SPECIFIC HEAT RATIOS AND ISENTROPIC EXPONENTS FOR CONSTANT-VOLUME COMBUSTION OF STOICHIOMETRIC MIXTURES OF HYDROGEN-OXYGEN DILUTED WITH HELIUM HYDROGEN

ANDRÉ BENOIT
UNIVERSITY OF TORONTO
TORONTO, CANADA

Contract No. AF 33(615)-2766
Project No. 7065

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United States Air Force
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INSTITUTE FOR AEROSPACE STUDIES
UNIVERSITY OF TORONTO
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AEROSPACE RESEARCH LABORATORIES
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UNITED STATES AIR FORCE
WRIGHT-PATTERSON AIR FORCE BASE, OHIO
FOREWORD

This interim technical report was prepared by Andre Benoit, University of Toronto, Canada on Contract AF33(615)-2766 for the Aerospace Research Laboratories, Office of Aerospace Research, United States Air Force. The research reported herein was accomplished on Task 7065-0015, "Fluid Dynamics Facilities Research" of Project 7065, "Aerospace Simulation Techniques Research" under the technical cognizance of Mr. John Goresh of the Fluid Dynamics Facilities Research Laboratory of ARL.

The author wishes to express his thanks to Dr. G. N. Patterson for the opportunity to complete this work.

He is also grateful to Dr. I. I. Glass who suggested the study reported herein, and to Professor J. Meinguet for the opportunity to perform the numerical calculations at the "Centre de Calcul Numerique" of the University of Louvain.
This note is complementary to UTIAS Technical Note No. 85, "Thermodynamic and Composition Data for Constant-Volume Combustion of Stoichiometric Mixtures of Hydrogen-Oxygen Diluted with Helium or Hydrogen", by A. Benoit. It includes the calculation of the equilibrium specific heats, the equilibrium specific heat ratios, the isentropic exponents, and the corresponding values of the speeds of sound. For convenience, the final-to-initial temperature ratio and the final-to-initial pressure ratio have also been included in the present tables. The results are presented for helium and hydrogen dilution respectively.
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NOTATION

\( a \)  
equilibrium speed of sound (m sec\(^{-1}\))

\( a_1 \)  
frozen speed of sound (m sec\(^{-1}\))

\( a^* \)  
fictitious speed of sound (m sec\(^{-1}\)) defined by (53) (*)

\( a_{ik} \)  
coefficients appearing in the linearized system ((15) to (17)) and given by relations ((18) to (29)).

\( b_{ik} \)  
coefficients appearing in the linearized system ((30) to (31)) and given by relations ((32) to (37)).

\( B_K \)  
functions of \( p \) and \( T \) defined by (9) and (10)

\( C_P \)  
molar equilibrium specific heat at constant pressure (cal mole\(^{-1}\) \(0^\circ\)K\(^{-1}\)) defined by (42)

\( C_{P1} \)  
molar frozen specific heat at constant pressure (cal mole\(^{-1}\) \(0^\circ\)K\(^{-1}\)) defined by (40)

\( C_{V1} \)  
molar frozen specific heat at constant volume (cal mole\(^{-1}\) \(0^\circ\)K\(^{-1}\))

\( d \)  
function of \( p \) and \( T \) defined by (39)

\( e \)  
function of \( p \) and \( T \) defined by (36)

\( H \)  
molar enthalpy (cal mole\(^{-1}\)) including sensible enthalpy and chemical energy at \(0^\circ\)K for gas state

\( K \)  
equilibrium constant based on partial pressures for reaction of formation from elements in gas state. The subscripts, 1, 2, 3 and 4 refer respectively to the formation of \( H_2O \), \( OH \), \( H_2 \) and \( O_2 \)

\( m \)  
number of moles of diluting hydrogen per mole of oxygen in reactants

(*) (53) refers to equation (53) etc.
number of moles of helium per mole of oxygen in reactants

number of moles of species "j" in reaction products per mole of oxygen in reactants

total number of moles of reactants per mole of oxygen in reactants \( (n_1 = m + n + 3) \)

total number of moles of products per mole of oxygen in reactants \( (n_f = \frac{m}{n_1} n_j) \)

pressure of reactants (atm)

pressure of products of reaction (atm)

universal gas constant \( (1.98718 \text{ cal mole}^{-1} \text{ } ^0\text{K}^{-1}) \)

universal gas constant \( (8.314. \text{ m}^2 \text{ sec}^{-2} \text{ } ^0\text{K}^{-1} \text{ gr}) \)

absolute temperature \( (^0\text{K}) \)

molar internal energy of reaction products (cal mole\(^{-1}\))

molar internal energy of reactants (cal mole\(^{-1}\))

isentropic exponent defined by (44)

frozen specific heat ratio defined by (49)

equilibrium specific heat ratio defined by (47)

molecular weight of products of reaction (gr. mole\(^{-1}\))

molecular weight of reactants (gr. mole\(^{-1}\))

molar fraction of species "j" in products of reaction

Subscripts

\( i \) refers to the reactants
j refers to species "j" according to the correspondence

1 for H$_2$O
2 for OH
3 for H$_2$
4 for O$_2$
5 for I$_2$
6 for O
7 for He

p at constant pressure

$\phi$ at constant entropy

v at constant volume
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1. **INTRODUCTION**

In order to complete the work on constant-volume combustion reported in Ref. 1, it was felt that the equilibrium quantities such as the specific heat, specific heat ratio, isentropic exponent and the equilibrium speed of sound should be computed and incorporated in the results. The same source of thermodynamic data was used (Ref. 2), but all the thermodynamic functions including the equilibrium constants were introduced in the program in the form of best-fitted analytical functions.

Instead of using the method of solution described previously (Ref. 1), the final thermodynamic conditions were obtained through an iteration procedure based on the linearization of the equations (Newton-Raphson iteration). This, at the same time, provided a verification of the results.

2. **BASIC EQUATIONS**

The general assumptions under which the calculations are performed have been described in Ref. 1.

The initial mixture

\[(2\text{H}_2 + \text{O}_2) + m \text{H}_2 + n \text{He}\]

at temperature \(T_1\) and the pressure \(p_1\), is transformed to give

\[n_1 \text{H}_2\text{O} + n_2 \text{OH} + n_3\text{H}_2 + n_4 \text{O}_2 + n_5 \text{H} + n_6 \text{O} + n_7 \text{He}\]

at temperature \(T\) and pressure \(p\). The symbols \(\text{H}_2\text{O}, \text{OH}, \text{etc.}\) represent one mole of \(\text{H}_2\text{O},\) one mole of \(\text{OH},\) etc. The equilibrium equations combined with the equations of conservation of mass of each chemical element yield the following equations:

\[
B_1 \mathbf{\Psi}_5^2 \mathbf{\Psi}_6 + B_2 \mathbf{\Psi}_5 \mathbf{\Psi}_6 + B_3 \mathbf{\Psi}_5^2 + B_4 \mathbf{\Psi}_5^2 \mathbf{\Psi}_6 + \mathbf{\Psi}_5 + \mathbf{\Psi}_6 + n \frac{n_f}{n_f} - 1 = 0 \tag{1}
\]

\[
2B_1 \mathbf{\Psi}_5^2 \mathbf{\Psi}_6 + B_2 \mathbf{\Psi}_5 \mathbf{\Psi}_6 + 2B_3 \mathbf{\Psi}_5^2 + \mathbf{\Psi}_5 - \frac{2(2 + m)}{n_f} = 0 \tag{2}
\]

\[
E_1 \mathbf{\Psi}_5^2 \mathbf{\Psi}_6 + B_2 \mathbf{\Psi}_5 \mathbf{\Psi}_6 + 2B_4 \mathbf{\Psi}_5^2 + \mathbf{\Psi}_6 - \frac{2}{n_f} = 0 \tag{3}
\]

and

\[
\mathbf{\Psi}_1 = K_1 \mathbf{\Psi}_5^2 \mathbf{\Psi}_6 \mathbf{p}^2 \tag{4}
\]

\[
\mathbf{\Psi}_2 = K_2 \mathbf{\Psi}_5 \mathbf{\Psi}_6 \mathbf{p} \tag{5}
\]

\[
\mathbf{\Psi}_3 = K_3 \mathbf{\Psi}_5^2 \mathbf{p} \tag{6}
\]

\[
\mathbf{\Psi}_4 = K_4 \mathbf{\Psi}_6^2 \mathbf{p} \tag{7}
\]
from which the composition can be determined when the temperature and pressure are known. In these relations, the \( \psi \)'s represent the molar concentrations, the K's are the equilibrium constants based on partial pressures and the subscripts of the K's refer to the following compounds: 1 to \( \text{H}_2\text{O} \), 2 to \( \text{OH} \), 3 to \( \text{H}_2 \) and 4 to \( \text{O}_2 \).

The B's are written for:

\[
B_1 = K_1 \rho_2 \\
B_j = K_j \rho \quad (j = 2, 3, 4)
\]

The two complementary equations required to determine the final pressure and temperature express the conservation of density and energy, i.e.,

\[
\rho = \rho_i \\
or \quad \frac{p}{p_i} = \frac{T_{nf}}{T_i n_i}
\]

and

\[
n_f U = n_i U_i \\
or \quad n_f (H - RT) = n_i (H_i - RT_i)
\]

3. METHOD OF SOLUTION

Equations (1) to (3) are linearized to provide a system from which \( \psi_5, \psi_6 \) and \( n_f \) are determined for any set of values, \( p, T \),

\[
a_{11} \psi_5 + a_{12} \psi_6 + a_{13} \delta \left( \frac{1}{n_f} \right) = a_{10}
\]

\[
a_{21} \psi_5 + a_{22} \psi_6 + a_{23} \delta \left( \frac{1}{n_f} \right) = a_{20}
\]

\[
a_{31} \psi_5 + a_{32} \psi_6 + a_{33} \delta \left( \frac{1}{n_f} \right) = a_{30}
\]

The coefficients \( a_{ij} \) are readily obtained

\[
a_{11} = 2B_1 \psi_5 + B_2 \psi_6 + 2B_3 \psi_5 + 1
\]
\[ a_{12} = B_1 \psi_5^2 + B_2 \psi_5 + 2B_4 \psi_6 + 1 \]  
\[ a_{13} = n \]  
\[ a_{10} = -(B_1 \psi_5^2 \psi_6 + B_2 \psi_5 \psi_6 + B_3 \psi_5^2 + B_4 \psi_6^2 + \psi_5 \psi_6 + \frac{n}{n_f} - 1) \]  
\[ a_{21} = 4B_1 \psi_5 \psi_6 + B_2 \psi_6 + 4B_3 \psi_5 + 1 \]  
\[ a_{22} = 2B_1 \psi_5^2 + B_2 \psi_5 \]  
\[ a_{23} = -2(2 + m) \]  
\[ a_{20} = -(2B_1 \psi_5^2 \psi_6 + B_2 \psi_5 \psi_6 + 2B_3 \psi_5^2 + \psi_5 \psi_6 - \frac{2(2 + m)}{n_f}) \]  
\[ a_{31} = 2B_1 \psi_5 \psi_6 + B_2 \psi_6 \]  
\[ a_{32} = B_1 \psi_5^2 + B_2 \psi_5 + 4B_4 \psi_6 + 1 \]  
\[ a_{33} = -2 \]  
\[ a_{30} = -(B_1 \psi_5^2 \psi_6 + B_2 \psi_5 \psi_6 + 2B_4 \psi_6^2 + \psi_6 - \frac{2}{n_f}) \]  

The linearized forms of equations (8) and (10) will provide the means of computing the temperature and the pressure

\[ b_{11} \delta p + b_{12} \delta T = b_{10} \]  
\[ b_{21} \delta p + b_{22} \delta T = b_{20} \]  

with

\[ b_{11} = \frac{-1}{p_i} - \frac{T}{T_i n_i} \left( \frac{\psi_{n_f}}{\delta p} \right) T \]  
\[ b_{12} = \frac{-1}{T_i n_i} \left( n_f + T \left( \frac{\psi_{n_f}}{\delta T} \right) \right) \]  
\[ b_{10} = \frac{p}{p_i} - \frac{T n_i}{T_i n_i} \]
\[ b_{21} = n_f e + (H - RT) \left( \frac{\partial n_f}{\partial T} \right)_p \]  \hspace{1cm} (35) \\
\[ b_{22} = n_f (C_{p_1} + d - R) + (H - RT) \left( \frac{\partial n_f}{\partial T} \right)_p \]  \hspace{1cm} (36) \\
\[ b_{20} = n_f (H - RT) - n_i (H_i - RT_i) \]  \hspace{1cm} (37) \\

where \[
\begin{align*}
    e &= \sum_{j=1}^{7} \left( \frac{\partial y_j}{\partial p} \right)_T \cdot H_j (T) \\
    d &= \sum_{j=1}^{7} \left( \frac{\partial y_j}{\partial T} \right)_p \cdot H_j (T)
\end{align*}
\]  \hspace{1cm} (38) \\
\]  \hspace{1cm} (39) \\

and \( C_{p_1} \) is the molar frozen specific heat, i.e.

\[ C_{p_1} = \sum_{j=1}^{7} y_j c_{pj} \]  \hspace{1cm} (40) \\

When the composition, the temperature and pressure have been determined, the following quantities are computed:

a) the molecular weight

\[ \mu = \sum_{j=1}^{7} y_j \mu_j \]  \hspace{1cm} (41) \\

b) the molar equilibrium specific heat

\[ C_p = \mu \left( \frac{\partial h}{\partial T} \right)_p \]  \hspace{1cm} (42) \\
\[ = C_{p_1} + d - \frac{H}{T} \left( \frac{\partial \ln \mu}{\partial \ln T} \right)_p \]  \hspace{1cm} (43) \\

c) the isentropic exponent

\[ \gamma = \left( \frac{\partial p}{\partial \mu} \right)_\mu \mu R_s T \]  \hspace{1cm} (44) \\
\[ = \left( \frac{\partial \ln P}{\partial \ln \mu} \right)_\mu \mu R_s T \]  \hspace{1cm} (45) \\
\[ = \left( 1 + \frac{\partial \ln P}{\partial \ln \mu} \right) \frac{R}{C_p} \left( 1 - \left( \frac{\partial \ln \mu}{\partial \ln T} \right)_p \right)^2 \]  \hspace{1cm} (46)
d) the equilibrium specific heat ratio

$$\gamma^* = \frac{C_p}{C_v}$$  \hspace{1cm} (47)

$$= \gamma (1 + \frac{\partial \ln \gamma}{\partial \ln \rho})_T$$  \hspace{1cm} (48)

e) the frozen specific heat ratio

$$\gamma_1 = \frac{C_p}{C_v}$$  \hspace{1cm} (49)

$$= \frac{C_p}{C_v}$$  \hspace{1cm} (50)

f) the frozen speed of sound (a), the equilibrium speed of sound (a) and a fictitious speed of sound computed using the equilibrium specific heat ratio instead of the isentropic exponent.

$$a_1 = R_o^{1/2} \left( \frac{\gamma_1 T}{\mu} \right)^{1/2}$$  \hspace{1cm} (51)

$$a = R_o^{1/2} \left( \frac{\gamma T}{\mu} \right)^{1/2}$$  \hspace{1cm} (52)

$$a^* = R_o^{1/2} \left( \frac{\gamma^* T}{\mu} \right)^{1/2}$$  \hspace{1cm} (53)

where $R_o$ is the universal gas constant for which the value 8314 m$^2$sec$^{-2}$ K$^{-1}$gr. has been used.

4. RESULTS

Computations have been performed for the initial conditions

$$T_1 = 298.15 \text{ K}$$

$$p_1 = 1. ; 5. ; 10. ; 30. ; 50. ; 100. ; 300. ; 500. \text{ atm.}$$

and the dilutions

1) m=0 and n varying from 0 to 12 in steps of 0.5
2) n=0 and m varying from 0 to 7.5 in steps of 0.5
The results are given Tables 1. and 2. and some are presented graphically in Figures 1. to 6. Incidentally, the intermediate results, p, T, n_f/n_i, C\textsubscript{p}, H\textsubscript{2}O, OH, ..., constitutes a verification of the data tabulated in Ref. 1 (*). A comparison is illustrated for the case T\textsubscript{i} = 298, 15 °K, p\textsubscript{i} = 1 atm., m = n = 0. The agreement is excellent.

<table>
<thead>
<tr>
<th>Quantity</th>
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<td>9.611</td>
</tr>
<tr>
<td>T/T\textsubscript{i}</td>
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<tr>
<td>n_f/n_i</td>
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<td>\mu</td>
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<tr>
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</table>

The effect of temperature on the composition (characterized by the partial derivatives of the molar fractions with respect to temperature) leads to values of the equilibrium specific heat (C\textsubscript{p}) appreciably higher than the values obtained neglecting the variations of the composition (C\textsubscript{p1}). The difference between C\textsubscript{p} and C\textsubscript{p1}

$$C_p - C_{p1} = \sum_{j=1}^{7} \left( \frac{\partial \ln \gamma_j}{\partial T} \right)_p \ln \frac{T}{T_j} - \frac{1}{T} \left( \frac{\partial \ln \mu_j}{\partial \ln T} \right)_p$$

(*) Note - In Ref. 1., The value used for R\textsubscript{0} was slightly larger than the accepted value, consequently all the values of a\textsubscript{f} and a\textsubscript{i} should be multiplied by the factor 0.9902 to obtain the correct values.
increases with decreasing initial pressure and decreases with increasing
dilution. The initial pressure has a much stronger influence on $C_p$ than on $C_{P1}$. 
For $m = n = 0$, $C_p$ is approximately five times as large as $C_{P1}$ for $p_i = 1$ atm, 
and about three times for $p_i = 100$ atm., while the variation of $C_{P1}$ remains 
of the order of 12% (see Fig. 1 and 2).

The difference between the isentropic exponent ($\gamma$) and the 
equilibrium specific ratio ($\gamma^*$) is as large as 10% for a stoichiometric mix-
ture of hydrogen-oxygen and an initial pressure of one atmosphere. This 
difference decreases rapidly with increasing dilution. Although the 
difference between $C_p$ and $C_{P1}$ is found to be a maximum for the stoichio-
metric mixture, the values of the specific heat ratios $\gamma^*$ and $\gamma$ are 
rather close in the neighborhood of $m = n = 0$. In fact the difference $|\gamma^* - \gamma|$ 
reaches a maximum for a value of the dilution index depending on the type of 
diluting gas and on the initial pressure. For instance, for hydrogen dilution 
and $p_i = 100$ atm., this maximum is close to $m = 2$. The isentropic exponent 
and the specific heat ratios are presented graphically in Fig. 3 and 4 for 
helium and hydrogen dilutions respectively.

The various expressions for the speed of sound are represented 
in Fig. 5 for helium dilution and in Fig. 6 for hydrogen dilution. According 
to the definitions (51), (52) and (53), what has been said for the $\chi$'s can be 
repeated for the sound speeds, (see Fig. 5 and 6).

5. CONCLUSIONS

The equilibrium specific heat ratio and the isentropic exponent 
have been computed for reaching gas mixtures composed of stoichiometric 
hydrogen-oxygen diluted with helium or hydrogen. The values of these 
quantities have been compared with the frozen specific heat ratio for initial 
pressures ranging from 1 to 500 atm. and diluting index ranging from 
1 to 7.5 in the case of hydrogen dilution, and from 1 to 12 in the case of 
helium dilution. In each calculation the initial temperature was chosen equal 
to 298.15°K, but the computer program does not include any such restriction.

Differences of the order of ten percent, were found between 
the isentropic exponent and the equilibrium specific heat ratio for initial 
pressure as high as one atmosphere. In both cases of helium and hydrogen 
dilutions, this difference was a maximum for the stoichiometric hydrogen-
oxygen mixture.

The difference between the equilibrium and frozen specific 
heat ratios, was a maximum for a dilution depending on the diluting gas and 
the initial pressure.
In the case of helium dilution, and $p_i = 1\text{ atm}$, this difference reaches about 15% for a dilution index $n$ of the order of 5.5 (approximately 65% of helium per volume in the initial mixture). The different values of the speed of sound evaluated using the isentropic exponent, the equilibrium and frozen specific heat ratios have also been computed and compared.
REFERENCES


LIST OF TABLES

1. Helium dilution, \( m = 0 \)
   
   \[
   T_i = 298.15^\circ\text{K}
   \]
   
   \[
   p_i = 1. ; 5. ; 10. ; 30. ; 50. ; 100. \text{ atm.}
   \]
   
   \[n = 0 \text{ to } 12 \text{ in steps of } 0.5\]

2. Hydrogen dilution, \( n = 0 \)

   \[
   T_i = 298.15^\circ\text{K}
   \]
   
   \[
   p_i = 1. ; 5. ; 10. ; 30. ; 50. ; 100. \text{ atm.}
   \]
   
   \[m = 0 \text{ to } 7.5 \text{ in steps of } 0.5\]

SYMBOLS USED IN TABLES 1 AND 2

Tables 1 and 2 are the direct outputs of the computer. The following symbols have been used:

- \( P_I \) : \( p_i \) (atm)
- \( T_I \) : \( T_i \) (\( ^\circ\text{K} \))
- \( N \) : \( n \)
- \( M \) : \( m \)
- \( C_P^I \) : \( C_{P_1} \) (cal mole\(^{-1}\))
- \( C_P \) : \( C_p \) (cal mole\(^{-1}\))
- \( \Gamma_1 \) : \( \gamma_1 \)
- \( \Gamma^* \) : \( \gamma^* \)
- \( \Gamma \) : \( \gamma \)
- \( A_1 \) : \( a_1 \) (m sec\(^{-1}\))
- \( A^* \) : \( a^* \) (m sec\(^{-1}\))
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**TABLE 1**

**TABLE 2**

**TABLE 3**
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\[ T_1 = 298.15 \, ^\circ\text{K} \]

\[ (2H_2 + O_2) + m \, H_2 \]

**FIG. 2**  
SPECIFIC HEAT VERSUS DILUTION (HYDROGEN DILUTION)
Initial conditions

\[ T_1 = 298.15 \, ^\circ\text{K} \]

\[
(2 \, \text{H}_2 + \text{O}_2) \rightarrow \text{n He}
\]
Initial conditions

\[ T_i = 298.15 \text{ K} \]

\[ (2H_2 + O_2) + m H_2 \]

**FIG. 4** SPECIFIC HEAT RATIOS AND ISENTROPIC EXPONENT (HYDROGEN DILUTION)
FIG. 5  SPEED OF SOUND VERSUS DILUTION (HELIUM DILUTION)

Initial conditions

* $T_i = 298.15 \, ^\circ C$
* $(2H_2 + O_2) + nHe$
Initial conditions

\[ T_1 = 298.15 \, ^\circ \text{K} \]

\[ \{ 2\text{H}_2 + \text{O}_2 \} + \text{m} \, \text{H}_2 \]
This note is complementary to UTIAS Technical Note No. 85, "Thermodynamic and Composition Data for Constant-Volume Combustion of Stoichiometric Mixtures of Hydrogen-Oxygen Diluted with Helium or Hydrogen", by A. Benoit. It includes the calculation of the equilibrium specific heats, the equilibrium specific heat ratios, the isentropic exponents, and the corresponding values of the speeds of sound. For convenience, the final-to-initial temperature ratio and the final-to-initial pressure ratio have also been included in the present tables. The results are presented for helium and hydrogen dilution respectively.
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