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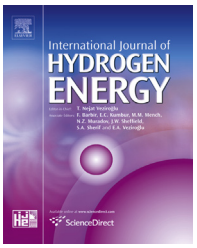

Assessment of the method for calculating the Lewis number of $H_2/CO/CH_4$ mixtures and comparison with experimental results

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ABSTRACT

This paper investigates the various techniques used in the literature to calculate the effective Lewis number of two-component (H_2/CO and H_2/CH_4) and three-component fuels ($H_2/CO/CH_4$ and $H_2/CO/CO_2$) over a wide range of equivalence ratios ($0.6 \leq \varphi \leq 1.4$) under laminar flame conditions. The most appropriate effective Lewis number formulation is identified through comparison with experimentally extracted Lewis numbers (*Le*). The paper first identifies the proper methodology to extract the experimental *Le* from the burned Markstein length of an outwardly propagating flame. Second, the different methodologies for the calculation of the effective *Le* are presented and compared to experimental results for H_2/CH_4 and H_2/CO mixtures. Based on the experimental results, it is shown that the calculation of the effective *Le* of mixtures can be divided into a three-step procedure depending on the equivalence ratio: (1) calculation of the *Le* for each fuel and the oxidizer; (2) use of the *Le* mixing rule; and (3) assessment of the necessity or not of combining the fuel's and oxidizer's Lewis numbers. The paper shows that, in rich mixtures, the oxidizer *Le* needs to be taken into account. Lastly, the methodology is validated for $H_2/CO/CH_4$ and $H_2/CO/CO_2$ fuels.

Introduction

Combustion is a process that relies profoundly on transport phenomena. Through heat transfer, the heat released by the flame is used to preheat the reactants to the activation temperature. Meanwhile, mass transfer ensures that the flame is continuously supplied with reactants. The imbalance of thermal diffusivity to mass diffusivity is known as nonequidiffusion [1]. It is represented by the Lewis number (*Le*), which is defined as the ratio of the thermal diffusivity to the mass diffusivity of the deficient reactant. Nonequidiffusion impacts the behavior of the stretched flames in many ways, such as on the minimum ignition energy and flame kernel [2–4] or on the determination of flame speed [5,6]. In particular, flame's stability characteristics depend on nonequidiffusion through a mechanism referred to as diffusional–thermal instability. While the flame is unconditionally unstable when *Le* is below the critical value Le^* (slightly smaller than 1 and thus often approximated as $Le^* = 1$) [7], diffusion helps to stabilize the flame when $Le > Le^*$. This instability mechanism is the cause of the apparition of

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Nomenclature

A	Flame area, m^2
A_1	Factor of mixture strength, –
C_p	Specific heat, J kg^{-1} K^{-1}
D_{ij}	Binary mass diffusivity, m^2 s^{-1}
$D_{i,mix}$	Mixture-averaged mass diffusivity, m^2 s^{-1}
$(dT/dx)_{max}$	Maximum temperature gradient, K m^{-1}
E_a	Activation energy, J mol^{-1}
k	Thermal conductivity, W m^{-1} K^{-1}
L_b	Burned Markstein length, m
L_u	Unburned Markstein length, m
l_{f_G}	Flame thickness calculated based on the temperature gradient, m
l_{f_K}	Flame thickness calculated based on the kinetic definition, m
Le	Lewis number, –
Le_{eff}	Effective Lewis number, –
Le_{f/O_2}	Combination of Le associated to the fuel and oxidizer, –
Le_{fuel}	Fuel Lewis number, –
Le_i	Individual Lewis number of each component i (fuel i or O_2 as the limiting reactant), –
$Le_{i,\alpha}$	Le_i with diffusion towards the whole air/fuel mixture, –
$Le_{i,\varphi}$	Le_i with diffusion towards air, using the quantity of air required to maintain φ of the overall mixture, –
$Le_{i,\varphi i}$	Le_i with diffusion towards air, using all the air available in the overall mixture, –
Le_D	Diffusion-based mixing formulation of Le_i, –
Le_H	Heat-based mixing formulation of Le_i, –
Le_V	Volume-based mixing formulation of Le_i, –
Le_{O_2}	Oxidizer Lewis number, –
Le_{BM}	Experimental Le extracted based on the work from Bechtold and Matalon [32], –
Le_{Chen}	Experimental Le extracted based on the work from Chen et al. [5,44], –
Le_{Law}	Experimental Le extracted based on the work from Law and Sung [51], –
q_i	Nondimensional heat release, –
Q_i	Heat of reaction, J kg^{-1}
R	Flame radius, m
R^0	Universal gas constant, J mol^{-1} K^{-1}
S_b	Laminar flame speed of the burned gas, m s^{-1}
S_b^0	Unstretched laminar flame speed of the burned gas, m s^{-1}
S_u	Laminar flame speed of the unburned gas, m s^{-1}
S_u^0	Unstretched laminar flame speed of the unburned gas, m s^{-1}
t	Time, s
T_{ad}	Adiabatic temperature, K
T_u	Unburned mixture temperature, K
Y_i	Mass fraction of species i in the fuel blend, –
$Y_{i,mix}$	Mass fraction of species i in the air/fuel mixture, –
Ze	Zel'dovich number, –

Greek symbols

α	Thermal diffusivity, m^2 s^{-1}
$\overline{\alpha}$	Factor accounting for the expansion ratio, –
κ	Stretch rate, s^{-1}
ρ_b	Density of the burned mixture, kg m^{-3}
ρ_u	Density of the unburned mixture, kg m^{-3}
σ	Flame expansion ratio, –
φ	Equivalence ratio, –
Φ	Ratio of excess-to-deficient reactants, –
χ_i	Mole fraction of species i in the fuel blend, –

cells on lean hydrogen and rich propane flames [7–10]. These cells wrinkle the propagating flame front, which creates a self-accelerating flame [7], induces turbulence in the unburned mixtures, and leads to engine knock [11]. The correct interpretation of many combustion phenomena therefore relies on the precise calculation of the Le which is a key parameter to discriminate between stable and unstable flame fronts.

While the definition of Le in a single-fuel mixture is quite straightforward, defining Le in a multifuel mixture is not trivial since the diffusivity characteristics of each fuel have to be considered. Because there is no consensus on the matter, three formulations of an "effective" Lewis number (Le_{eff}) have been reported in the literature. Bouvet et al. [12] performed an experimental investigation examining the validity of the three Le_{eff} formulations in lean mixtures (equivalence ratios of 0.6 and 0.8) of H_2/CO, H_2/CH_4, H_2/C_3H_8, and H_2/C_8H_{18}. Using the theoretical link proposed by Chen and Ju [13] between the burned Markstein length (L_b) and Le, Bouvet et al. [12] compared the experimentally determined Markstein lengths to the Markstein lengths calculated using each formulation of Le_{eff}. They identified a qualitatively accurate formulation for the prediction of Le in H_2/alkane mixtures, while no Le_{eff} formulation has been found to adequately fit the results obtained with H_2/CO mixtures.

Over the last few years, there has been a marked interest for hydrogen-based fuels, since hydrogen can reduce the emission of pollutants from combustion by being a non-carbon-based source of energy and by extending the lean operating limit [14–16]. Many studies have been devoted to the evaluation of the combustion characteristics and performance of such fuels in gas turbines [17,18] and spark ignition engines [19,20]. A precise characterization of combustion properties of hydrogen-based fuels, including Le, is of importance in theoretical and numerical analyses [21–24]. The goal of this paper is thus to focus on investigating syngas Le while broadening the investigation of Bouvet et al. [12] by (1) assessing the validity of the three Le_{eff} formulations using two additional theoretical equations linking Le to the Markstein length; (2) expanding the investigation from lean to stoichiometric and slightly rich conditions; (3) evaluating the impact of CH_4 and CO_2 addition to H_2/CO. This broad investigation is performed at equivalence ratios (φ) ranging from 0.6 to 1.4, with respect to the rich flammability limit of CH_4. The volumetric content of H_2, CO and CH_4 was varied from 0 to 100%,[1] while the CO_2 content was

[1] 2% of H_2 (5% at $\varphi = 0.6$) was required to ensure the combustion of pure CO.

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