Magnetocombustion: A Thermodynamic Analysis

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The impact of a uniform magnetic field on equilibrium combustion characteristics has been explored. An expression for the Gibbs free energy, which includes a magnetic field contribution, has been developed. Using the method of Lagrange multipliers, changes in the Gibbs free energy for a mixture of paramagnetic and diamagnetic ideal gases are minimized. A model reaction of methane in air is used to quantitatively examine the changes in equilibrium compositions in the presence of a uniform magnetic field. Plots are presented showing the equilibrium mole fractions as a function of temperature and magnetic induction for all product species. In general, the results indicate that within certain temperature ranges a magnetic field decreases the mole fraction of major product species and increases the mole fraction of minor product species. The maximum equilibrium mole fraction of NO was observed to decrease an order of magnitude for an increase in magnetic induction of 0.00–0.04 T. The impact of a magnetic field was seen to affect equilibrium combustion characteristics of NO at temperatures well above those existing in practical combustion applications.

Nomenclature

a_{ij} = atoms of element j in product i
B = magnetic induction
b_j = atoms of element j in reactants
C = damping constant
C_W = Curie–Weiss constant
E = iteration error
G = Gibbs free energy
$\bar{g}^0$ = molar specific reference Gibbs free energy
H = magnetic field strength
I = enthalpy
M = intensity of magnetization
n = number of moles
n_e = number of constituent elements
n_s = number of product species
n_T = total number of moles
p = pressure
p_i = partial pressure of species i
R_u = universal gas constant
S = entropy
T = temperature
U = internal energy
V = volume
W = work
y_i = mole fraction of species i
θ = Curie–Weiss constant
λ_i = Lagrange multiplier of species i
μ_0 = permeability of free space
χ = magnetic susceptibility

Subscript

i = species i

Superscript

0 = reference conditions

I. Introduction

SINCE the time of Faraday,[1] the impact of magnetic fields on combustion behavior has been recognized. This interaction has primarily been attributed to the diamagnetic and paramagnetic nature of the gases involved in the combustion process. Diamagnetic behavior is observed in gases consisting of atoms with no permanent magnetic dipole moments. In the presence of an external magnetic field, the atoms of a diamagnetic substance develop a net dipole moment. This induced moment opposes the applied field, and thus a diamagnetic gas exhibits a weak repulsion to an applied magnetic field. The stronger the external magnetic field is, the stronger the repulsion. On the other hand, a paramagnetic gas is a gas consisting of atoms with at least one unpaired electron, and thus the atoms exhibit permanent dipole moments. In the absence of a magnetic field, the magnetic dipole moments of a paramagnetic substance are randomly oriented and exhibit only a weak interaction. When a magnetic field is applied, the dipole moments of the individual atoms align with the magnetic field producing a mild attraction to the magnetic field. The strength of this attraction is proportional to the strength of the externally imposed magnetic field. In a paramagnetic gas the magnetic behavior associated with the dipole moments must compete with the randomizing effect of temperature. The experimentally developed Curie–Weiss law describes this behavior. Noting that the magnetic susceptibility is defined as the ratio of the magnetization vector to the magnetic field strength, the Curie–Weiss law is given as

$$\chi = C_W/(T - \theta) \qquad (1)$$

On the other hand, diamagnetic susceptibility is independent of temperature.[2] Note that for a paramagnetic substance $\chi > 0$, and for a diamagnetic substance $\chi < 0$.

Early work involving chemical reactions and magnetic fields predicted that prohibitively large magnetic field strengths would be required to product a significant interaction between the two.[3−5] These conclusions were based upon order-of-magnitude estimates and did not account for the highly nonlinear nature of chemical reactions. Recent work, involving both permanent and electromagnets, has shown that magnetic fields can significantly affect combustion characteristics. Hayashi found that in oxygen-propane and oxygen-hydrogen combustion the presence of a nonuniform magnetic field increased the emission intensity of the OH radical.[6] Ueno and Harada[7] found that the application of a magnetic field with a magnetic induction of 0.5–1.4 T, with magnetic field gradients on

the order of 20–200 T/m, lowered the combustion temperature of alcohol in the presence of a platinum catalyst by 100–200°C.

Aoki found that the presence of a nonuniform magnetic field produced an increase in emission intensity for OH, CH, and C_2 as well as a decrease in soot production for diffusion flames.[8] Later, Aoki reported a decrease in the emission intensity of OH, CH, and C_2 for a configuration that possessed both uniform and nonuniform magnetic field regions.[9] This decrease was attributed to the arrangement used to produce the uniform field. Wakayama found that while magnetic field gradients had little effect on premixed flames an increasing field gradient decreased the combustion rate in diffusion flames.[10] A later investigation by Wakayama and coworkers found that the application of a magnetic field decreased the formation of soot in diffusion flames.[11] Additionally, the interaction of the nonuniform magnetic field with the paramagnetic and diamagnetic behavior of the constituent gases was found to induce convective transport that would otherwise not be present in their microgravity experiments.

The motivation for this investigation is the recent appearance of commercially available devices that claim to increase combustion efficiency significantly through the use of magnetic fields. Given that one of the primary sources of atmospheric pollution is the combustion of fossil fuels, such devices hold great promise from both a financial and environmental perspective. In this paper a thermodynamic analysis has been used to examine the impact of a uniform magnetic field on equilibrium combustion characteristics. An expression for the Gibbs free energy of a mixture of ideal gases is presented in Sec. II. The technique used to obtain the equilibrium composition is outlined in Sec. III. In Sec. IV, results are presented, and the significance of these results is discussed. The paper is summarized and conclusions are given in Sec. V.

II. Formulation of Gibbs Free Energy

Consider a closed, homogeneous, isotropic, stationary system subjected to a constant uniform magnetic field. For a reversible process the first law of thermodynamics states that

$$T\,dS + \delta W = dU \tag{2}$$

The work contribution, including both boundary and magnetic field work, may be written as[12]

$$\delta W = -p\,dV + d\left(V\int H\,dB\right) \tag{3}$$

Note that Eq. (3) has been modified compared to that appearing in Rosenweig to account for a difference in the sign convention. Expanding Eq. (3) gives

$$\delta W = -p\,dV + H^2\mu_0\chi\,dV + VH\mu_0\chi\,dH + VH^2\mu_0\,d\chi \tag{4}$$

where it has been assumed that the system consists of either a paramagnetic or diamagnetic substance, i.e., $M = \chi H$. Also note that the relation $B = \mu_0(H + M)$ was used in the development of Eq. (4). Substituting Eq. (4) into the first law of thermodynamics, Eq. (2), and using the definition of enthalpy $I \equiv U + pV$ and Gibbs free energy $G \equiv I - TS$, the change in Gibbs free energy for the system just described may be written as

$$dG = V\,dp + H^2\mu_0\chi\,dV + VH^2\mu_0\,d\chi \tag{5}$$

The preceding equation has been simplified by noting that the magnitude of magnetic field is constant, i.e., $dH = 0$, and assuming that isothermal conditions exist, i.e., $dT = 0$. Recall that the system has been taken to be either a paramagnetic or diamagnetic substance. Assuming that the magnetic susceptibility obeys the Curie–Weiss law, Eq. (1), and is thus only a function of temperature, $d\chi = 0$ from the assumption of isothermal conditions. Given this, the change in the Gibbs free energy is described as

$$dG = V\,dp + H^2\mu_0\chi\,dV \tag{6}$$

Assuming that the system consists of an ideal gas, i.e., $pV = nR_uT$, Eq. (6) may be written as

$$d\left(\frac{G}{R_uT}\right) = n\left(\frac{dp}{p} - H^2\mu_0\chi\,\frac{dp}{p^2}\right) \tag{7}$$

where the Gibbs free energy was normalized to be consistent with the Chemkin thermodynamic database.[13] Integrating Eq. (7) from a reference state to a specified state yields (assuming that $p^0 = 1$ atm as)

$$G/R_uT = n\left[\bar{g}^0\big/R_uT + \ell n\,p + H^2\mu_0\chi(1/p - 1)\right] \tag{8}$$

The Gibbs free energy for a mixture of ideal gases is therefore

$$\frac{G}{R_uT} = \sum_{i=1}^{n_s} n_i\left(\frac{\bar{g}_i^0}{R_uT} + \ell n\,p_i + H^2\mu_0\chi_i\left(\frac{1}{p_i} - 1\right)\right) \tag{9}$$

For a mixture of ideal gases, $p_i = y_i p$, where the mole fraction is defined as $y_i = n_i/n_T$ and the total number of moles is simply

$$n_T = \sum_{i=1}^{n_s} n_i$$

Substituting these definitions into Eq. (9) gives

$$\frac{G}{R_uT} = \sum_{i=1}^{n_s} n_i\left(\frac{\bar{g}_i^0}{R_uT} + \ell n\,y_i + \ell n\,p + \frac{H^2\mu_0\chi_i}{y_iP} - H^2\mu_0\chi_i\right) \tag{10}$$

Equation (10) is an expression for the Gibbs free energy of a mixture of ideal paramagnetic and diamagnetic gases. Including the contribution of a uniform magnetic field results in the addition of two terms, one that is inversely proportional to the pressure. Also note that the sign of the last two terms in Eq. (10) will depend on whether the respective component is diamagnetic or paramagnetic.

III. Determination of Equilibrium Composition

Equilibrium combustion characteristics have primarily been computed using the equilibrium constant method[14] or by minimizing the changes in the Gibbs free energy.[15–18] In this paper equilibrium combustion characteristics have been determined by minimizing the Gibbs free energy. Recall that at a specified temperature and pressure equilibrium will be achieved for $(dG)_{T,P} = 0$. In addition to the requirement that the change in the Gibbs free energy be equal to zero, the conservation of mass must also be satisfied. The conservation of mass for each of the constituent elements involved in the chemical reaction is given as

$$\sum_{i=1}^{n_s} a_{ij}n_i - b_j = 0 \tag{11}$$

Note that Eq. (11) is a system of n_e equations. To establish the equilibrium composition for a given reaction at a specified temperature and pressure, one must minimize changes in Gibbs function subject to mass conservation as specified by Eq. (11).

To obtain the solution of the preceding system of equations, the method of Lagrange multipliers was used. Following the formulation outlined by Morley,[16] the following pseudoalgorithm was used to determine equilibrium compositions:

1) Initialize the number of moles for each of the product species to 0.1, calculate various constants, populate the mass conservation matrix and the magnetic susceptibility array, read in the thermochemical data, calculate magnetic field contribution, and evaluate the normalized reference Gibbs free energy for each of the product species.

2) Solve the following system of equations:

$$\sum_{i=1}^{n_s} M_i = \sum_{j=1}^{n_e} \lambda_j \sum_{i=1}^{n_s} \tilde{n}_i a_{ij} \tag{12}$$

$$\sum_{k=1}^{n_e} \lambda_k \sum_{i=1}^{n_s} a_{ij}a_{ik}\tilde{n}_i + \frac{n_T}{\tilde{n}_T}\sum_{i=1}^{n_s} a_{ij}\tilde{n}_i + \sum_{i=1}^{n_s}(-a_{ij}M_i) - b_j = 0 \tag{13}$$

where

$$M_i \equiv n_i \left[\frac{g_i^0}{R_u T} + \ell_n\left(\frac{\tilde{n}_i}{\tilde{n}_T}\right) + \ell_n\, p - H^2 \mu_0 \chi_i + \sum_{m=1}^{n_s} \frac{H^2 \mu_0 \chi_m}{p} \right] \tag{14}$$

Note that Eq. (13) provides n_e equations, and Eq. (12) provides a single equation. The unknowns for the system are $n_T/\tilde{n}_T$ and n_e values of λ_k. This system of equations is solved using standard Gaussian elimination with pivoting.[19]

3) Calculate a new temporary estimate of the number of moles of each component using

$$\tilde{n}_i^{m+1} = \tilde{n}_i^m \left(\frac{n_T}{\tilde{n}_T} + \sum_{j=1}^{n_e} \lambda_j a_{ij} \right) - M_i \tag{15}$$

The new estimate of the equilibrium number of moles is calculated using

$$\tilde{n}_i^{m+1} = \tilde{n}_i^{m+1} + C\left(\tilde{n}_i^{m+1} - \tilde{n}_i^m\right) \tag{16}$$

where C, the damping constant, is adjusted to ensure convergence.
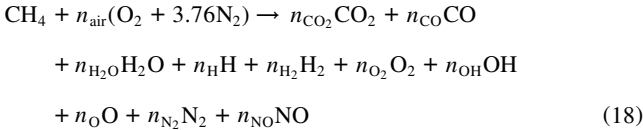
4) Calculate the normalized max norm of the solution

$$E = \max_i \frac{\left| n_i^{m+1} - n_i^m \right|}{n_i^{m+1}} \tag{17}$$

If $E < 10^{-15}$, the solution is taken to be converged, the results are written to a file, and the program stops. If $E \geq 10^{-15}$, then $n_i^m = n_i^{m+1}$, and the program proceeds to step 2.

IV. Results and Discussion

The equilibrium composition of a model reaction was used to investigate the impact of a magnetic field on combustion characteristics. The specific model reaction considered in this investigation is the combustion of methane in air, i.e.,

$$CH_4 + n_{air}(O_2 + 3.76N_2) \rightarrow n_{CO_2}CO_2 + n_{CO}CO$$

$$+ n_{H_2O}H_2O + n_H H + n_{H_2}H_2 + n_{O_2}O_2 + n_{OH}OH$$

$$+ n_O O + n_{N_2}N_2 + n_{NO}NO \tag{18}$$

The equilibrium composition model developed in the preceding section was validated by comparison with a GASEQ.[16] GASEQ is a Windows-based program for calculating equilibrium composition using the same technique used to generate the results presented in this paper, i.e., the minimization of Gibbs free energy. Results from the equilibrium model and from GASEQ were compared for temperatures ranging from 500 to 5000 K, pressure ratios ranging from 1 to 100, and equivalence ratios ranging from 0.1 to 1. Setting the magnetic induction to zero produced results that matched exactly (to within the number of significant digits in the output) to the results from GASEQ for the same conditions.

All results reported here have assumed an equivalence ratio of unity. Values for the magnetic susceptibilities for elements/compounds were obtained from the *CRC Handbook of Chemistry and Physics*.[20] Magnetic susceptibility values for ions were obtained from Mulay.[21] Recall that in the introduction section of this paper the statement was made that the magnetic susceptibilities of paramagnetic materials are a function of temperature. This temperature dependence was not included in the values of magnetic susceptibility used in the equilibrium model because of the scarcity of data on the conditions for which reported values were obtained. The range of magnetic induction examined in this paper is 0.00–0.04 T. The decision to examine this range was an ad hoc decision based solely on the fact that the impact of a magnetic field on equilibrium characteristics was easily observable in this range.

Figure 1 is a plot of the equilibrium mole fraction for carbon dioxide. From the figure one can see that in a certain temperature range
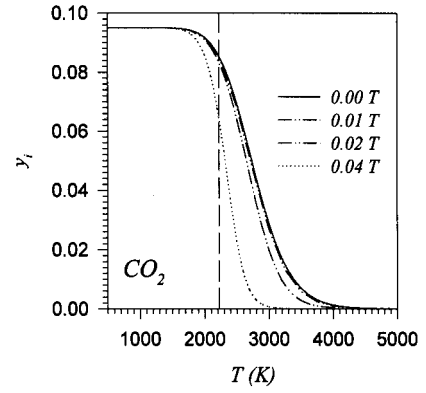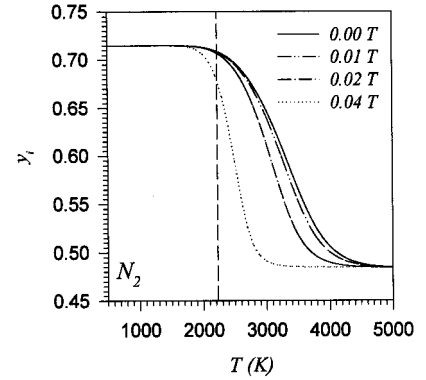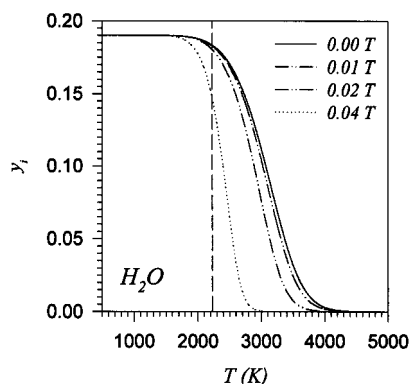

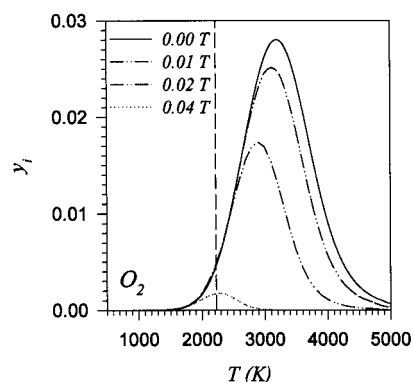**Fig. 1   Equilibrium mole fraction of CO_2.**

Fig. 2   Equilibrium mole fraction of N_2.

there is a nonlinear decrease in the mole fraction of CO_2 for increasing magnetic induction at a specified temperature. The influence of a magnetic field on the mole fraction becomes significant above a temperature of approximately 1500 K. For all cases the mole fraction was observed to decrease to a negligible level as the temperature increased beyond a certain value. The mole fraction dropped below 0.002 at a temperature of slightly less than 3900 K when no magnetic field was applied. This compares to the mole fraction dropping below 0.002 at a temperature near 2850 K when a 0.04 T magnetic field was applied, i.e., a drop of nearly 1000 K for an increase of 0.04 T. For stoichiometric conditions with no applied magnetic field, the adiabatic flame temperature for methane-air combustion[14] is 2226 K. On all of the plots presented in this paper, the adiabatic flame temperature for a stoichiometric mixture of methane and air without an applied magnetic field is represented as a vertical broken line. Of course, because the magnetic field affects the equilibrium composition, it will also affect the adiabatic flame temperature. At this point in time, no conclusions are drawn with regard to how a magnetic field will affect the adiabatic flame temperature. Considering the equilibrium composition in the vicinity of this temperature, one can see that there is a significant decrease in the mole fraction of CO_2 because of the application of the magnetic field.

Figure 2 is a plot of the equilibrium mole fraction for diatomic nitrogen. As with CO_2, the application of a magnetic field decreased the mole fraction of N_2 for a given temperature in a certain range of temperatures. Magnetic field effects became significant for temperatures above approximately 1700 K. As the temperature was increased, an asymptotic value of 0.485 was observed for the mole fraction in all cases. The temperature at which this asymptotic value was reached decreased with increasing magnetic induction. For no magnetic field the asymptotic value was observed for temperatures above 4800 K. This value decreased to 3400 K for a magnetic field of 0.04 T. As with CO_2, at the nonmagnetic stoichiometric adiabatic flame temperature the presence of a magnetic field produced a significant drop in the predicted mole fraction of N_2.
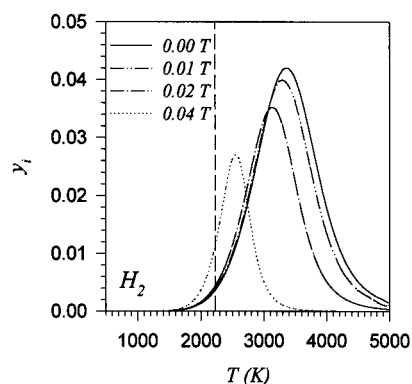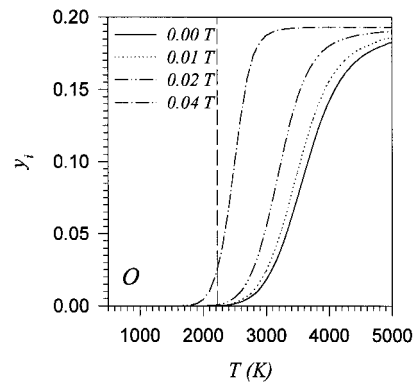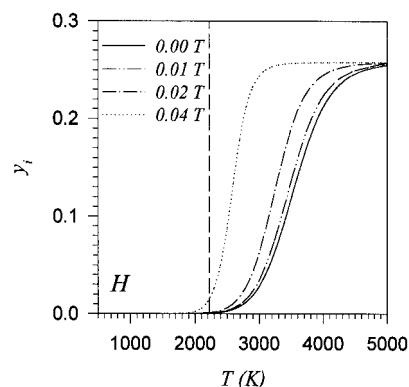
Figure 3 is a plot of the equilibrium mole fraction for water. As in the preceding two figures, an increase in the magnetic field strength

Fig. 3   Equilibrium mole fraction of H₂O.

Fig. 4   Equilibrium mole fraction of O₂.

Fig. 5   Equilibrium mole fraction of H₂.

Fig. 6   Equilibrium mole fraction of O.

Fig. 7   Equilibrium mole fraction of H.

decreased the mole fraction of water in a certain range of temperatures. Magnetic field effects became significant above a temperature of 1600 K. Above this temperature the mole fraction of water decreased to negligible levels as the temperature increased. For the case of no applied magnetic field, the mole fraction dropped below 0.005 at a temperature around 3900 K. This compares to a temperature of approximately 2800 K for the case of a magnetic field strength of 0.04 T. As with CO_2 and N_2, at the nonmagnetic stoichiometric adiabatic flame temperature the presence of a magnetic field produced a significant drop in the predicted mole fraction of H_2O.

Figure 4 is a plot of the equilibrium mole fraction for diatomic oxygen. As can be seen from the figure, as the temperature increases the mole fraction of O_2 increases to a maximum and then steadily decreases. A maximum value of 0.028 was observed at a temperature of approximately 3200 K when no magnetic field was applied. This compares with a maximum of 0.0017 at a temperature of near 2500 K when a magnetic field of 0.04 T was applied. The general trend is that there is a decrease in the maximum mole fraction with an increase in the magnetic field strength and that the temperature at which this maximum occurs decreases with increasing magnetic induction. At a temperature of 2226 K, the plot shows that for a magnetic induction of 0.04 T there is a significant drop in the mole fraction, but for the other values of the magnetic induction considered the change was negligible.

Figure 5 is a plot of the equilibrium mole fraction for diatomic hydrogen. Diatomic hydrogen exhibited the same general behavior as diatomic oxygen. As the temperature increased, H_2 increased to a maximum and then steadily decreased. The maximum mole fraction decreased with increasing magnetic field strength, and the temperature at which this maximum occurred also decreased with increasing magnetic field strength. A magnetic field decreased the temperature where the maximum mole fraction occurred. This behavior was somewhat more pronounced for H_2 than for O_2. There was nearly an order-of-magnitude decrease in the maximum mole fraction of O_2 for an increase in magnetic field strength of 0.00–0.04 T. For H_2 the maximum value decreased only about 43% for

a similar increase in magnetic field strength. At a temperature of 2226 K (the adiabatic flame temperature without an applied magnetic field), the plot shows that an increasing magnetic induction increases the mole fraction of H_2.

Figure 6 is a plot of the equilibrium mole fraction for monatomic oxygen. Increases in the magnetic field strength decreased the temperature where the equilibrium composition behavior began to differ from the behavior observed in the absence of a magnetic field. In general, there was an increase in the mole fraction of monatomic oxygen as a function of magnetic field strength at a specified temperature above the temperature where magnetic field effects became significant. An asymptotic value of approximately 0.1925 was observed when a magnetic field of 0.04 T was applied. Whereas none of the other cases reached this value in the temperature range, the curves do indicate that the other cases may be approaching this value. At a temperature of 2226 K, the plot shows that an increasing magnetic induction increases the mole fraction of monatomic oxygen.

Figure 7 is a plot of the equilibrium mole fraction for monatomic hydrogen as a function to temperature. As for monatomic oxygen,

there was an increase in the mole fraction of H at a specified temperature for an increase in the magnetic field strength within a given temperature range. The temperature where magnetic field effects became significant was observed to decrease with increasing magnetic field. An asymptotic value of approximately 0.2575 was observed for the largest applied magnetic field, i.e., 0.04 T. The behavior of the other cases indicated that this asymptotic value would hold for all of the cases. Because the maximum temperature considered was 5000 K, the asymptotic value was not achieved by the other cases. As with the other minor product species, an increasing magnetic field increased the mole fraction of monatomic hydrogen at the stoichiometric adiabatic flame temperature with no magnetic field.

Figure 8 is a plot of the equilibrium mole fraction for the hydroxyl radical as a function of temperature. As for O_2 and H_2, there is a decrease in the temperature where the maximum value of the mole fraction for OH occurs. Although the general behavior of OH is similar to that observed for O_2 and H_2, i.e., there is an increase in OH to a maximum value and then a steady decrease as the temperature increases, there is a significant difference. The maximum value of the mole fraction of OH initially increases with increasing magnetic field. As the magnetic field increases beyond a certain value, the maximum value of the mole fraction of OH then begins to decrease. As can be seen from Fig. 8, the maximum value of the mole fraction for OH is approximately the same for no applied magnetic field and for a magnetic field of 0.04 T. At a temperature of 2226 K, the plot shows that an increasing magnetic induction increases the mole fraction of the OH radical.

Figure 9 is a plot of the equilibrium mole fraction for carbon monoxide as a function of temperature. As can be seen from the plots, the general trend above approximately 1550 K is for the mole fraction to increase to a maximum and then decrease to a nonzero asymptotic value. This asymptotic value of the mole fraction is approximately 0.065. The magnetic field has the effect of decreasing the peak in mole fraction. At a magnetic field strength of 0.04 T, there is no significant peak in the mole fraction of CO. At a temperature of approximately 2800 K, for an applied magnetic field of 0.04 T CO achieves the asymptotic value of mole fraction. In
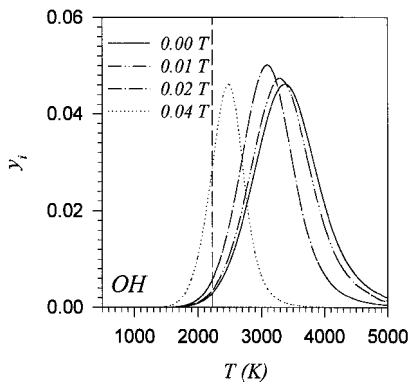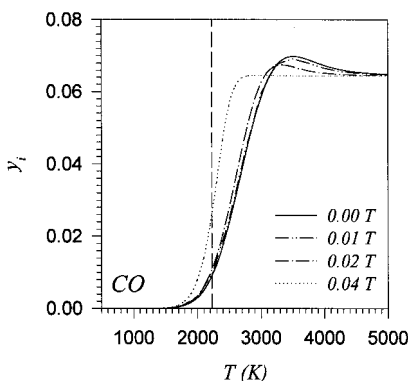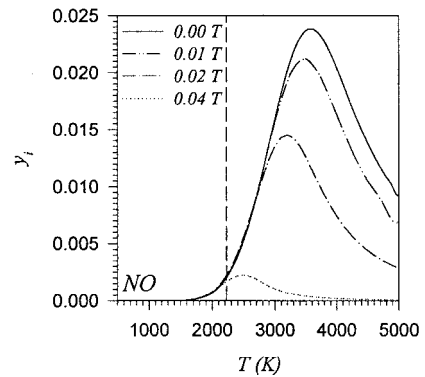

Fig. 10   Equilibrium mole fraction of NO.

the region where the peak in the mole fraction is distinguishable, increasing the magnetic field has the effect of decreasing the temperature where this peak occurs. In the region where the mole fraction of CO is increasing to the asymptotic value, the trend is that there is an increase in the mole fraction at a specified temperature as the magnetic field strength increases. Again, the plot shows that at 2226 K, an increasing magnetic induction increases the predicted mole fraction of CO.

Figure 10 is a plot of the equilibrium mole fraction for nitric oxide. Above a temperature of approximately 1500 K, NO was observed to increase to a maximum and then steadily decrease. As with O and H, the magnitude of the maximum mole fraction observed for NO decreased with increasing magnetic field strength. In fact, from an increase of 0–0.04 T, the maximum mole fraction of NO was observed to decrease nearly an order of magnitude. As had been the case with other species, an increase in the magnetic field strength decreased the temperature at which the maximum mole fraction occurred. At a temperature of 2226 K, the plot indicates that there is a relatively small change in the mole fraction of NO compared to the changes observed for larger temperatures. For a magnet induction of 0.01 and 0.02 T, the presence of a magnetic field actually increased the mole fraction slightly.

V.   Conclusions

Results of a thermodynamic investigation of equilibrium combustion characteristics in the presence of a uniform magnetic field have been presented. An expression for the Gibbs free energy was developed. The method of Lagrange multipliers was used to minimize the changes in the Gibbs free energy while ensuring that the conservation of mass was satisfied. Results, in the form of plots of the equilibrium mole fraction as a function of temperature and magnetic induction, were presented. Based upon the information gained from these plots, the following qualitative conclusions may be drawn:

1) When they are of sufficient strength, magnetic fields can have a significant impact on equilibrium combustion characteristics.

2) At the stoichiometric adiabatic flame temperature for no applied magnetic field, the application of a magnetic field had the effect of decreasing the mole fraction of the major product species.

3) In general, at the stoichiometric adiabatic flame temperature for no applied magnetic field, the application of a magnetic field had the effect of increasing the mole fraction of the minor product species.

4) At higher temperatures the application of a magnetic field greatly decreased the equilibrium composition of NO. Given that the predicted values for NO are likely to overpredict the actual values (because of the speed of the formation reaction), application of a magnetic field may be useful in decreasing NO emissions. At 2226 K for all conditions but the largest magnetic induction value, application of a magnetic field slightly increased the mole fraction of NO. The impact of a uniform magnetic field primarily affects NO equilibrium compositions at temperatures well above those occurring in practical engineering applications, however. Additional research is needed to predict the impact of magnetic fields on NO production for practical engineering applications.

Fig. 8   Equilibrium mole fraction of OH.

Fig. 9   Equilibrium mole fraction of CO.

Acknowledgments

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