + pentafluoroethane (HFC-125) + DME was named R419 (Valtz, Gicquel, Coquelet, & Richon, 2005). Bi et al. found that the performance of dimethyl ether with carbon dioxide (Bi, Chen, Wu, & Liu, 2009) or 1,1,1,2,3,3,3-heptafluoropropane (HFC-227ea) (Bi, Chen, Wu, & Zhou, 2009) is more excellent compared to chlorodifluoromethane (HCFC-22) under the same conditions. The diluent effect of HFC-125 on the flammability of dimethyl ether was investigated in our previous work (Zhang, Wu, Gao, & Xue, 2010). However, the flammability limits of the mixtures of dimethyl ether with other diluent refrigerants remain unknown. This work reports the flammability limits of binary mixtures of dimethyl ether with HFC-134a, HFC-227ea, HCFC-22, nitrogen and carbon dioxide, and compares the obtained results with the estimated values and the correlated data.

2. Experimental method

2.1. Apparatus

The measurements of flammability limits were performed essentially by ASHRAE method, which is a revised version of ASTM E681 (2004). The explosion vessel was a 12-L spherical glass flask. The flask was settled in a chamber which was kept at room temperature of (23 ± 2) °C. Schematic diagram of the experimental system is described in Fig. 1. Gas mixtures were directly prepared in the explosion vessel by the partial pressure method, and the progress was realized using solenoid valves controlled by a software developed under the environment of Microsoft Visual C++. The pressure was measured with a transducer MPM 4730 (Micro Sensor Co., Ltd., China, ±0.1% FS). The transducer was calibrated by a Paroscientific digiquartz pressure sensor (Model 223A-101) with an accuracy of ±0.01% FS. Ignition of the mixture was attempted by a 15 kV neon transform attached to a pair of tungsten electrodes 6.4 mm apart, with a spark duration limited to 0.4 s. The electrodes were positioned at one third of the diameter of the flask from the bottom of the flask. Whether the mixture was flammable or not was judged by the ASHRAE 90° flame propagation criterion (Rowley, Rowley, & Wilding, 2010).

In the experiments, the flask was evacuated below 200 Pa before introduction of the samples. The system was capable of maintaining a vacuum within 100 Pa in 10 min. Then the samples and air

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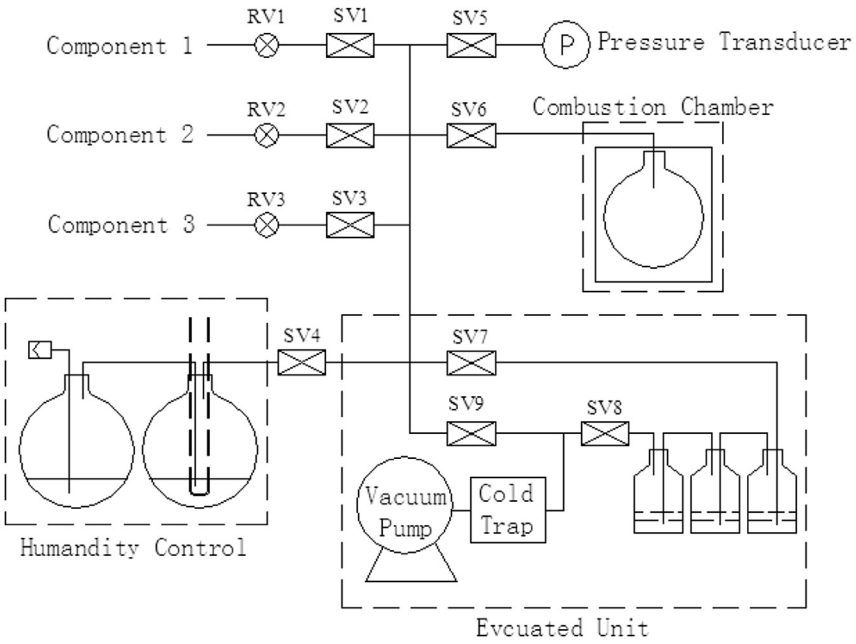

Fig. 1. Schematic diagram of experimental system: SV1–SV9, solenoid valves; RV1 ~ RV3, gas regulators.

were introduced into the flask in sequence. The prepared mixture was agitated with a magnetic stirrer for at least 5 min to obtain complete mixing. Shortly before ignition, the stirrer was turned off and the mixture was left for 1 min to eliminate turbulence, and the clamps on the cover were released. The combustion exhaust gas was filtered after ignition through three bottles with dilution alkali solution. A cold trap with liquid nitrogen was installed to prevent evacuating the water vapor to the vacuum pump. This process was repeated, varying the volume ratio of the sample until a flame fulfilled the flame propagation criterion, and the volume ratio steps were 0.05 vol% for the lower flammability limit (LFL) and 0.2 vol% for upper flammability limit (UFL). The uncertainties of the experimental results were estimated considering the volume ratio step and the flame propagation angle of the mixtures at different concentration in air.

2.2. Validation of the apparatus

Performance of the apparatus was validated by determining the LFL and UFL of HFC-152a, HFC-32 and HC-600a. The experimental results were listed in Table 1 together with the reference values. The average absolute deviation of the experimental results of the LFL and UFL from reference values was 0.23 vol% for the three refrigerants. The maximum deviation was 0.7 vol% which was the situation of the lower flammability limits of HFC-32. Deviations of the experimental results from the reference values were mostly under the uncertainty of each measurement, which indicated the reliability of the apparatus.

Table 1
Comparison between the experimental results (*exp*) and references values (*ref*) for HFC-152a, HFC-32 and HC-600a.

Fuel		Flammability limits (vol%)		
		exp	*ref*	*exp* − *ref*
HFC-152a	LFL	4.42 ± 0.08	4.32[a]	0.10
	UFL	17.6 ± 0.4	17.35[a]	0.25
HFC-32	LFL	13.7 ± 0.4	13.0[b]	0.7
	UFL	27.2 ± 0.6	27.0[b]	0.2
HC-600a	LFL	1.78 ± 0.04	1.80[c]	−0.02
	UFL	8.50 ± 0.18	8.40[c]	0.10

[a] Kondo, Takizawa, Takahashi, Tokuhashi, and Sekiya (2008).
[b] Kondo et al. (2006).
[c] Grosshandler, Donnelly, and Womeldorf (2000).

3. Results

Flammability limits for pure dimethyl ether were measured, and the results were listed in Table 2. The lower flammability limit of dimethyl ether reported by different authors was in good agreement with each other, while large deviation was shown with regard to the upper flammability limit. Jones and Scott (1947) found that the upper flammability limit of dimethyl ether was related to the experimental methods. It was reported that with upward propagation of flame in a tube 5 cm in diameter, opened at the firing end, the upper flammability limit is 18.2 vol%. When the firing end is closed and ignition is caused by a heated platinum wire instead of a flame, the upper flammability limit becomes 26.7 vol% and the propagation is by "cool flame". In narrow tubes of 1–2.5 cm in diameter, the flammability range was (3.93–16.6) vol%. Kondo, Takizawa, Takahashi, and Tokuhashi (2011) measured the flammability limits of dimethyl ether by ASHRAE 12-L method, the upper flammability limit of 25.2% was obtained at the temperature of 20 °C. In this work, pale blue flame was observed near the upper flammability limit, which indicated the existence of the effect of cool flame upon the upper flammability limit. ASHRAE 90° criterion was strictly used to determine the flame propagation, the results of

Table 2
Literature values of flammability limits for pure dimethyl ether.

LFL (vol%)	UFL (vol%)	References
3.42	21.0	This work
3.45	18.2	Jones and Scott (1947)
3.45	26.7	Jones and Scott (1947)
3.93	16.6	Coward and Jones (1952)
3.35	25.2	Kondo et al. (2011)
3.4	17	Granryd (2001)
3.4	18	Bhide et al. (2003)
3.4	27.0	NFPA 325 (1994)