



A model for predicting temperature effect on flammability limits



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ABSTRACT

A model for predicting the variation of flammability limits by the initial temperature was derived from the energy balance equation. All parameters of this proposed model can be sourced from the literature and do not need to be regressed by any experimental data. The proposed model was validated using 20 sets of experimental data consisting of 11 compounds, including alkanes, alkenes, alcohols, a ketone, an ester and inorganic fuels. Some samples were measured experimentally and other sample data were adopted from the literature. Overall, the proposed model predicted the temperature variation data of the flammability limits well when based on the adiabatic assumption. Application of this model is not restricted to organic fuels, but also inorganic fuels, such as carbon monoxide and hydrogen.

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1. Introduction

Explosions blamed on the leakage of flammable gas or vapor may result in industrial losses and public catastrophe, such as the gas explosions in Kaohsiung, Taiwan, which killed 32 and injured 321 people in 2014 [1,2]. Gas and vapor can be ignited and burned in the air only between the composition range of the upper and lower flammability limits, the UFL and LFL, respectively [3,4]. Flammability limits are the properties most frequently used in assessing the flammability hazards of gases and vapors.

The tested values of flammability limits for a given fuel are not constants. They are associated with a variety of external factors, such as the scale and configuration of the test apparatus, initial temperature and pressure, the presence of inert gases, and others [3,5]. In many commercial processes, such as oxidation processes, air or oxygen and fuel coexist in one system, and that create the potential hazards of fire and explosion. Published data for flammability limits were generally tested at the ambient temperatures, such as the flammability limits reported by Coward and Jones [6]. However, the process temperature is always highly apart from the ambient temperature. The UFL and LFL increase and decrease with the initial temperature, respectively [6]. Thus, process designs based on the flammability limits conducted at ambient temperature could result in a potentially serious risk of fire and explosion

for processes with process temperature greater than the ambient temperature. In fact, such process designs were found for some oxidation processes in Taiwan. A model to upgrade the flammability limit data originally based on ambient temperatures to those at process temperatures is critical in practice.

Based on the experimental observation of paraffins, from methane to decane, Zabetakis used an empirical linear equation to describe the variation of LFL with initial temperature [7]. These data adhered well to the modified Burgess–Wheeler law [8]. Zabetakis also applied the law to the analogous UFL data [7]. Vanderstraeten et al. described the temperature variation data of the UFL for methane with a linear equation and determined the slope of the equation by regressing it against experimental data [9]. Van der Schoor and Verplaetsen extended the work of Vanderstraeten et al. to ethane, ethylene, propane, propylene and *n*-butane [10]. Based on the assumption of the LFL approaching zero at the lower limit flame temperature, proposed by Zabetakis [7], Britton and Frurip suggested that the LFL was linear within the difference between the lower limit flame temperature and initial temperature [11]. Catoire and Nauder suggested an experimental formula to estimate the LFL of fuels made up of the elements C, H, O, and N at different temperatures [12].

Theories relating flammability limits to heat loss indicate that the flame temperature for a given fuel should be greater than the minimum temperature for the flame to propagate [13–15]. Application of the flame temperature, which is equivalent to such a minimum temperature, in the energy balance explains the critical behavior of combustion, such as flammability limits [16]. The assumption that the adiabatic flame temperature remains

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Nomenclature

A_s	heat transfer surface area per mole mixture, $\text{m}^2 \text{mol}^{-1}$
C_p	heat capacity, $\text{J mol}^{-1} \text{K}^{-1}$
e	emissivity
h	specific enthalpy, J mol^{-1}
L	lower flammability limit
N	number of experimental data
n	mole, mol
Q_r	heat loss through thermal radiation, J mol^{-1}
T	temperature, K
T_0	a specific temperature, K
T_i	initial temperature, K
T_L	flame temperature at LFL, K
T_U	flame temperature at UFL, K
U	upper flammability limit

Greek letters

α	radiation heat transfer surface area efficiency factor
ΔH_{298}°	standard reaction heat at 298 K, J
$\Delta h_{\text{H}_2\text{O}}^\circ$	standard heat of formation of H_2O , J mol^{-1}
Δh_c°	standard combustion heat for a fuel, J mol^{-1}
Δt	the duration of flame propagation, s
σ	Stefan–Boltzmann constant, $\text{W m}^{-2} \text{K}^{-4}$

Subscripts

c	combustion
$exp.$	experimental data
f	fuel
i	species i
$pred.$	predictive value

constant for a given fuel, which does not consider heat loss, was frequently used in the estimation of the flammability limits of fuel-inert mixtures [17–23]. Previously, a model assuming a constant flame temperature, with heat loss taken into account, was developed for approximating the flammability limits of fuel and inert admixtures [24]. Now, a model to predict the variation of flammability limits by initial temperature is proposed, based on the assumption of a constant flame temperature, and validated with experimental data, data adopted from the literature or measured in this study. The fuels included methane, propylene, butylene, and hydrogen (H_2).

2. Experimental procedures

2.1. Materials

Methane (99.50%), propylene (99.50%), butylene (99.50%), and hydrogen (99.0%) were purchased from Jing De Gases Co. (Taiwan).

2.2. Spherical glass flask

A 5-l spherical glass flask, which was designed to meet the requirement of ASTM E681 [25], was employed to test the flammability limits of various fuel gases (methane, propylene, butylene, and hydrogen) mixed with air at different initial temperatures. The system diagram of the 5-l spherical glass flask is shown in Fig. 1. The 5-l flask was sealed to keep it at atmospheric pressure with water and was placed in an air bath to control the temperature of the gas mixture. The ignition source, a spark from a 16–18 kV power supply with spark duration of 0.5 s, was placed in the center of the sphere.

The apparatus operation was conducted according to the ASTM E681 [25] and E918-83 [26] standards. Except for hydrogen, the determination of whether a mixture was flammable or not was assessed according to the ASTM E681 standard [25]. Flammable was defined as “flames that spread upward and outward to the walls of the flask, continuously along an arc greater than that subtended by an angle equal to 90° , as measured from the point of ignition to the walls of the flask” [25]. In the case of hydrogen, the flame is invisible. Thus, flammable was defined as a pressure increase that pushed the water seal up to 54 cm from its original location and outside the water sealed hose.

The composition of the gas mixture was determined using a partial pressure procedure (sensitivity of the pressure sensor = 0.07 torr). After the evacuation of the spherical flask (maximum vacuum = 3 torr), the flask was flushed with air three

times before loading the fuel. The air was then introduced, and finally the air bath was used to control the temperature of the gas mixture.

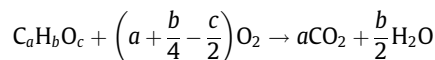
3. Theory

For a constant pressure flammability limits test apparatus, such as the vertical tube, spherical glass flask, the energy balance equation is described as [24]:

$$\Delta H_{298}^\circ + \sum_{\text{reactants}} \int_{T_i}^{298} n_i C_{p_i} dT + \sum_{\text{products}} \int_{298}^T n_i C_{p_i} dT = Q_r \quad (1)$$

where T_i and T are the initial and final temperatures of the mixture before and after burning.

In this study, fuel compounds made up of C, H, and O were taken into account. Because of sufficient oxygen to transform the fuel ($\text{C}_a\text{H}_b\text{O}_c$) completely into carbon dioxide (CO_2) and steam (H_2O), the combustion stoichiometry at the LFL is described as [24]:



Based on expending 1 mol of the combustion blend at the LFL, the quantity of the components after and before burning is listed in Table 1. The standard reaction heat at 298 K is [24]:

$$\Delta H_{298}^\circ = L(\Delta h_c^\circ) \quad (2)$$

and

$$\begin{aligned} \sum_{\text{reactants}} n_i C_{p_i} &= L C_{p_f} + 0.79(1-L) C_{p_{\text{N}_2}} + 0.21(1-L) C_{p_{\text{O}_2}} \\ &= L(C_{p_f} - P_L) + P_L \end{aligned} \quad (3)$$

$$\begin{aligned} \sum_{\text{products}} n_i C_{p_i} &= 0.79(1-L) C_{p_{\text{N}_2}} + [0.21(1-L) - f_L L] C_{p_{\text{O}_2}} \\ &\quad + g_L L C_{p_{\text{CO}_2}} + h_L L C_{p_{\text{H}_2\text{O}}} \\ &= L[-0.79 C_{p_{\text{N}_2}} - (0.21 + f_L) C_{p_{\text{O}_2}} + g_L C_{p_{\text{CO}_2}} + h_L C_{p_{\text{H}_2\text{O}}}] \\ &\quad + [0.79 C_{p_{\text{N}_2}} + 0.21 C_{p_{\text{O}_2}}] = L(Q_L - P_L) + P_L \end{aligned} \quad (4)$$

where

$$f_L = a + \frac{b}{4} - \frac{c}{2}$$

$$g_L = a$$

$$h_L = \frac{b}{2}$$

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