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Assessment of the method for calculating the Lewis number of H₂/CO/CH₄ mixtures and comparison with experimental results

Denis Lapalme^{*}, Romain Lemaire, Patrice Seers

Department of Mechanical Engineering, École de technologie supérieure, 1100 Notre-Dame Ouest, Montréal, QC, H3C 1K3, Canada

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ABSTRACT

This paper investigates the various techniques used in the literature to calculate the effective Lewis number of two-component (H₂/CO and H₂/CH₄) and three-component fuels (H₂/CO/CH₄ and H₂/CO/CO₂) over a wide range of equivalence ratios ($0.6 \le \varphi \le 1.4$) 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 H₂/CH₄ and H₂/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 H₂/CO/CH₄ and H₂/CO/CO₂ fuels.

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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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^{*} Corresponding author.

E-mail address: denis.lapalme.1@ens.etsmtl.ca (D. Lapalme).

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AFlame area, m^2 Bechtol and Matalon [32], $-$ A1Factor of mixture strength, $ Le_{Chen}$ Experimental Le extracted based on the work from Chen et al. [5,44], $ D_{ij}$ Binary mass diffusivity, $m^2 s^{-1}$ Le_{Law} Experimental Le extracted based on the work from Law and Sung [51], $ (dT/dx)_{max}$ Maximum temperature gradient, K m^{-1} Q_i Heat of reaction, $J \ kg^{-1}$ E_a Activation energy, J mol ⁻¹ RFlame tarelease, $ (dT/dx)_{max}$ Maximum temperature gradient, K m^{-1} Q_i Heat of reaction, $J \ kg^{-1}$ L_b Burned Markstein length, mSbLaminar flame speed of the burned gas, m s ⁻¹ L_b Burned Markstein length, mSbLaminar flame speed of the burned gas, m s ⁻¹ L_i Unburned Markstein length, mSuLaminar flame speed of the burned gas, m s ⁻¹ L_e Flame thickness calculated based on the temperature gradient, mSuLaminar flame speed of the unburned gas, m s ⁻¹ L_e Effective Lewis number, $-$ tTime, s L_{effr} Effective Lewis number, $-$ TTad Adiabatic temperature, K Le_{int} full full full full full full full full	Nomenclature	Le _{BM}	Experimental Le extracted based on the work from
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$ \begin{array}{llllllllllllllllllllllllllllllllllll$	A Flame area, m	Le _{Chen}	Experimental Le extracted based on the work from
	A_1 Factor of mixture strength, –		Chen et al. [5,44], –
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	C_p Specific near, j kg K	Le _{Law}	Experimental Le extracted based on the work from
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	D_{ij} Binary mass diffusivity, m ² s ⁻¹		Law and Sung [51], –
	$D_{i,mix}$ Mixture-averaged mass diffusivity, m ⁻ s ⁻¹	qi	Nondimensional heat release, –
L_a Activation energy, J mol $^{-1}$ K $^{-1}$ RFlame radius, mkThermal conductivity, W m $^{-1}$ K $^{-1}$ RUniversal gas constant, J mol $^{-1}$ K $^{-1}$ L_b Burned Markstein length, mSbLaminar flame speed of the burned gas, m s $^{-1}$ L_u Unburned Markstein length, mSbUnstretched laminar flame speed of the burned gas, m s $^{-1}$ L_u Unburned Markstein length, mSbUnstretched laminar flame speed of the burned gas, m s $^{-1}$ L_u Flame thickness calculated based on the kinetic definition, mSuLaminar flame speed of the unburned gas, m s $^{-1}$ Le Lewis number, -Time, sTime, s Le_{eff} Effective Lewis number, -TadAdiabatic temperature, K Le_{fuel} Fuel Lewis number, -TadMass fraction of species i in the fuel blend, - Le_{fuel} Fuel Lewis number of each component i (fuel i or O_2 as the limiting reactant), - $Zel dovich number, -Le_{i,a}Le_i with diffusion towards air, using the quantity of air required to maintain \varphi of the overall mixture, -\alphaThermal diffusivity, m^2 s^{-1}Le_{i,qi}Le_i with diffusion towards air, using all the air available in the overall mixture, -\sigmaFlame expansion ratio, -Le_{i,qi}Le_i with diffusion of Le_i, -\varphiDensity of the unburned mixture, kg m^{-3}Le_{i,qi}Le_i with diffusion of Le_i, -\varphiRatio of excess-to-deficient reactants, -Le_{i,qi}Le_i with diffusion of Le_i, -\varphiRatio of excess-to-deficient r$	(a1/ax) _{max} Maximum temperature gradient, K m ⁻²	Qi	Heat of reaction, J kg ⁻¹
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Le _{O2} Oxidizer Lewis number, –	Le_V Volume-based mixing formulation of Le_i , –	χ_i	Mole fraction of species i in the fuel blend, –
	Le _{O2} Oxidizer Lewis number, –		

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 (Leeff) have been reported in the literature. Bouvet et al. [12] performed an experimental investigation examining the validity of the three Leeff 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 Leeff. They identified a qualitatively accurate formulation for the prediction of Le in H2/alkane mixtures, while no Leeff formulation has been found to adequately fit the results obtained with H₂/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 noncarbon-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 Leeff 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 CH4. The volumetric content of H2, CO and CH4 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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