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A Fuel-Independent Measure of Mixture Stoichiometry**

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The Oxygen Ratio:

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Abstract

The pollutant-formation characteristics and other properties of a combustion reaction typically depend strongly on the proximity of the mixture to its stoichiometric condition, *i.e.*, the “mixture stoichiometry.” A quantitative, widely applicable measure of this mixture property is therefore a critical independent variable in the study of combustion systems. Such a parameter enables the clear separation of mixture stoichiometry effects from other effects (*e.g.*, fuel molecular structure, product temperature, diluent concentration, pressure). The parameter most often used to quantify mixture stoichiometry is the equivalence ratio. Unfortunately, the equivalence ratio fails to properly account for oxygen in oxygenates, *i.e.*, compounds that have oxygen chemically bound within the fuel molecule. This manuscript introduces the oxygen ratio, a parameter that properly characterizes mixture stoichiometry for a broader class of reactants than does the equivalence ratio, including oxygenates. A detailed definition of the oxygen ratio is provided and used to show its relationship to the equivalence ratio. The definition is also used to quantify errors involved when the equivalence ratio is used as a measure of mixture stoichiometry with oxygenates. Proper usage of the oxygen ratio is discussed and the oxygen ratio is used to interpret results in a practical example.

Keywords: oxygenate; equivalence ratio; oxygen ratio

1. Introduction

The properties of a combustion reaction (*e.g.*, adiabatic flame temperature, flame speed, product concentrations) typically depend strongly on the proximity of the mixture to its stoichiometric condition. A mixture is said to be at its stoichiometric condition when the ratio of fuel to oxidizer is such that all fuel and oxidizer could be completely converted to stable product species. A quantitative, widely applicable measure of the proximity of a mixture to its stoichiometric condition is therefore a critical independent variable in the study of combustion systems.

Many parameters are used, either implicitly or explicitly, to quantify “mixture stoichiometry” (*i.e.*, the proximity of a mixture to its stoichiometric condition). Although mass and mole fractions of certain species may correlate with mixture stoichiometry for certain mixtures, it is easy to show that they are not general measures of mixture stoichiometry. Similarly, the carbon-to-oxygen atomic ratio is not a general measure of mixture stoichiometry because it neglects the oxidation of hydrogen. On the other hand, the equivalence ratio is widely used to quantify mixture stoichiometry. The equivalence ratio is denoted ϕ and defined as

$$\phi \equiv \frac{(m_f/m_o)}{(m_f/m_o)_{st}} \quad (1)$$

where m_f and m_o are the masses of fuel and oxidizer in the mixture, respectively, and the subscript *st* indicates evaluation at the stoichiometric condition [1,2]. It will be shown below that ϕ is an accurate measure of mixture stoichiometry under some conditions, for instance when the fuel is a hydrocarbon (*i.e.*, composed of only hydrogen and carbon atoms). However, ϕ is not necessarily an accurate measure of mixture stoichiometry when oxygenated fuels are used. An

oxygenated fuel is defined as one that has oxygen chemically bound into the fuel molecule (*e.g.*, alcohols, ethers, esters). Investigations into the potential of non-traditional fuels to reduce soot emissions from diesel engines have highlighted the need to define a parameter that is more general than the equivalence ratio to accurately measure mixture stoichiometry when oxygenated fuels are used [3-5].

The following two mixtures illustrate the problem. The first is a fuel-rich mixture of methane (CH_4) and oxygen (O_2):



The second is a fuel-rich mixture of carbon monoxide (CO) and oxygen:



The first and second mixtures have equivalence ratios of 1.67 and 5.00, respectively. Such very different equivalence ratios would suggest that the mixture stoichiometries are very different, but they are not. This can be shown by considering a parameter, denoted Ω , that is defined as the ratio of the number of oxygen (O) atoms in a mixture divided by the number of O -atoms required for stoichiometric combustion of the mixture. By definition, Ω is a direct measure of mixture stoichiometry.

The CH_4/O_2 mixture contains 4 moles of O -atom and it would require $20/3$ moles of O -atom for stoichiometric combustion, yielding

$$\Omega_1 = \frac{4}{20/3} = \frac{3}{5} = 60\%. \quad (4)$$

The CO/O_2 mixture contains 6 moles of O -atom and would require 10 moles of O -atom for stoichiometric combustion, yielding

$$\Omega_2 = \frac{6}{10} = 60\%. \quad (5)$$

Each of the mixtures has 60% of the oxygen required for stoichiometric combustion. Thus, *the two mixtures are equally close to their stoichiometric conditions, but they have different equivalence ratios*. The equivalence ratios are different because the definition of ϕ does not properly account for oxygen that is chemically bound in the fuel. Thus, if ϕ were used to characterize the CO/O_2 mixture stoichiometry, the mixture would appear significantly more fuel-rich than it actually is.

Because ϕ is such a fundamental parameter, its misrepresentation of mixture stoichiometry when oxygenated fuels are used can be harmfully misleading. For example, if the sooting characteristics of a hydrocarbon and an oxygenated fuel are compared at the same rich ϕ -value, the oxygenated-fuel mixture will likely produce less soot. As shown above, this is likely because the oxygenated-fuel mixture will actually be closer to its stoichiometric condition (*i.e.*, it will be leaner) than the hydrocarbon-fuel mixture. Without this knowledge, considerable effort might be spent searching for a mechanism to explain the differences in soot concentrations. This manuscript resolves such problems by introducing a parameter called the “oxygen ratio” that provides a quantitative, fuel-independent measure of mixture stoichiometry.

2. Definition of the Oxygen Ratio

The reactions of greatest practical importance in combustion are oxidation-reduction (or redox) reactions. In redox reactions, the oxidation number of at least one element must change as a result of the reaction. Oxidation numbers can be used to determine which elements in the reactant mixture act as oxidizers, which act as fuels (*i.e.*, reducing agents), and which are stable.

Once the elements that comprise the reactant mixture are classified, the proximity of the reactant mixture to its stoichiometric condition can be quantified.

The following classification procedure can be used to determine whether an element in the reactants acts as a fuel, an oxidizer, or a stable species. First, the equation for the stoichiometric reaction must be known. Each product of the stoichiometric reaction is called a saturated stoichiometric product, or SSP. A species is an SSP if and only if the valence orbitals of all its constituent atoms are filled (*i.e.*, saturated or closed-shell). Then the oxidation number of each element in the SSPs is determined using established guidelines that can be found in textbooks on general chemistry (see *e.g.*, [6,7]). Elements with positive, negative, and zero oxidation numbers in the SSPs act as fuels, oxidizers, and stable atoms, respectively, in the reactants.

Once each element in the reactants has been classified as a fuel, an oxidizer, or a stable atom, one could calculate a general “oxidizer ratio” by dividing the amount of oxidizer present in the given mixture by the amount required for stoichiometric reaction of the same quantity of fuel. This general procedure has been used before (*e.g.*, see definition in Eq. 204 of [8]), and would apply to systems that use metallic fuels, oxidizing elements other than oxygen, *etc.* While this is the most general measure of mixture stoichiometry, it can be challenging to implement in its full generality; for instance, certain elements can have many different oxidation numbers in the SSPs (depending on the reactants), and some reactions will have two or more elements acting as oxidizers of different strengths. For these reasons, it is desirable to trade off some of the generality of the “oxidizer ratio” for a parameter that is simpler to use. This parameter is called the oxygen ratio.

The oxygen ratio of a mixture is defined as the amount of oxygen in the mixture divided by the amount of oxygen required for stoichiometric combustion of the same quantity of fuel, where atoms bound in stable species are neglected. The oxygen ratio is a valid measure of mixture stoichiometry in any mixture for which the SSPs are CO_2 , H_2O , N_2 , and/or noble gases (NG). This definition of the oxygen ratio is applicable to a broad class of combustion problems related to transportation, power generation, and other industrial processes. The determination of mixture stoichiometry is easier within such a system because each element has a unique oxidation number in the SSPs. These oxidation numbers are presented in Table 1. Table 1 shows that oxygen is the only element acting as an oxidizer in this system.

The definition of the oxygen ratio requires the identification of stable reactant species. A stable reactant species is defined as one that is neither a fuel nor an oxidizer. In the discussion above, oxidation numbers were used to determine whether certain *elements* act as oxidizers, fuels, or stable atoms. A similar approach is used to determine whether certain *reactant species* act as oxidizers, fuels, or stable compounds [9]. This is accomplished by calculating the sum

$$S_i \equiv \sum_j n_{j,i} s_j \quad (6)$$

for each reactant species i . In Eq. 6, $n_{j,i}$ is the number of atoms of the j^{th} element in the i^{th} reactant species, and s_j is the oxidation number of the j^{th} element in the SSPs (taken from Table 1). If S_i is > 0 , then the i^{th} reactant species is a fuel. If S_i is < 0 , then the i^{th} reactant is an oxidizer. If $S_i = 0$ and the i^{th} reactant is an SSP, then the i^{th} reactant is a stable species. If $S_i = 0$ but the i^{th} reactant is not an SSP, then the i^{th} reactant is a fuel. An example of a fuel for which $S_i = 0$ is the monopropellant $C_2H_4(NO_3)_2$. A logic diagram for the classification of reactant species is shown in Fig. 1. Note from Fig. 1 that all stable species have $S_i = 0$, but not all species for which $S_i = 0$ are stable.

The definition of the oxygen ratio can be extended for classes of problems where it is valuable to distinguish between fuel-bound oxygen and oxygen originating from the ambient environment. For instance, when the combustion of an oxygenated fuel is mixing-controlled [4,5], a certain fraction of the oxygen required for stoichiometric combustion comes from the fuel, while a different fraction comes from entrained oxygen. This motivates splitting the total oxygen ratio of the mixture into an oxygen ratio of the fuel and an oxygen ratio of the oxidizer. Consider a general reactant mixture of the form:

$$\sum_i a_i C_{n_{C,i}} H_{n_{H,i}} O_{n_{O,i}} N_{n_{N,i}} NG_{n_{NG,i}} \quad (7)$$

where i is an index over all reactant species, a_i is the number of moles of the i^{th} species, and $n_{[element],i}$ is the number of atoms of $[element]$ in the i^{th} species. The oxygen ratio of this reactant mixture is defined as:

$$\Omega = \frac{\sum_k a_k n_{O,k} + \sum_m a_m n_{O,m}}{\sum_r a_r \left(2n_{C,r} + \frac{1}{2}n_{H,r} \right)} \quad (8)$$

where k , m , and r are indices over all fuel, oxidizer, and fuel-plus-oxidizer reactant species, respectively. (In mathematical terms, if each species in the reactants has a unique index then indices k and m have no intersection and their union is the set of indices r .) The first summation in the numerator of Eq. 8 accounts for the contribution to the total oxygen ratio from oxygen in all fuel species. The second summation in the numerator accounts for the contribution to the total oxygen ratio from oxygen in all oxidizer species. The summation in the denominator quantifies the total amount of oxygen required for stoichiometric combustion, *i.e.*, the amount of oxygen required to convert all carbon to carbon dioxide and all hydrogen to water. The first summation in the numerator of Eq. 8 divided by the denominator is called the oxygen ratio of the

fuel and is denoted Ω_f . The second summation in the numerator of Eq. 8 divided by the denominator is called the oxygen ratio of the oxidizer and is denoted Ω_{ox} . Thus, the total oxygen ratio of a given mixture can be expressed as the sum of the oxygen ratio of the fuel and the oxygen ratio of the oxidizer:

$$\Omega = \Omega_f + \Omega_{ox}. \quad (9)$$

The distinction between oxygen from the fuel and oxygen from the oxidizer is used in the following section to help show how the oxygen ratio is related to the equivalence ratio.

3. Properties and Proper Use of the Oxygen Ratio

By definition, the oxygen ratio is unity when the equivalence ratio is unity. It can be shown that the relationship between the oxygen ratio and the equivalence ratio is

$$\Omega = \Omega_f + \frac{1 - \Omega_f}{\phi} \quad (10)$$

when the oxidizer species (*i.e.*, compounds for which $S_i < 0$ in Eq. 6) contain no hydrogen or carbon. Equation 10 shows that the oxygen ratio is simply the reciprocal of the equivalence ratio when the fuel does not contain oxygen (*i.e.*, $\Omega_f = 0$). The oxygen ratios of some fuels of interest are presented in Table 2. Note that Ω_f cannot exceed unity because then the compound would be an oxidizer rather than a fuel. Figure 2 uses Eq. 10 to show the relationship between the oxygen ratio and the reciprocal of the equivalence ratio for hydrocarbon fuels as well as oxygenated fuels with Ω_f equal to 10% and 50%. It is clear from Fig. 2 that the three types of fuels will have different actual mixture stoichiometries for any given $\phi \neq 1$.

Using Eq. 10, the error (ϵ) involved when ϕ is used as a measure of mixture stoichiometry can be shown to be:

$$\varepsilon = \frac{\phi^{-1} - \Omega}{\Omega} = \left[\frac{1}{\Omega_f(1-\phi)} - 1 \right]^{-1}. \quad (11)$$

Error values (greater than/less than) zero indicate that a mixture is (richer/leaner) than its ϕ -value would suggest. Figure 3 shows a plot of Eq. 11 as a function of ϕ for fuels with oxygen ratios of 0%, 1%, 10%, and 50%. For fuels with $\Omega_f = 0$ (e.g., hydrocarbon fuels), ϕ is a perfect measure of mixture stoichiometry, but errors in mixture stoichiometry can be large when $\Omega_f \neq 0$. Figure 3 also shows that a non-stoichiometric mixture with $\Omega_f \neq 0$ will always be closer to its stoichiometric condition than its ϕ -value would indicate.

The oxygen ratio is a measure of the proximity of a reactant mixture to its stoichiometric condition. This is its sole purpose, and it accomplishes this purpose for a broader set of reactants than the equivalence ratio. It is not a parameter that should be expected to correlate all combustion observables by itself. For example, the definition of the oxygen ratio requires that atoms bound in stable species are neglected. While classified as stable, H_2O and CO_2 will participate in reactions at sufficiently elevated temperatures (e.g., water-gas shift reactions and dissociation). The rate of such reactions will depend strongly on mixture temperature and other variables. While the oxygen ratio may affect these variables, one should not expect to completely capture the physics of temperature-dependent processes with the oxygen ratio since the oxygen ratio is independent of temperature. This should be kept in mind in the study of reactant mixtures containing large or variable concentrations of H_2O or CO_2 at high temperatures. In such situations, additional parameters (e.g., H_2O concentration, product temperature) should be used in the interpretation of the results. Likewise, since the oxygen ratio contains no information about the arrangement of atoms within a given reactant molecule, one should not expect that it would capture molecular-structure effects.

One application that is well suited to the use of the oxygen ratio would be a comparison of the effects of mixture stoichiometry on adiabatic flame temperature for a hydrocarbon and an oxygenated fuel. For example, consider CH_4/O_2 and CO/O_2 mixtures like those discussed in the Introduction. Figure 4 shows adiabatic flame temperature results for the two fuels, using ϕ and Ω as measures of mixture stoichiometry, for comparison. The calculations were performed using the EQUIL module of the CHEMKIN software package [10], which is available from Reaction Design, Inc. The initial mixture temperatures and pressures are 25°C and 101 kPa, respectively.

Figure 4a shows what appears to be an interesting trend wherein the CO/O_2 mixtures have higher adiabatic flame temperatures outside given lean and rich boundaries, but lower values within the boundaries. This could spawn a research effort to determine the exact numerical values of the boundary points, reasons for the crossovers, and so on. Figure 4b shows the data from Fig. 4a re-plotted as a function of the oxygen ratio. The crossovers observed in Fig. 4a are gone because they were simply due to ϕ being an inaccurate measure of the CO/O_2 mixture stoichiometry. Adiabatic flame temperature values for the CO/O_2 mixtures are always less than those for the CH_4/O_2 mixtures when the comparison is made at the same mixture stoichiometry (*i.e.*, the same oxygen ratio). The results in Fig. 4b are consistent with the error analysis above that showed that a non-stoichiometric mixture with $\Omega_f \neq 0$ will always be closer to its stoichiometric condition than its ϕ -value would indicate. By properly accounting for the effects of mixture stoichiometry, the researcher can more readily focus on the reasons for the observed differences in adiabatic flame temperature. Problems caused by using by ϕ as a measure of mixture stoichiometry have often occurred in studies of the sooting tendencies of oxygenated fuels, and will likely be important in other studies of combustion processes that use oxygenated fuels.

4. Summary and Conclusion

This paper shows how the equivalence ratio fails to properly quantify mixture stoichiometry (*i.e.*, the proximity of a mixture to its stoichiometric condition) when oxygenated fuels are used. The concept of an “oxidizer ratio” as determined from basic chemistry considerations was explored to resolve the problem with complete generality, but the calculation of such a parameter was found to be cumbersome. The oxygen ratio was introduced as a good compromise between complete generality and ease of use.

The oxygen ratio of a mixture is defined as the amount of oxygen in the mixture divided by the amount of oxygen required for stoichiometric combustion of the same quantity of fuel, where atoms bound in stable species are neglected. The oxygen ratio is a valid measure of mixture stoichiometry in any mixture for which the saturated stoichiometric products are CO₂, H₂O, N₂, and/or noble gases (NG). The oxygen ratio is straightforward to calculate and yet general enough to apply to most practical combustion systems of interest, including those using oxygenated fuels.

A detailed mathematical definition of the oxygen ratio is provided. The definition is used to show the relationship between the oxygen ratio and the equivalence ratio, and to quantify the errors involved when the equivalence ratio is used as a measure of mixture stoichiometry with oxygenated fuels. An example application is provided to show how the oxygen ratio can simplify data analysis when it is used as an independent variable to characterize mixture stoichiometry. It is recommended that the oxygen ratio be used to quantify the proximity of a reactant mixture to its stoichiometric condition whenever oxygenated fuels are used.

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Element	Oxidation Number, <i>s</i>
Carbon, <i>C</i>	+4
Hydrogen, <i>H</i>	+1
Oxygen, <i>O</i>	-2
Nitrogen, <i>N</i>	0
Noble Gases, <i>NG</i>	0

Table 1. Oxidation numbers of elements of interest in saturated stoichiometric products (SSPs)

Fuel Name (Chemical Formula)	Oxygen Ratio of Fuel, Ω_f
Hydrocarbon (C_nH_m)	0%
Ethanol (C_2H_5OH)	14.3%
Methanol (CH_3OH)	25.0%
Carbon monoxide (CO)	50.0%
Nitromethane (CH_3NO_2)	57.1%
Ethylene glycol di-nitrite ($C_2H_4(NO_3)_2$)	100.0%

Table 2. Oxygen ratios of some fuels of interest

Figure Captions

Figure 1. Logic diagram for the classification of a reactant species as a fuel, oxidizer, or stable compound.

Figure 2. Relationship between reciprocal of equivalence ratio and oxygen ratio for hydrocarbon ($\Omega_f = 0$) and oxygenated ($\Omega_f \neq 0$) fuels, from Eq. 10. Proximity to stoichiometric mixture as indicated by equivalence ratio is inaccurate and depends on fuel type when oxygenated fuels are used.

Figure 3. Percentage error involved when equivalence ratio is used as a measure of mixture stoichiometry. Error is zero for hydrocarbon fuels, but increases rapidly when oxygenated fuels are used. Error is such that a non-stoichiometric oxygenated-fuel mixture is always closer to its stoichiometric condition than its equivalence ratio would indicate.

Figure 4. Adiabatic flame temperatures of CH_4/O_2 and CO/O_2 mixtures as a function of **a.** equivalence ratio and **b.** oxygen ratio. Data analysis is simplified when oxygen ratio is used to properly account for effects of variations in mixture stoichiometry.

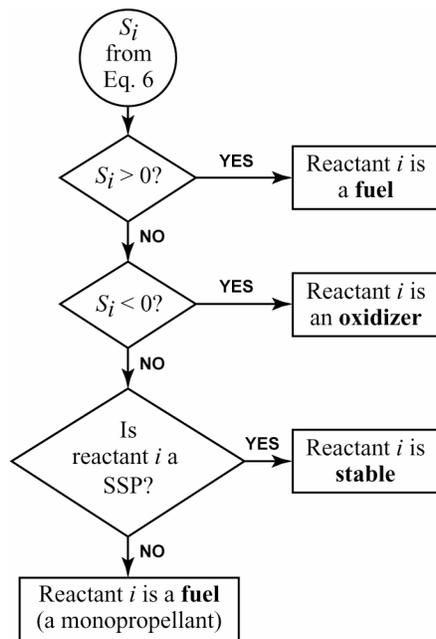


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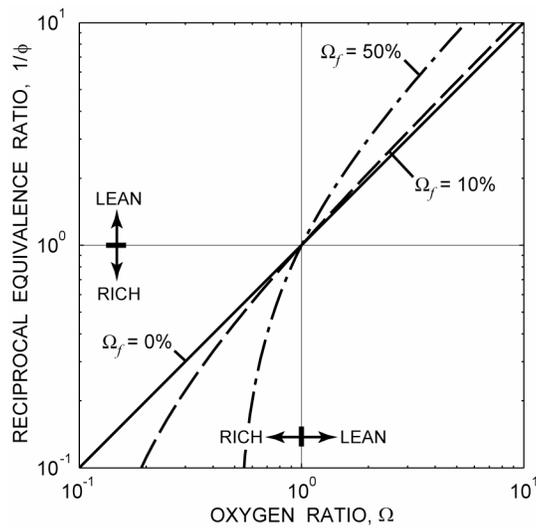


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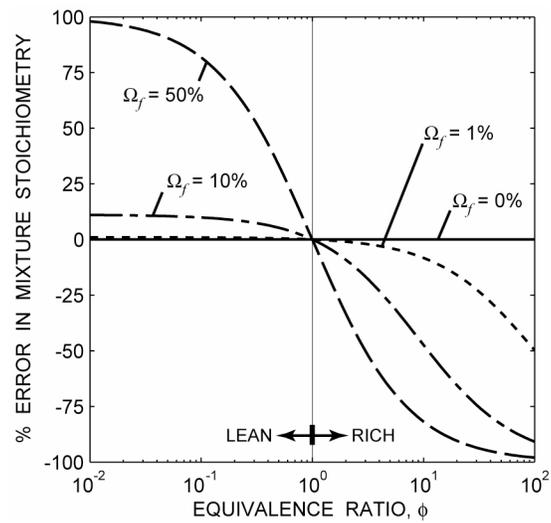


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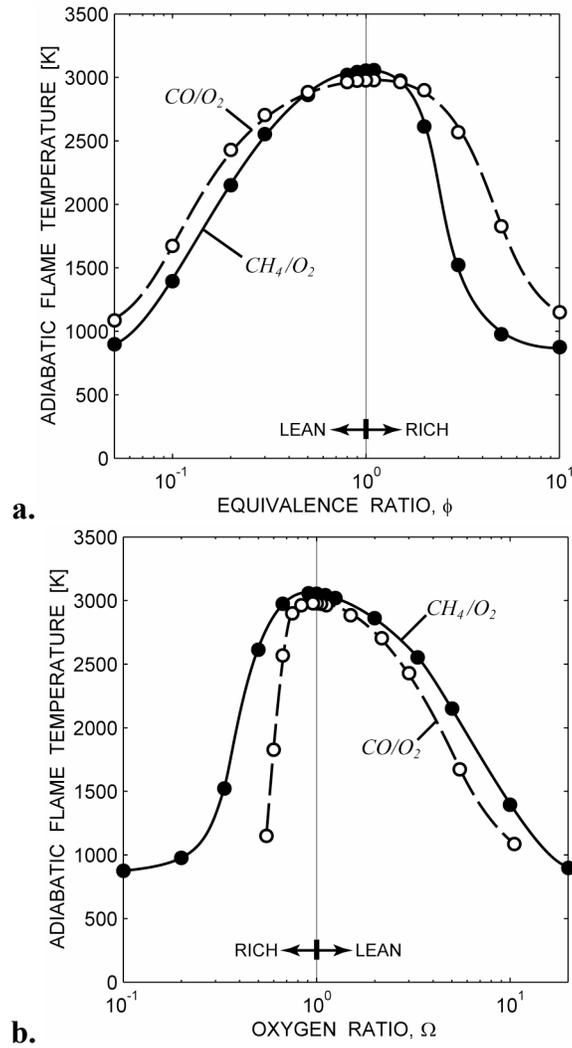


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