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A Novel Method for Evaluating the Chemical Effect of Diluents on Burning Velocity of Premixed Methane/Air Flames

It is proposed a new method to determine the effects of diluents on laminar burning velocity in premixed methane/air flames. The present study shows in what way the chemical effect of dilution with CO_2 can be determined using thermal theory of flame propagation without the computer code GRI-Mech3.0. This must be compared not only the effect of CO_2 and N_2 with equal dilution ratio, but also the effect of those diluents in mixtures with the same adiabatic flame temperature. The new method is also applicable to not enough studied combustion systems including which the computer code GRI-Mech3.0 is not built for now. *Bibl. 8, Fig. 2, Table 5.*

Key words: diluents, burning velocity, chemical effect.

Introduction

Comprehensive experimental and numerical data on the effects of typical diluents in premixed methane/air flames were published recently [1, 2]. The effects were studied in terms of measured (S^0_L) or computed (S_L) laminar burning velocity. Several diluents have been tested: N_2, CO_2, H_2O (vapor), inert gases Ar and He.

Experiments [1] were carried out in constant volume bomb (24.3 L) under spherically expanding flames at atmospheric pressure and T_u = 393 K. Such inlet temperature was chosen to full vaporization of water. Laminar burning velocity decreases from 56 to 14 cm/s at growth of CO_2 dilution ratio from 0 to 20 %.

Simulations were based on GRI Mechanism and PREMIX code of the CHEMKIN package. As a result of this study the chemical effect of dilution was found only with CO_2. Its value amounts up to 56 % at dilution percentage 5 %. With grows of dilution ratio the chemical effect gradually vanishes and becomes negligible.

The effects of diluents CO_2, N_2, Ar, and He on computed laminar burning velocity were studied numerically [2] at normal temperature and pressure conditions (NTP) using the CHEMKIN package. The results show that diluents will reduce the flame temperature and the concentrations of radicals H, OH, and O.

It should be noted that the chemical effect of CO_2 dilution on premixed methane flames was before studied theoretically [3] by comparison of two numerical solutions: one for replacement of N_2 in air (equivalence ratio ER = 1) by real CO_2

and other for the same replacement by «inert» CO_2. «Inert» CO_2 or according authors terminology «fictitious molecule CO_2» possess thermal and transport characteristics of real CO_2 but does not react in the kinetic scheme of GRI Mechanism. It is appeared that in case of «inert» CO_2 burning velocity is always higher in comparison with real CO_2. For example, in result of replacement 30 % N_2 in combustion air by real CO_2 burning velocity decreased from 39 to 17 cm/s, however, by «inert» CO_2 to 25 cm/s. The dominant reaction pathway for chemical participation of real CO_2 in a flame is H + CO_2 ↔ CO + OH. The competition of CO_2 for H atoms through this reaction via the most important chain branching reaction H + O_2 ↔ OH + O plays a chemically inhibiting role that reduces overall rate of combustion. Sensitivity analyses show this reaction exhibits the largest sensitivities over the entire dilution range.

In all works the chemical effect is determined with diluents equal on volume or at the same dilution ratio. The dilution ratio is defined as β = D/(CH_4 + 2 O_2 + 7.52 N_2 + D), where CH_4, O_2, N_2, and D are mole fractions of methane, air (O_2 + 3.76 N_2), and diluents in mixture (ER = 1). However, equal on volume diluents give a burning mixtures with non equal flame temperature because of a difference their molar heat capacity. Therefore, the determination of the chemical effect is complicated by strong (exponential) influence of flame temperature on burning velocity and hence on the effect of dilution. On this account it is necessary to introduce such hypothetical diluents as «fictitious» [1] or «false» [2]

molecules CO_2» (F-CO_2 ≡ GRI-CO_2) and then to use corresponding kinetic mechanism.

In premixed methane/air flames the chemical effect of CO_2 dilution remains significant in the narrow temperature «window» (T_b = 2150−2050 K) and reaches 40−60 %. The chemical effect has practical significance, since CO_2 is a part of biogas (CO_2 = 35−40 %) and used to NO_x suppression by recirculation of combustion products.

Method of equal diluents ($β_{N2}$ = $β_{CO2}$)

Experimental data on laminar burning velocity CH_4/Air flames with different diluents are presented in Tables 1 and 2 where different diluents are compared on their ability to reduce flame propagation.

These data confirm the well known order of diluents activity: CO_2 > H_2O > N_2 > Ar > He. The same order exists for molar heat capacities (J/(mol·K) at 393 K) those diluents: 41.4 (CO_2) > 34.0 (H_2O) > 29.1 (N_2) > 20.9 (Ar and He). From the standpoint of modern theory to better compare the adiabatic flame temperatures T_b. For example, with dilution ratio β = 10 %

Table 1: Measured [1] laminar burning velocity of CH_4/Air preheated spherical flames (Conditions: T_u = 393 K, ER = 1, P_u = = 0.1 MPa)

Diluents	Molar dilution ratio (relative to mixture) β, %						
	5	10	15	20	25	30	$β_{limit}$[1)
Laminar burning velocity S^0_L, cm/s							
CO_2	39[2)	26	20	14	–	–	24
H_2O	46	38	29	21	15	–	29
N_2	48	43	34	30	23	18	38
Ar	52	45	41	35	30	25	51
He	54	49	44	41	36	32	40

Notes: [1) Maximum dilution ratio corresponding to the low explosion limit of premixed methane/air flame; [2) Values S^0_L are rounded to integer number, since ones were taken from Figures with the scale 5 cm/s. Without dilution (β = 0) velocity S^0_L = 55.6 cm/s.

Table 2: Calculated [2] laminar burning velocity of CH_4/Air spherical flames (ER = 1) at NTP conditions

Diluents	Molar dilution ratio (relative to mixture) β, %					
	5	10	15	20	25	30
Laminar burning velocity S_L, cm/s[1)						
CO_2	25.8	17.8	12.0	7.8	–	–
N_2	31.5	26.7	22.1	17.8	13.9	10.4
Ar	33.1	29.5	26.0	22.6	19.3	16.0
He	35.2	33.4	31.3	28.9	26.2	23.2

Notes:[1) Non-dilution condition (β = 0) velocity S_L = 36.7 cm/s.

we have respectively the following distribution of T_b (T_u = 393 K): 2078 K (CO_2) < 2118 K (H_2O) < 2162 K (N_2) < 2201 K (He and Ar). Adiabatic flame temperature is identical with dilution of Ar and He, but the burning velocity in the case of He is higher than Ar because of considerably more high thermal and mass diffusivity of helium.

Concentrations of reactants (CH_4 and O_2) and products (CO_2 and H_2O) are clearly lower for diluted flame comparing to undiluted flame. Authors of the work [1] proposed to evaluate the chemical (ch) and the thermal (th) effects (E) via the difference [S_L(GRI-CO_2) − S^0_L(CO_2)]:

$$E_{ch} = [S_L(GRI\text{-}CO_2) − S^0_L(CO_2)]/ /[S^0_L(N_2) − S^0_L(CO_2)]; \quad (1)$$

$$E_{th} = [S^0_L(N_2) − S_L(GRI\text{-}CO_2)]/ /[S^0_L(N_2) − S^0_L(CO_2)]. \quad (1, a)$$

Here nitrogen is a marker for comparison under the condition $β_{N2}$ = $β_{CO2}$ (equal diluents). Thus, effect of a trivial dilution is removed from initial factors. In case of air (O_2 + 3.76 N_2) namely nitrogen should be the marker. Indeed, undiluted methane-air mixture may be regarded as CH_4 + 2 O_2 mixture diluted by 7.52 N_2 resulting that S^0_L is reduced from 330 to 37 cm/s.

Necessary to underline a key role of the computational burning velocity S_L (GRI−CO_2), because only if S_L (GRI−CO_2) > S^0_L (CO_2) the chemical effect will be non-zero.

Similar formula for the chemical effect is used in the article [2]

$$E_{ch} = [S_L(GRI\text{-}CO_2) − S^0_L(CO_2)]/ /[S^0_L(β = 0) − S^0_L(CO_2)]. \quad (2)$$

Here the non-dilution condition ($β_{CO2}$ = 0) is chosen as a basis for comparison instead of N_2. Equation (2) is less convincing and gives the underestimated results.

Method of equivalent diluents (T_{bN2} = T_{bCO2})

Now will move directly to the purpose of this study and show how the chemical effect may be found without rather artificial conception based on the fictitious molecule CO_2.

In ZFK (Zeldovich, Frank-Kamenetsky) approach of chemical reaction within a narrow flame zone the main parameter of flame propagation is

Table 3: The chemical effect determined by the method of equivalent diluents (Conditions: ER = 1, T_u = 393 K, $T_{b, \beta=0}$ = 2271 K)

Given parameters [1]				
Dilution ratio β_{CO2}, %	5	10	15	20
Flame temperature $T_b(CO_2)$, K	2172.5	2075.2	1977.2	1876.9
Thermal diffusivity $\chi_b(CO_2)$, cm²/s	6.09	5.46	4.88	4.34
Equivalent ($T_{b\,N2} = T_{b\,CO2}$) parameters				
Dilution ratio β_{N2}, %	8.9	16.5	23.3	29.8
Flame temperature $T_b(N_2)$, K	2172.5	2075.7	1977.7	1876.1
Thermal diffusivity $\chi_b(N_2)$, cm²/s	6.23	5.73	5.26	4.80
Coefficient $m_b(N_2)$	0.989	0.976	0.963	0.951
S^0_L(GI-N₂)[1]), cm/s	44.0	34.1	25.1	16.7
S_L(ZFK-CO₂), cm/s	44	33	24	16
S_L(GRI-CO₂), cm/s	44	33	25	17
The chemical effect of dilution with CO_2, %				
By GI-method[2]) via S_L(ZFK-CO₂)	56	41	29	14
By GRI-method[3]) via S_L(GRI-CO₂)	56	41	36	19

Note: [1]) Used the approximation of data [1]: $S^0_L(N_2)$ = 55.6−1.305 β_{N2}, cm/s; [2]) The Gas Institute (GI), Ukraine; [3]) The Gas Research Institute (GRI), USA.

Table 4: The chemical effect determined by the method of equivalent diluents (Conditions: ER = 1, T_u = 298 K, $T_{b, \beta=0}$ = 2225 K)

Given parameters [2]				
Dilution ratio β_{CO2}, %	5	10	15	20
Flame temperature $T_b(CO_2)$, K	2123.6	2022.7	1920.9	1816.9
Thermal diffusivity $\chi_u(CO_2)$, cm²/s	0.220	0.213	0.206	0.199
Equivalent ($T_{b\,N2} = T_{b\,CO2}$) parameters				
Dilution ratio β_{N2}, %	8.6	16.2	23.0	29.4
Flame temperature $T_b(N_2)$, K	2123.9	2022.2	1919.4	1815.9
Thermal diffusivity $\chi_u(N_2)$, cm²/s	0.227	0.227	0.227	0.227
Coefficient $m_u(N_2)$	0.984	0.968	0.953	0.937
S_L(GI-N₂), cm/s	28.0	21.0	15.2	10.8
S_L(ZFK-CO₂), cm/s	27.6	20.3	14.5	10.1
S_L(GRI-CO₂), cm/s	28.5	21.4	15.5	10.6
The chemical effect of dilution with CO_2, %				
By GI-method[1]) via S_L(ZFK-CO₂)	31.6	28.1	34.7	28.0
By GRI-method[2]) via S_L(GRI-CO₂)	47.4	40.4	34.7	28.0

Note: [1]) The Gas Institute (GI), Ukraine; [2]) The Gas Research Institute (GRI), USA.

adiabatic flame temperature T_b. As is generally known, methane burning velocity in inverse ratio depends on root pressure (mainly through the density ρ_u of initial mixture).

So, for first order reaction (n = 1) we have the classical ZFK solution [4]:

$$\rho_u \, S_L{}^2 = 2 \, (\lambda_b/C_{pb}) \, (\rho_b/\rho_u) \times$$
$$\times \, Ze^{-2} \, k_0 \cdot \exp \, (-E/RT_b) \approx \qquad (3)$$
$$\approx 2 \, (\lambda_b/C_{pb}) \, (T_u/T_b) \times$$
$$\times \, Ze^{-2} \, k_0 \cdot \exp \, (-E/RT_b). \qquad (3,a)$$

Zeldovich number $Ze \equiv E \, (T_b - T_u)/RT_b{}^2$ serves as a criterion for burning similarity.

Expressions (3, 3a) may be simplified by introducing the thermal diffusivity of combustion products $\chi_b = \lambda_b/(\rho_b C_{pb})$ at temperature T_b

$$S_L{}^2 = 2 \, \chi_b(\rho_b/\rho_u)^2 \, Ze^{-2} \, k_0 \cdot \exp \, (-E/RT_b) \approx$$
$$\approx 2 \, \chi_b(T_u/T_b)^2 \, Ze^{-2} \, k_0 \cdot \exp \, (-E/RT_b). \qquad (4)$$

It should be noted an approximate character of the correlation $(\rho_b/\rho_u) \approx (T_u/T_b)$, which is correct only for weak dissociation of combustion products. Exact correlation looks like $(\rho_b/\rho_u) = (T_u/T_b) \times (M_b/M_u)$ and takes into account the change of molecular mass in result of burning ($2 \, H_2 + O_2 = 2 \, H_2O$) or dissociation ($T_b > 2500$ K).

For example, in case of CH_4/Air we have $(T_u/T_b) = 298/2225 = 0.134$ and also $(\rho_b/\rho_u) = 0.150/1.131 = 0.133$; but in case of CH_4/O_2 will be $(T_u/T_b) = 298/3054 = 0.0976$, that differs from $(\rho_b/\rho_u) = 0.0858/1.091 = 0.0786$ and we need to make the correction $(T_u/T_b)(M_b/M_u) = 0.0976 \, (21.51/26.68) = 0.0787$.

Let consider a task of premixed combustion under the condition of the same adiabatic flame temperature T_b [5]. The next relation is obtained for burning velocities with CO_2 dilution and *equivalent nitrogen* (GI-N₂) as the marker (reference case)

$$S_L(ZFK\text{-}CO_2) \approx [\chi_b(CO_2)/$$
$$/\chi_b(N_2)]^{0.5} \, S^0_L(GI\text{-}N_2). \qquad (5)$$

Temperature factor $(T_u/T_b) \, Ze^{-2}$ is reduced since should be compared burning mixtures with identical initial T_u and final T_b temperature. The rate constant of overall chemical reaction $k_0 \cdot \exp \, (-E/RT_b)$ is also reduced because namely inert diluents are examined for the same fuel.

Thus, correlation (5) may be rewritten in more simple form:

$$S_L(ZFK\text{-}CO_2) = m_b(N_2) \, S^0_L(GI\text{-}N_2), \quad (6)$$

where the coefficient $m_b(N_2)$ is

$$m_b(N_2) \approx [\chi_b(CO_2)/\chi_b(N_2)]^{0.5}. \qquad (7)$$

Table 5: The test problem: He (diluent) — N_2 (marker) (Conditions: ER = 1, T_u = 298 K, $T_{b,\beta=0}$ = 2225 K)

Given parameters [2]						
Dilution ratio β_{He}, %	5	10	15	20	25	30
Flame temperature T_b(He), K	2189.3	2149.7	2106.2	2057.6	2004.8	1945.5
Thermal diffusivity χ_u(He), cm2/s	0.252	0.279	0.310	0.344	0.381	0.423
Equivalent ($T_{b\,He} = T_{b\,N2}$) parameters						
Dilution ratio β_{N2}, %	3.18	6.56	10.0	13.7	17.4	21.4
Flame temperature $T_b(N_2)$, K	2189.5	2149.4	2106.7	2057.2	2004.7	1945.0
Thermal diffusivity $\chi_u(N_2)$, cm2/s	0.227	0.227	0.227	0.227	0.227	0.227
Coefficient m_u(He)	1.053	1.109	1.167	1.230	1.295	1.364
S_L(GI-N_2), cm/s	33.4	30.2	26.7	23.4	20.1	16.7
S_L(ZFK-He), cm/s	35.2	33.5	31.2	28.8	26.0	22.8
S_L(GRI-He), cm/s	35.2	33.4	31.3	28.9	26.2	23.2

The coefficient $m_b(N_2)$ is rather closed to unit: $m_b(N_2)$ = 0.95−1.0. In general, if molecular mass of diluents (M_D) is greater than mass of the marker N_2 (M_{N2}), the coefficient $m_b(N_2)$ < 1 (M_D > M_{N2}), and conversely.

The results of new method are represented in Tables 3, 4.

Accuracy of the laminar burning velocity determination and deviation of coefficient $m_b(N_2)$ from unit have the same value. So, for a rough estimate it is possible to take S_L(ZFK-CO_2) ≈ ≈ S^0_L(GI-N_2). But important only that diluents will be *equivalent*, that ones are should be selected under the condition of the same burning temperature $T_b(N_2)$ = $T_b(CO_2)$.

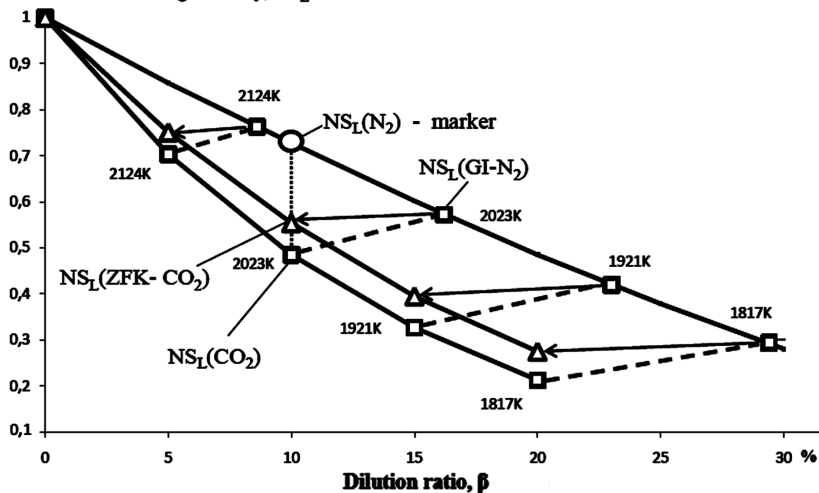**Normalized burning velocity, NS$_L$**

Fig. 1. Determining the chemical effect of dilution with CO_2 by the GI-method. Calculated [2] burning velocities are normalized about the S_L = 36.7 cm/s (non-dilution condition); the equivalent points are connected by the dotted line; the correlation S_L(ZFK-CO_2) = $m_u(N_2)·S_L$(GI-N_2) is shown by the arrows, $m_u(N_2)$ < 1; difference between the points NS$_L$(ZFK-CO_2) and NS$_L$(CO_2) characterizes the chemical effect; value of the chemical effect see Table 4.

Now, the chemical effect of dilution whis CO_2 is determined by the equation

$$E_{ch} = [S_L(ZFK\text{-}CO_2) - S^0_L(CO_2)]/$$
$$/[S^0_L(N_2) - S^0_L(CO_2)] \approx$$
$$\approx [m_b(N_2)\, S^0_L(GI\text{-}N_2) -$$
$$- S^0_L(CO_2)]/[S^0_L(N_2) - S^0_L(CO_2)]. \quad (8)$$

In the numerator enter *equivalent diluents* ($T_{bN2} = T_{bCO2}$) and in the denominator — *equal diluents* ($\beta_{N2} = \beta_{CO2}$).

This level of problem understanding has been made in our article [5], when the very important paper [2] was published. Chinese scientists have made a complete analysis of the thermal and chemical effects CO_2 dilution on computed laminar burning velocity S_L. One of the advantages their study (see Table 2) is the tabulation of computed laminar burning velocity S_L(GRI-CO_2), S_L(CO_2), and $S_L(N_2)$ according to dilution ratio β_{CO2}. Thus, it is become possible to correct comparison of both methods.

A modern version of the thermal theory of flame propagation [6] is used in paper [2]. According to the modern version of thermal theory S_L depends on thermal diffusivity of inlet mixture χ_u instead of combustion products χ_b as in ZFK theory. This difference is insignificant, since the coefficients $m_b(N_2)$ and $m_u(N_2)$ differ little from each other.

As see from Tables 3 and 4, ZFK theory of flame propagation gives the results identical to the kinetic mechanism GRI-Mech3.0. Burning velocities S_L(ZFK-CO_2) and S_L(GRI-CO_2) coincide practically. Advantage of ZFK theory consists in determination of the chemical effect via experimental burning velocity S^0_L (N_2) without rather artificial «fictitious» or «false» its values.

Figure 1 shows the graphical procedure of proposed method.

The test problem for GI-method

In order to test a novel method the problem with known solution was proposed. The best pair for this is inert couple: He (diluent) — N_2 (marker). Helium in its physical properties is most strong differs from other diluents in the set of
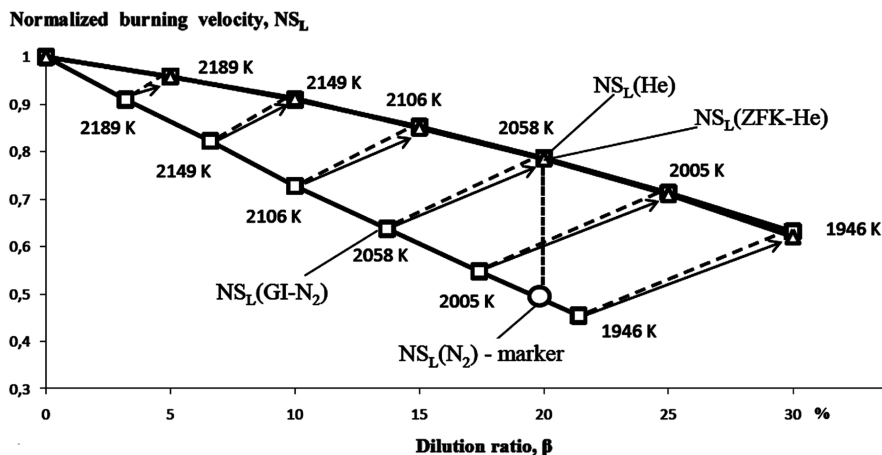
Normalized burning velocity, NS_L

Fig. 2. The test problem: He (diluent) − N_2 (marker). Calculated [2] burning velocities are normalized about S_L = 36.7 cm/s (non-dilution condition); the equivalent points are connected by the dotted line; the correlation S_L(ZFK-He) = m_u(He)·S^0_L(GI-N_2) is shown by the arrows, m_u(He) > 1; difference between the points NS_L(ZFK-He) and NS_L(He) is negligible, so the chemical effect is absent.

Ar, CO_2, N_2, and H_2O. For helium, thermal diffusivity of inlet mixture χ_u(He) increases with dilution ratio β_{He} (Table 5). In this case, the coefficient m_u(He) changes more significant to verify the applicability of ZFK theory. Besides, helium is preferable to other diluents since the greatest dilution ratio (up to 30 %) may be achieved namely with He.

In result of the test problem He-N_2 we would expect to receive S_L(ZFK-He) = S_L(He). This means absence of the chemical effect in case of dilution with helium. The data given in Table 5 and illustrated in Figure 2 fully confirm this conclusion.

In Figure 2 the arrows directed upwards, since molecular mass of helium (M_{He} = 4) is significantly smaller than mass of the marker (M_{N2} = 28), while in Figure 1 the arrows directed slightly downwards, because mass of carbon dioxide M_{CO2} = 44 > 28.

In this paper, we have considered only stoichiometric CH_4/Air mixtures. The next task is to extend a possible value of fuel/air ratio for lean and rich mixtures just as it was done in the article [7] for dilution of methane with water vapor.

Finally, it is interesting to mark that surprisingly large effect of CO_2 addition on burning velocity was first discovered a long time ago [8], and the following was written: «under the extreme conditions of the flame carbon dioxide can interfere with the chemical reactions, for example, the reaction CO_2 + H ↔ CO + OH might replace rapidly diffusing hydrogen atoms (D_H = 1.06 cm^2/s) by heavier and larger hydroxyl radicals (D_{OH} = 0.26 cm^2/s)». According this view the effect CO_2 dilution is rather physical than chemical.

Summary

Among the typical diluents CO_2, N_2, Ar, and He, which suppress combustion because of decrease in adiabatic flame temperature, only carbon dioxide has the chemical effect on laminar burning velocity.

It is possible to find the chemical effect of CO_2 dilution on burning velocity without rather artificial conception based on a fictitious molecule CO_2 (GRI-method). This necessary to compare not only the effect of diluents with equal dilution ratio, but also the effect of those diluents in mixtures with equal adiabatic flame temperature (GI-method).

Advantage of the GI-method consists in determination of the chemical effect via experimental burning velocities without rather artificial «fictitious» or «false» its values. Novel method is applicable also to not enough studied combustion systems, including which the computer code GRI-Mech3.0 is not built for now.

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Новый метод определения химического действия разбавителей на скорость распространения пламени в смеси метана с воздухом

Предложен новый метод определения химического действия разбавителей на нормальную скорость распространения пламени в смеси метана с воздухом. Показано, как можно найти химическую составляющую действия CO_2 без использования компьютерного кода GRI-Mech3.0. Для этого надо сравнить действие не только равных по объему добавок CO_2 и N_2, но и тех же добавок при одинаковой теоретической температуре горения. Новый метод применим также к процессу горения недостаточно изученных систем, для которых пока не разработан компьютерный код. *Библ. 8, рис. 2, табл. 5.*
Ключевые слова: скорость распространения пламени, влияние разбавителей, химический эффект добавки CO_2.

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Новий метод визначення хімічної дії розріджувачів на швидкість розповсюдження полум'я у суміші метану з повітрям

Запропоновано новий метод визначення хімічної дії розріджувачів на нормальну швидкість розповсюдження полум'я у суміші метану з повітрям. Показано, яким чином можливо знайти хімічну складову дії CO_2 без використання комп'ютерного коду GRI-Mech3.0. Для цього треба порівняти дію не тільки рівних за об'ємом домішок CO_2 та N_2, але й тих самих домішок за умови однакової теоретичної температури горіння. Новий метод може також застосуватися до процесу горіння недостатньо вивчених систем, для яких ще не розроблено комп'ютерний код. *Бібл. 8, рис. 2, табл. 5.*
Ключові слова: швидкість розповсюдження полум'я, вплив розріджувачів, хімічна дія домішки CO_2.