

A Theoretical Study to Compare Flame Characteristics of H₂ and CO Flames for Containment Safety Analysis

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1. Introduction

The hydrogen combustion in a containment is one of the vulnerable phenomena threatening the safety of a nuclear power plant (NPP). During a postulated severe accident, a large amount of hydrogen can be generated dominantly through an exothermic reaction of zircaloy and steam. As hydrogen promptly diffuses throughout the containment atmosphere, highly combustible hydrogen mixtures can be formed. The combustion of these mixtures can impose high pressure and temperature loads to the containment structure depending on the thermodynamic and chemical properties. Owing to this potential risk of hydrogen combustion, most countries operating and constructing NPPs is devoting significant attention to prevent such hydrogen risk [1].

On the other hand, if the accident progresses to the ex-vessel phase, carbon monoxide and hydrogen are generated in the containment cavity through molten corium-concrete interaction (MCCI). Carbon monoxide is also a flammable gas, which raises the combustion risk when coexisting with hydrogen. However, most of the previous studies and regulations have mainly focused on the hydrogen risk and paid relatively less attention to the risk posed by carbon monoxide [2]. For this reason, the IRSN in France announced a plan to develop a methodology for combustion risk assessment of the H₂/CO mixture in lately [3].

Recently, the calculated non-adiabatic flame temperature (CNAFT) model was developed to predict the LFL of hydrogen mixtures [4]. The CNAFT model, which shows a reasonable accuracy for various hydrogen mixtures, considers the heat loss mechanisms during upward propagation to overcome the previous adiabatic models. On the contrary, the LFL of carbon monoxide has been properly predicted by the calculated adiabatic flame temperature (CAFT) model [5]. For this reason, Kim et al. suggested a combined methodology for H₂/CO mixture including the CNAFT and CAFT model separately based on Le Chatelier's law [2]. However, it is not yet clear why adiabatic assumption for predicting the LFL is selectively available for fuel types. In this study, we theoretically compared the characteristic of hydrogen and carbon monoxide flame to explain the need for different models. The flame analysis focused on the preferential diffusion effects dependent on the Lewis number.

2. Flammability limit of H₂ and CO mixtures

The LFL is the minimum fuel concentration where the flame can continuously propagate. If a fuel concentration of local gas mixture in containment exceeds the LFL, the mixture can combust by ignition. These kinds of combustion are a major threat for containment integrity due to possibility of overpressure. Furthermore, a more detailed risk analysis of the NPP should be performed to verify the potential risk of flame acceleration (FA) and detonation if the concentration exceeds the threshold. The importance of prediction of fuel lower flammability limit (LFL) in characterizing fire and explosion hazards has long been recognized in nuclear industry [4].

Table 1 shows that the LFL concentration varies widely depending on the mixture condition for both hydrogen and carbon monoxide. In FITS experiments, LFL of hydrogen mixtures was measured according to the steam concentration for nuclear reactor safety analysis. The LFL of H₂-air mixture at room temperature is known to be 4.1 vol%. Due to the diluent effect of steam, the LFL increase up to 8% depending on the steam concentration [6]. Although not as vigorously studied as hydrogen, the LFL of carbon monoxide has been measured by several experiments. Among the experiments, White observed the variation of the LFL according to the initial temperature. These results show a wide range of 11 to 16 vol% depending on the mixture conditions [7]. The MELCOR code, a representative severe accident analysis code, still uses conservative constant values for LFL prediction. Therefore, the need for a reasonable model for predicting the LFL of combustible gas has been continuously required.

Table 1. LFL concentration by fuel type

Reference	LFL (vol%)	
	Hydrogen	Carbon monoxide
FITS experiments [6]	4-8	-
White' experiments [7]	-	11-16*
MELCOR	4.1	12.5

*12.4 vol% at 300°C

2.1 Hydrogen mixtures

The CAFT model was developed by assuming that the flame peak temperature can be estimated by the

calculated flame temperature with adiabatic condition. It has been remarked that the peak temperature was proportional to CAFT, and therefore the LFL concentration can be predicted as a concentration reaching the threshold CAFT [8, 9]. The CAFT value can be calculated as shown Eq. (1) [10, 11]. In adiabatic processes, the heat produced during the exothermic chemical reaction is transferred only to the products, and the temperature of the products increases. Vidal et al. insisted that the CAFT is a powerful tool for estimating the LFL of gaseous mixtures [9]. In the case of hydrogen mixtures, the threshold CAFT for the LFL is known to be approximately 600 K [12].

Figure 1 shows the accuracy of the CAFT model for hydrogen mixtures. Initial conditions for the hydrogen mixtures can be referred in Ref. [4]. Although the model shows the reasonable accuracy for various mixtures, other mixtures show significant differences as shown. This discrepancy is the most pronounced when the initial temperature is high or the steam is included. This limitation was caused by the simplified assumption of adiabatic flame propagation.

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

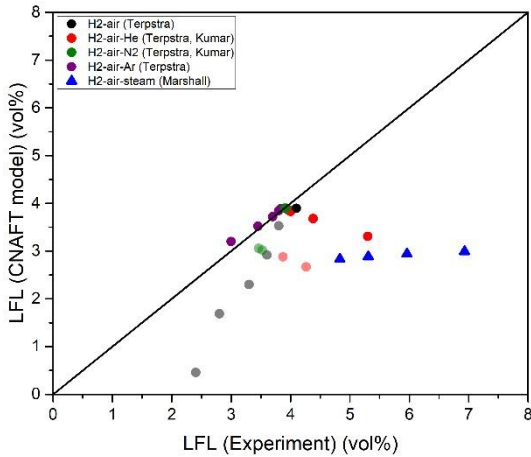


Figure 1. Validation of CAFT model for hydrogen mixtures. The reduced accuracy was identified for the mixtures with dilution of steam.

Figure 2 shows the improved accuracy of the CNAFT model for same hydrogen mixtures. The CNAFT can be calculated by the energy equation considering heat loss mechanism at the flame skirt as shown Eq. (2). The model can estimate the amount of heat loss according mixture properties based on the heat loss rate from the reaction zone due to conduction into the cooling post-reaction zone. The model reliability was confirmed for H₂-air mixtures up to 300 °C and H₂-air-Steam mixtures up to 40 vol. % steam concentration as shown Figure 2 [3]. The detailed derivation process of the heat loss amount can be identified in Refs. [4, 13]. In conclusion, it was noted that the heat loss mechanism, which is

neglected in adiabatic assumption, should be considered to predict the LFL of hydrogen mixtures.

$$\sum_{\text{reactants}} n_i [\Delta H_{f,i}^0 + \bar{c}_{p,i}(T_i - T_{ref})] - \sum_{\text{products}} n_i [\Delta H_{f,i}^0 + \bar{c}_{p,i}(T_{CNAFT} - T_{ref})] = Q_{rad,1} \quad (2)$$

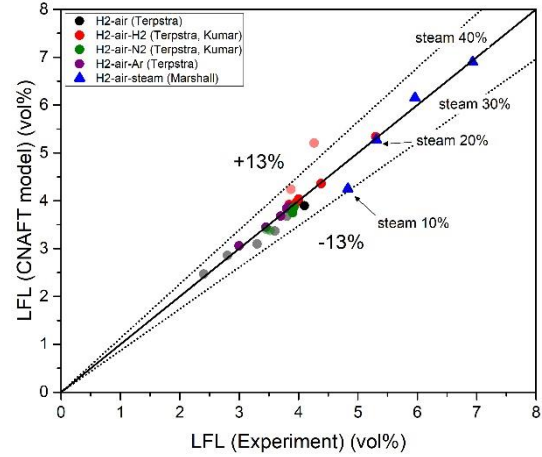


Figure 2. Validation of CNAFT model for hydrogen mixtures

2.2 Carbon monoxide mixtures

Table 2 shows the measured LFL and the CAFT values from the limiting mixtures by White [7]. The initial temperature of the tested mixture increases up to 673 K at room temperature. It was noted that, although the measured LFL shows significant difference depending on the mixture condition, the CAFT for limiting mixtures were almost constant. The maximum relative difference is only 6% for CAFT (30% for LFL) based on the mixture at room temperature. It means that the LFL of carbon monoxide can be well predicted by the CAFT model. In next section, we theoretically compared the hydrogen and carbon monoxide flame to understand the distinction of adiabatic model accuracy.

Table 2. Influence of temperature on CAFT at limiting mixtures of CO-air mixtures [7]

Temperature (K)	LFL (vol%)	CAFT (K)
290	16.3	1490
323	15.7	1465
373	14.8	1435
423	14.2	1425
473	13.5	1410
523	12.9	1405
573	12.4	1405
623	12.0	1410
673	11.4	1410

3. Comparison of H₂ and CO mixtures based on the Lewis number

The Lewis number is defined as the ratio of the thermal diffusivity to the mass diffusivity. Previous

studies insisted that the number has significant importance for understanding the transport process associated in gaseous mixtures, especially in the field of premixed combustion [14]. Unlike theoretical one dimensional flame, the coupled effect of preferential diffusion and flame stretch affects the flame temperature depending on the Lewis number in real propagation.

For $Le < 1$ flames (H_2 -air), the local burning intensity increases at the flame tip due to strong preferential mass diffusion as shown **Figure 3**. Burning intensity is a scale of the amount of heat produced by combustion reaction. In other words, the diffusive mass transport from the unburned gas to the stretched flame is concentrated at the flame edge [15]. On the contrary, for $Le > 1$ flames (CO-air), the strong preferential heat diffusion spreads the high temperature generated by combustion at the flame edge over a wide area. It means that the local flame temperature can be reduced than planar flame. This trend is reversed when the fuel direction for curvature changes.

The coupled effect to flame structure also affects the extinction process of each flame. First, the extinction of hydrogen flame ($Le < 1$) proceeds as the flame skirt on both sides become shorter. Since the local burning intensity is focused on the flame tip, the intensity at the both sides is relatively weak (critical point) as shown **Figure 4**. In the same vein, for $Le > 1$ flames, the local intensity at the edge is relatively weak due to the diffusive heat transport. Jarosinski et al. experimentally observed that the extinction of methane flames occurred from the flame tip [16]. The Lewis number of methane flames has a value close to 1, which is much larger than hydrogen flames. We can deduce that the extinction mechanism at flame tips will be more pronounced for CO-air flames with a higher Lewis number.

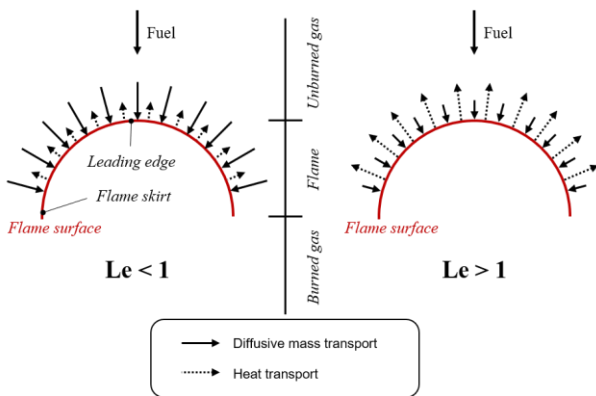


Figure 3. Diffusive heat and mass transport near the flame surface dependent on Lewis number

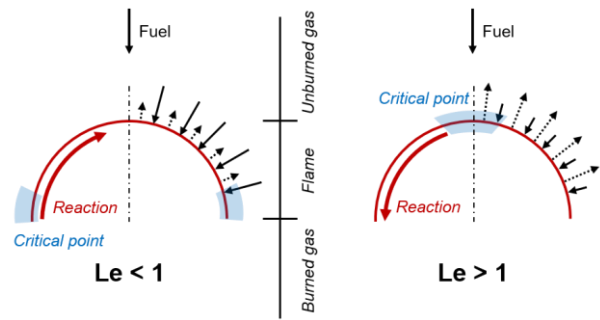


Figure 4. Critical point for flame extinction dependent on Lewis number

We can speculate why adiabatic assumption for predicting the LFL is selectively available, through these flame structure analyzes based on the Lewis number. For $Le < 1$ flames, the heat loss mechanism at the flame skirt on the both sides is important for LFL prediction. The balance of generated combustion heat and heat loss on the sides determines whether a flame continuously propagates. On the other hand, for $Le > 1$ flames, balance of combustion heat and heat loss on the flame tip is important. Turns noted that the indirect radiation (conduction) mechanism can account for the LFL of gas mixtures [17]. The relative amount of the indirect radiation from the flame to the burned gas is significantly greater on the bottom (flame skirt) than the flame tip due to the concave flame structure. In conclusion, it seems that the characteristics of local burning intensity with Lewis number determine the availability of the adiabatic model. Our speculation was based on phenomenological analysis of flame structure based on the Lewis number. Although it has been experimentally identified that the flame extinction occurs at each critical point predicted by the Lewis number, the effect of heat loss needs to be evaluated quantitatively. This is our future study.

Figure 5 shows the distribution of burning intensity for the binary fuel flames simulated by Zhou et al [15]. The left contour shows the lean limit flames for a fuel consisting of 40% hydrogen and 60% methane. The equivalence ratio of the fuel is 0.320. The focused burning intensity at the flame tip was identified. The right contour shows the lean limit flames with a higher mole fraction of methane ($\phi = 0.323$). It is interesting that the maximum burning intensity shifted to the side from the edge as Lewis number increases. It means that the preferential diffusion effects with the Lewis number can valid for binary fuel flames. Due to the larger Lewis number of carbon monoxide than methane, this change in local intensity distribution will be more pronounced. We expected that the CAFT and CNAFT model can be selected based on the Lewis number for the H_2/CO mixture range except for the transition range. For transition range where the adiabatic assumption is partially effective, the Kim's combined methodology can be used for predicting LFL as mentioned [2].

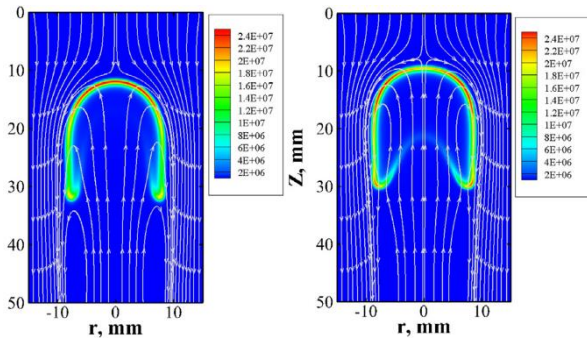


Figure 5. Distribution of chemical heat release with different H₂ contents in ultralean fuel gas (left: 40% H₂+60% CH₄ fuel, right: 20% H₂+80% CH₄ fuel) [15]

4. Conclusions

The combustion risk assessment of the H₂/CO mixture is essential for investigating containment integrity since the severe accident can progress to the ex-vessel phase. In this study, we theoretically compared the flame characteristic of hydrogen and carbon monoxide flame to clarify the reason why the heat loss effects on the flammability differs. For $Le < 1$ flames (H₂-air), the local burning intensity increases at the flame tip due to strong preferential mass diffusion. It means that the heat loss mechanism at the flame skirt on the both sides is important for LFL prediction. The balance of generated combustion heat and heat loss on the sides determines whether a flame continuously propagates. On the contrary, for $Le > 1$ flames (CO-air), the strong preferential heat diffusion spreads the high temperature generated by combustion at the flame edge over a wide area. Balance of combustion heat and heat loss on the flame tip is important. Turns noted that the indirect radiation (conduction) mechanism can account for the LFL of gas mixtures [17]. The relative amount of the indirect radiation from the flame to the burned gas is significantly greater on the bottom (flame skirt) than the flame tip due to the concave flame structure. In conclusion, it seems that the characteristics of local burning intensity with Lewis number determine the availability of the adiabatic model. We expect that this analysis with Lewis number can be valid for combustion risk assessment of the H₂/CO. This is our future study.

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