

Development of Calculated Non-Adiabatic Flame Temperature Model to Predict Flammability Limits of H₂-Air-Diluent Mixtures Based on Heat Transfer Mechanism

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1. Introduction

The lower flammability limit (LFL) of hydrogen is of considerable interest in the nuclear industry because of the potential hydrogen risk as a consequence of severe accidents [1]. If the released hydrogen gas exceeds LFL, a more detailed risk analysis of the plant will be required to verify the potential possibility of flame acceleration and detonation. [2]. Although many experimental studies have been conducted to determine LFL of various mixtures extensively, it is still difficult to identify the limits of all possible mixture conditions under a nuclear reactor severe accident. For this reason, a theoretical model for predicting LFL is needed to evaluate the hydrogen risk according to mixture conditions such as mixture compositions and different initial temperatures.

Through numerous studies [3-5], it was found that the calculated adiabatic flame temperature (CAFT) remains nearly constant regardless of the properties of limiting mixtures if fuel type is the same. These studies concluded that the threshold flame temperature for flame propagation is proportional to the CAFT. This concept was deemed to yield reasonable estimates for a variety of mixtures and initial temperatures. However, this concept exhibits limited accuracy depending on the mixture condition as shown **Table 1**. The limitation of the CAFT model deviates from the assumption on an adiabatic condition, whereas the actual flame propagation involves the heat loss mechanism. Thermal theories of the flammability limits of fuels have attributed the existence of these limits to heat loss from the reaction zone. The amount of heat loss affects the temperature of the reaction zone and hence determines the intrinsic LFL value of the mixture.

Therefore, the objective of this study is to develop the calculated non-adiabatic flame temperature (CNFT) model, which relaxes the assumption of adiabatic condition. The model facilitating prediction of the LFL of hydrogen mixtures assumes combustion in a non-adiabatic condition considering heat loss due to the radiative heat transfer. It is because the radiative heat transfer dominantly determines the total amount of the heat loss from reaction zone to environment. It is assumed that, if fuel type is the same, the CNFT would remain constant regardless of the property of limiting mixtures. The verification of the model was based on an upward flame propagation experiment. The experiment used a similar apparatus to that used by the US Bureau of Mines, with a 51 mm diameter and a one-meter length of pipe.

2. Modeling

2.1 Adiabatic flame temperature

The adiabatic flame temperature can be calculated from the energy balance of the reaction at equilibrium as shown **Equation (1)**. Where $\Delta H_{f,i}^0$ is formation enthalpy and T_i is initial temperature of mixture. Previous studies concluded that the threshold flame temperature for flame propagation is proportional to the calculated adiabatic flame temperature (CAFT). This concept was reported to yield reasonable estimates for a variety of mixtures and temperatures [3-5].

$$\sum n_i [\Delta H_{f,i}^0]_{\text{reactants}} - \sum n_i [\Delta H_{f,i}^0 + \bar{c}_{p,i} (T_{\text{CAFT}} - T_{\text{initial}})]_{\text{products}} = 0 \quad (1)$$

The experimental flammability limits of various hydrogen-diluent fuel mixtures in air for upward propagation at ambient pressure were determined by Terpstra [6]. The experiment used a similar apparatus to that used by the US Bureau of Mines, with a 51mm diameter and a 1 m length. These dimensions allowed the experimenter to observe the widest flammability limits that would propagate on their own in air while minimizing the influence of the ignition source. The flammability limits were measured at constant pressure to ensure that the mechanisms of flame propagation remained constant as the flame traveled the length of the tube.

Table 1 shows the CAFT for each condition of limiting mixture determined by Terpstra. Although the temperature remains nearly constant for some mixtures around 600 K, other mixtures show significant differences. This is especially true when the initial temperature is high or the diluent gas is helium. As a result, it was confirmed that the CAFT model showed limited accuracy depending on the mixture condition. The limit of the CAFT model can be inferred from **Equation (2)**. In real mechanism of flame propagation, the flame does not propagate under adiabatic conditions and it involves heat loss process.

$$dh = du + pdv + vdp = q + vdp = Q_{\text{loss}} \quad (2)$$

Table 1. CAFT for each limiting mixture

Mixture	T_i (K)	H ₂ vol%	Diluent vol%	$T_{adiabati}$ (K)
H ₂ -Air	293	3.90	0	581
H ₂ -Air	323	3.80	0	604
H ₂ -Air	373	3.60	0	639
H ₂ -Air	423	3.30	0	667
H ₂ -Air	473	2.80	0	680
H ₂ -Air	573	2.40	0	750
H ₂ -Air-He	293	3.84	0.96	583
H ₂ -Air-He	293	3.96	2.64	594
H ₂ -Air-He	293	4.00	4.00	598
H ₂ -Air-He	293	4.00	6.00	601
H ₂ -Air-He	293	4.38	17.52	645
H ₂ -Air-He	293	5.30	47.7	777
H ₂ -Air-Ar	293	3.84	0.96	583
H ₂ -Air-Ar	293	3.80	3.80	583
H ₂ -Air-Ar	293	3.70	14.80	587
H ₂ -Air-Ar	293	3.45	31.05	586
H ₂ -Air-Ar	293	3.00	57.00	579
H ₂ -Air-N ₂	293	3.92	0.98	588
H ₂ -Air-N ₂	293	3.90	3.90	586
H ₂ -Air-N ₂	293	3.90	15.60	587

2.2 Effects of heat transfer mechanism on lean flammability limit

In many previous studies, it has been suggested that a threshold flame temperature must be reached in the flame front in order for the premixed flame to propagate [3-5]. Zel'dovich explained that the actual flame speed at flammability limit cannot be zero but rather it has to take a finite value [7]. The finite value can be calculated from the Davies and Taylor [8]. Experimental results proved that an upward propagating flame at the limit of flammability shared the same properties with a rising Taylor bubble [9]. Because the actual speed is a strong function of the peak temperature, the prediction of the peak temperature can identify the flammability of gas mixture.

The heat loss from reaction zone to post-reaction zone affects substantially in determining the peak temperature. If there is no heat loss, all the combustion heat will be transferred to the unburned gas during flame propagation. **Equation (3)** shows the energy balance equation in the flame front for the adiabatic flame [10]. Where $\rho_u S_u^a$ is the adiabatic mass flux, c_p is the gas specific heat, H^a is the energy released per unit mass of the mixture, T_u is the unburnt mixture temperature and T_f^a is the peak flame temperature. Changes in the specific heat were assumed to be negligible. In this case, the peak temperature of the flame can continue to increase once the heat above the activation energy is supplied.

$$\rho_u S_u^a [c_p T_u + H^a] - \rho_u S_u^a c_p T_f^a = 0 \quad (3)$$

If the heat loss to post-reaction zone is taken into account, the peak temperature assumed to occur near the end of the reaction zone can be calculated by an energy balance equation considering the heat loss as shown **Equation (4)**. Where the q_{loss} is the heat loss rate per unit area of the flame front. S_u is the actual flame speed. The particular heat loss mechanisms of essential in the application of **Equation (4)** to the prediction of the flammability limits are convective and radiative heat transfer from a flame to its environment [10].

$$\rho_u S_u [c_p T_u + H^a] - \rho_u S_u c_p T_f - q_{loss} = 0 \quad (4)$$

However, convective heat loss can be ignored if the tube diameter exceeds certain size of diameter. This approach is acceptable for a standard apparatus for determining flammability limits by Coward and Jones. It is a vertical tube 51 mm in diameter and 1.8 m long, closed at the upper end and open to the atmosphere at the bottom [11]. The flame propagation in the corresponding geometry was analyzed in this study. Therefore the radiative heat loss was considered dominant for energy balance in the flame front. The effects of reduced radiative heat loss from the flame can be classified as conduction of heat into the post-reaction zone, which is cooled by radiative heat loss $q_{rad,1}$ and radiative heat loss from reaction zone itself $q_{rad,2}$ as shown **Figure 1** [10].

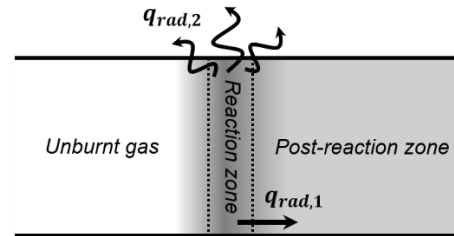


Figure 1. Radiative heat loss mechanisms

First, the conduction of heat into the post-reaction zone is caused by the temperature gradient near the end of the reaction zone. In steady-state one-dimensional flame propagation, the temperature distribution in the post-reaction zone can be defined as **Equation (5)**. Where R is the volumetric heat loss dependent of the mixture properties and the local temperature distribution in the post-reaction zone. k is the thermal conductivity. Usually, the conduction heat transfer is much smaller compared to the heat transfer by transport term near the peak temperature [10]. Therefore, a peak temperature gradient under the influences of heat losses can be solved by **Equation (6)**.

$$\rho_u c_p S_u \frac{dT}{dx} - \frac{d}{dx} k \frac{dT}{dx} = -R(T) \quad (5)$$

$$\left(\frac{dT}{dx} \right)_{x=x_f} = - \frac{R(T_f)}{\rho_u c_p S_u(T_f)} \quad (6)$$

From **Equation (8)**, the heat loss rate from the reaction zone per unit area of the flame front due to conduction in to the cooling post-reaction zone can be calculated by **Equation (7)**. It consists of several variables determined by the mixture properties.

$$q_{rad,1} = k_f \frac{R(T_f)}{\rho_u c_p S_u(T_f)} \quad (7)$$

Second, the radiative heat loss from reaction zone itself is integrated by **Equation (8)**.

$$q_{rad,2} = \int_0^{\delta} R_r dx \quad (8)$$

However, Mayer insisted that the radiative heat loss from reaction zone itself can be ignored inasmuch because the space-average value of R_r in the reaction zone is much lower than $R(T)$ [10]. As a result, the amount of heat loss for calculating the peak temperature can be estimated only by considering the conduction of heat related to the radiative heat loss.

2.3 Development of calculated non-adiabatic flame temperature (CNFT) model

In this study, **Equation (1)** was modified to **Equation (9)** to develop a LFL prediction model by considering the radiative heat transfer. This equation assumed combustion in a non-adiabatic condition by considering heat loss due to the radiative heat transfer. Because the radiative heat transfer dominantly determines total amount of heat loss from reaction zone to environment. It took into account the radiative heat loss mechanism based on the energy equation for flame front. As shown in **Equation (10)**, the magnitude of radiative heat loss can be determined by the thermal diffusivity, flame speed, and volumetric heat loss. The flame speed, as mentioned above, is independent of the mixture properties for the limiting mixtures. The volumetric heat loss determined by the flame temperature also becomes constant because the flame temperature is independent of the mixture properties according to previous studies [3-5]. So the ratio of these two variables was expected as constant if fuel type is the same. It was concluded that the magnitude of the radiative heat loss can be estimated by the function of thermal diffusivity. Also, it was expected that the temperature would remain constant as the initial temperature of the mixtures changes or the diluent is added for limiting mixture as similar as CAFT model. This is the characteristic concept of the calculated non-adiabatic flame temperature (CNFT) model developed in this study. This approach can be used to predict LFL by accounting for the change in heat capacity of mixture.

$$\sum n_i [\Delta H_{f,i}^0]_{reactants} - \sum n_i [\Delta H_{f,i}^0 + \bar{c}_{p,i} (T_{CNFT} - T_{initial})]_{products} = Q_{rad,1} \quad (9)$$

$$q_{rad,1} = k_f \frac{R(T_f)}{\rho_u c_p S_u(T_f)} = \frac{\alpha R(T)}{S_u(T_f)} \quad (10)$$

3. Results and discussion

3.1 Validation of CNFT model

Mathematically derived **Equation (10)**, which predicts that the radiative heat loss and thermal diffusivity would be linearly proportional, was validated with experimental results in **Table 1**. The heat loss magnitude of each mixture was inversely estimated using the experimental results according to **Equation (9)**. The thermal diffusivity was divided by the molar concentration C to predict the heat loss in mole units. This parameter was defined as a CNFT coefficient π . The CNFT of the H_2 -air mixture at 20 °C was assumed equal to the CAFT, 579 K because a lower CNFT coefficient indicates a lower amount of radiative heat loss. The coefficient was calculated as the mean value of each gas component of the mixture with physical properties.

Figure 2 shows calculated radiative heat loss for each mixture with the coefficient. A proportional relationship between the two variables was confirmed and **Equation (11)** was determined by a least-squares analysis with R^2 of 0.98. It implies that they show a strong linear relationship except for the mixture including steam. Also, it should be noted that the elevated initial temperature mixtures or helium mixtures have higher coefficient than other mixtures. Because they are representative mixtures, prediction of the LFL is difficult with the CAFT model. In other words, these mixtures with high CNFT coefficient require more combustion heat to compensate for higher radiative heat loss than other mixtures.

$$Q_{rad,1}(\pi) = 0.246(\pi - \pi_{Air,20^\circ C}), \quad \pi = \alpha / C [10^3 \cdot \frac{cm^5}{mol \cdot sec}] \quad (11)$$

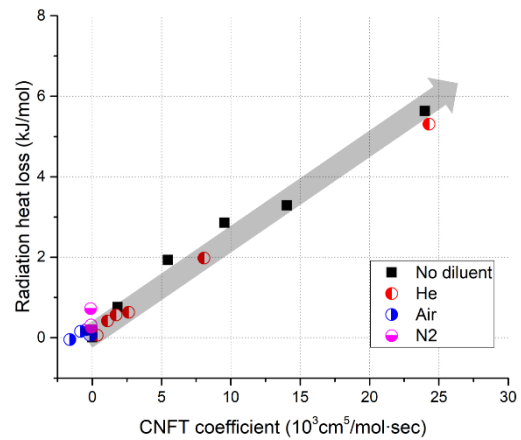


Figure 2. Linear relationship between CNFT coefficient and radiative heat loss

3.2 Prediction of the LFL by CNFT model

The accuracy of the CNFT and CAFT models was compared. First, **Figure 3** shows the prediction of the LFL at H₂-air mixtures with elevated initial temperature by the both models. As the initial temperature increases, the CNFT coefficient of the mixtures become larger. So the reliability of the CAFT model without considering the radiative heat loss is decreased. The difference between the predicted value and the experimental results increases up to 2%. In contrast, the CNFT model reflects the magnitude of the radiative heat loss determined by CNFT coefficient. A maximum difference from the experimental results does not exceed 0.4%.

Figure 4 shows the prediction of the LFL at H₂-air-helium mixtures by the both models. As the composition of helium increases, the LFL in the experiment increases. Only the CNFT model reflecting increase of radiative heat loss according to the helium concentration exhibits reliable accuracy in comparison with experimental results. The CNFT model shows a maximum difference of 0.3%, while the CAFT model shows 2.0%. As a result, it was confirmed that consideration of radiative heat loss is essential for LFL prediction in that the error for mixtures with relatively high thermal is greatly reduced.

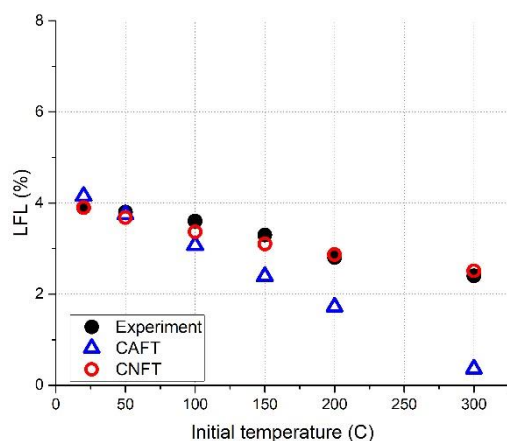


Figure 3. Prediction of LFL by the CNFT and CAFT models in H₂-air mixtures

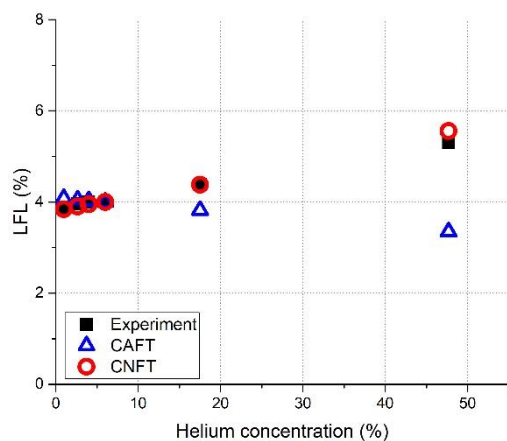


Figure 4. Prediction of LFL by the CNFT and CAFT models in H₂-air-He mixtures

4. Conclusions

In this paper, the CNFT model was proposed for prediction of LFL following concept of CAFT model. Agreement with experiment on H₂-air-diluent mixtures was significantly improved especially for the elevated initial temperature and steam mixtures, where the previous models showed inherent limitation. It was confirmed that consideration of radiative heat loss is essential to predict intrinsic LFL of each mixture. Developed CNFT model is expected to contribute on hydrogen safety analysis of nuclear power plants.

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