# The Effects of Non-Uniform Magnetic Field on the Concentration of Methane-Air Reaction Species

#### Ali. Saeedi\*, Javad. Khadem, and Hojjat. Raznahan

Department of Mechanical Engineering, University of Birjand, Birjand 9717434765, Iran

(Received 29 September 2019, Received in final form 24 March 2020, Accepted 24 March 2020)

It is a well-known fact that the effects of magnetic fields on combustion can be used to control and optimize the flame deformation and the flame brightness. The kinetics and equilibrium properties of chemical reactions of combustion are influenced by the magnetic force exerted on paramagnetic species. In this study, the effects of non-uniform magnetic fields on one-stage methane combustion reaction are numerically investigated. It is known that NO, OH, and  $O_2$  are paramagnetic species and the other species and methane have diamagnetic behavior. Considering these facts, the effects of non-uniform magnetic field on 10 main product species of methane combustion are studied, by minimization of the Gibbs free energy. The results indicate that variation of non-uniform magnetic fields from 0 to 0.08 Tesla leads to decrease in NO mole fraction by 99.6 % in temperature range 1500-2500 K. Furthermore, the combination of non-uniform magnetic field and raising the pressure have the beneficial result in decreasing NO and CO mole fractions as well as rise in temperature.

Keywords : methane-air reaction, non-uniform, magnetic field, pressure, species

## 1. Introduction

It is well-known that a magnetic field affects flame [1]. The application of a magnetic field serves as a means of controlling combustion behavior [2]. Magnetic field is known to influence combustion in three ways:

Through the effects a magnetic field has on ionized species by Lorentz force.

Through the effects a magnetic field has on paramagnetic and diamagnetic species.

Through the effects a magnetic field has on equilibrium thermodynamics and chemical kinetics of combustion.

There are three different types of magnetism: diamagnetism, paramagnetism, and ferromagnetism. Diamagnetic substances are repelled by both poles of a magnet. In paramagnetic materials, such as oxygen, the magnetic dipole moments of the material get lined up with the external magnetic field to produce a net magnetic dipole moment and are slightly attracted to the magnets.

It has been demonstrated that the effect of magnetic field on combustion ionized species is negligible. Aoki [3] tested and showed the infinitesimal effect of the

```
©The Korean Magnetics Society. All rights reserved.
*Corresponding author: Tel: +98-56-32202301
Fax: +98-56-32202517, e-mail: Ali.saeedi@birjand.ac.ir
```

Lorentz force on the combustion characteristics.

Motivated by the possibility of utilizing magnetic body force exerted on paramagnetic and diamagnetic gases of combustion, most of the research studies have been conducted on the subject of the effects of magnetic fields on combustion control and pollutants emission of diffusion flames [4]. Wakayama studied the behavior of gas flow in air under gradient magnetic fields [5]. Oxygen or nitrogenmixed oxygen were observed to be attracted by a magnetic field. In contrast, nitrogen gas escaped from magnetic fields of high intensity. Wakayama suggested the possibility of controlling combustion in diffusion flames by means of a magnetic field. In another study, Wakayama [6] observed an increase of about 120 °C in flame temperature. The flame also became shorter and more brilliant. Gillon et al. [7, 8] observed that gradient magnetic field causes flame lift-off height to decrease. This observation is attributed to the magnetic force which develops through the air via its action on the paramagnetic oxygen molecules. Baker et al. [9] investigated the behavior of laminar jet diffusion flames in the presence of non-uniform magnetic fields. Their results indicated that, compared with flames to which no magnetic field is applied, the average flow rate related to the visible soot inception point decreased and the average flow rate associated with the near extinction point increased. Yamada et al. [10] numerically examined

the effect of a magnetic field on OH radical distribution in a hydrogen–oxygen diffusion flame. They noted the impact of the magnetic field gradient intensity on OH emission in the flame.

A few research studies have discussed the effects of magnetic field on the equilibrium thermodynamics and chemical kinetics of combustion. By considering the magnetic field work, the Gibbs free energy equation was modified for use in the presence of magnetic field. The modified equation can be minimized to determine the mole fraction of species [3]. Hayashi [11] studied the effects of magnetic fields on chemical reactions. Their results showed that a uniform magnetic field can increase the emission intensity of the OH radical, while no effect was observed on the CH and C2 radicals. Baker et al. [12] investigated the thermodynamic characteristics of the methane-air equilibrium combustion in the presence of a uniform magnetic field. Their results indicated that magnetic field has a significant impact on equilibrium combustion characteristics. Also, they noted that application of a magnetic field reduced the mole fraction of the major product species of the methane combustion. Shinoda et al. [13] studied PLIF measurement and numerical simulation of methane-air premixed jet flame in a magnetic field. They investigated the effect of magnetic field on OH density distribution. They noted that the magnetic force acting on  $O_2$  in the premixed gas injected from the burner influenced the OH density distribution in the premixed flame. Gupta et al. [14] studied the impact of a uniform magnetic field on equilibrium combustion characteristics under constant volume conditions. Their results showed that increase in the magnetic induction caused NO mole fraction to decrease. Also, the application of magnetic field decreased the equilibrium mole fraction of CO, H, O, H<sub>2</sub>, O<sub>2</sub>, OH and NO product species and increased that of CO<sub>2</sub>, N<sub>2</sub>, H<sub>2</sub>O, and CH<sub>4</sub>. In another study[15], they showed that the equilibrium flame temperature increases with increase in magnetic-field strength. Wu et al. [2] investigated the influence of magnetic fields on the temperature and pollutant properties of methane laminar combustion. Their results indicated that gradient magnetic field has a great effect on the production of thermal  $NO_X$ . They noted that the concentration of NO<sub>X</sub> decreased by, at least, up to 60 % on average. Barmina et al. [16] provided the effects of magnetic field on combustion dynamics in order to control the process of swirling flame. The magnetic field effect on the flame temperature and rate of reactions leads to increase in the flow vorticity. Jocher et al. [17] elucidated spontaneous flame instability that is mitigated as the non-premixed sooting flame experiences a magnetic perturbation. They show that the flow modification by a

magnetic gradient severely changes the non-premixed flame base structure and consequently soot production in the flame. Jocher *et al.* [18] unveiled a domain for different oxidizer mixtures composed of  $CO_2$ ,  $N_2$  and  $O_2$  which can control the stability of an ethylene non-premixed flame. Flame stabilization in the magnetic field was explained through enhanced soot formation and subsequent flame cooling in the flame.

The effects of non-uniform magnetic field on a combustion reaction is not a well-considered issue. Therefore, this paper is going through to this and considering the effects of a linear non-uniform magnetic field on methaneair reaction. Accordingly, in this study, thermodynamic analysis has been done to examine the effects of nonuniform magnetic field on equilibrium combustion characteristics. For this purpose, the mole fraction of the product species is calculated by the method of the Gibbs free energy under constant pressure condition. The present study focuses on emission reduction by making use of the effects of magnetic fields on equilibrium thermodynamics. By reviewing the previous relevant research, we acknowledge the considerable impact of pressure on combustion. Therefore, in this paper, the combination of the effects of magnetic field and pressure on the production of species will be investigated.

#### 2. Formulation

Consider a homogeneous, isotropic, stationary system that is subjected to a magnetic field. The change in the internal energy of the system is described as:

$$TdS + \delta W = dU \tag{1}$$

The work term may be written as:

$$\delta W = -p \, dV + d(V \int H \, dB) \tag{2}$$

The system consists of either a paramagnetic or diamagnetic substance, *i.e.*, M = x H. It is also notable that the relation  $B = \mu_0 H(1 + x)$  is used to further simplify Eq. (2). Therefore, the total work done on the system can be represented by either:

In the case of a uniform magnetic field:

$$\delta w = -pdV + H^2 \mu_0 x dV + V H \mu_0 x dH + V H^2 \mu_0 dx \quad (3)$$

Or

In the case of a non-uniform magnetic field:

$$\delta w = -pdV + \mu_0 dV (\int Hx dH + H^2 dx) + VH\mu_0 x dH + VH^2 \mu_0 dx$$
(4)

By substituting Eq. (3 or 4) into the first law of thermo-

dynamics, Eq. (2), and using the Gibbs free energy equation (G = I + TS) along with the definition of enthalpy (I = U + PV), we can obtain the Gibbs free energy of the system. Eq. (3 or 4) is simplified by considering the fact that the system is isothermal (dT = 0), and the magnetic susceptibility, which obeys the Curie-Weiss law, is only a function of temperature, which means dx = 0. Given this, the change in the Gibbs free energy is obtained for:

A Uniform magnetic field:

$$dG = Vdp + H^2 \mu_0 x dV \tag{5}$$

Or

A non-uniform magnetic field:

$$dG = Vdp + H^2 \mu_0 x dV + HV x \mu_0 dH \tag{6}$$

Assuming that the mixture of gases only consists of ideal gases, the Gibbs free energy of a mixture of ideal paramagnetic and diamagnetic gases is given by:

In the case of a uniform magnetic field:

$$\frac{G}{R_u T} = \sum_{i=1}^{n_{Sp}} n_i \left( \frac{g_i^0}{R_u T} + \ln(y_i) + \ln(p) + H^2 \mu_0 x_i \left( \frac{1}{y_i p} - 1 \right) \right)$$
(7)

Or

In the case of a non-uniform magnetic field:

$$\frac{G}{R_u T} = \sum_{i=1}^{n_{Sp}} n_i \left( \frac{g_i^0}{R_u T} + \ln(y_i) + \ln(p) + H^2 \mu_0 x_i \left( \frac{1}{y_i p} - 1 \right) + \frac{\mu_0 x_i}{2y_i p} (H^2 - H_0^2) \right)$$
(8)

In this paper, the mole fractions of combustion product species are calculated by minimizing the Gibbs free energy. Therefore, the change in the Gibbs free energy must be hypothesized to be equal to zero. The conservation of mass of the constituent elements is defined as:

$$\sum_{i=1}^{n} a_{ij} x_i - b_j = 0$$
(9)

As mentioned above, to compute the mole fraction of the equilibrium composition, we must minimize variation of the Gibbs function in accordance to mass conservation as specified by Eq. (9). In order to solve equation (8), the following pseudo algorithm was used to determine equilibrium compositions [19]:

$$\sum_{i=1}^{n_{Sp}} h_i = \sum_{j=1}^{n_{EI}} \lambda_j \sum_{i=1}^{n_{Sp}} y_i a_{ij}$$
(10)

$$\sum_{i=1}^{n_{Sp}} (-a_{ij}h_i) + \frac{\sum x_i}{\sum y_i} \sum_{i=1}^{n_{Sp}} y_i a_{ij} + \sum_{j=1}^{n_{El}} \lambda_k \sum_{i=1}^{n_{Sp}} a_{ik} a_{ij} y_i - b_j = 0$$
(11)

Where

$$h_{i} = y_{i} \left[ \frac{g_{i}^{0}}{R_{u}T} + \ln\left(\frac{y_{i}}{y_{t}}\right) + \ln p - H^{2}\mu_{0}x_{i} + \sum_{m=1}^{n_{Sp}} \frac{H^{2}\mu_{0}x_{m}}{p} + \sum_{m=1}^{n_{Sp}} \frac{\mu_{0}x_{m}}{2p} (H^{2} - H_{0}^{2}) \right]$$
(12)

Eq. (10) gives a single equation while Eq. (11) gives  $n_{El}$  equations. This system of equations is solved using standard Gaussian elimination. A new estimate of the number of moles is calculated by:

$$y_i = -h_i + y_i \left( \frac{\sum x_i}{\sum y_i} + \sum_{j=1}^{n_{El}} \lambda_j a_{ij} \right)$$
(13)

## 3. Results and Discussion

Methane-air 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-air as follows:

$$CH_4 + n_{air} (O_2 + 3.76N_2)$$
  
 $\rightarrow n_{CO_2}CO_2 + n_{CO}CO + n_{H_2O}H_2O + n_HH + n_{H_2}H_2$   
 $+ n_{O_2}O_2 + n_{OH}OH + n_OO + n_{N_2}N_2 + n_{NO}NO$ 

An equivalence ratio of unity was assumed for all the reported results. Magnetic susceptibility was used to quantify paramagnetic and diamagnetic behavior in the present study. The magnetic susceptibilities to diamagnetic product species were obtained from the CRC Handbook of Chemistry and Physics [4], which provides the values listed in Table 1. Among the product species, NO (nitric oxide),  $O_2$  (oxygen), and OH (hydroxyl) are paramagnetic gases. In a paramagnetic gas, the magnetic behavior is reversed with the effect of temperature. The behavior can be determined by the Curie–Weiss law. The Curie–Weiss

 Table 1. Magnetic susceptibility of some species in methane combustion [4].

Species	x (cgs unit)
CO <sub>2</sub>	$-21 \times 10^{-6}$
CO	$-9.8 \times 10^{-6}$
$H_2O$	$-12.63 \times 10^{-6}$
Н	$-2.93  imes 10^{-6}$
$H_2$	$-3.99  imes 10^{-6}$
$N_2$	$-12 \times 10^{-6}$
OH	$+7.25 \times 10^{-6}$
NO	$+1.45 \times 10^{-3}$
$O_2$	$+3.43 \times 10^{-3}$

	T(K)	1600	2000	2200	2400
$\mathbf{B}=0\left(\mathbf{T}\right)$	Baker <i>et al.</i> [12]	0.095	0.0921	0.0864	0.0771
	Present work	0.0942	0.0918	0.0864	0.0763
	Error (%)	0.7	0.3	0.07	1
B = 0.04 (T)	Baker et al. [12]	0.0946	0.0864	0.0682	0.0398
	Present work	0.0945	0.0863	0.0722	0.0469
	Error (%)	0.1	0.2	5.9	17.6

Table 2. Comparison between the results of the present work and those of Baker *et al.* [12] for  $CO_2$ .

law is given as:

$$x_{i} = \frac{N_{A}g_{L}^{2}\mu_{B}^{2}S_{i}(S_{i}+1)\mu_{0}}{2kTM_{i}}$$
(14)

The value of Lande's g factor is 2.0 for all paramagnetic species. The value of S is taken as 1.0, 0.5, and 0.5 for O, NO, and OH, respectively. Based on this explanation, the modeling of methane combustion in the presence of a magnetic field is done. We studied the effects of the non-uniform magnetic field at different pressures on 10 methane combustion main product species. The non-uniform magnetic field considers value of 0 to 0.08 Tesla.

Table 2 is a comparison of the values of the equilibrium mole fraction of  $CO_2$  as reported by the present work against Baker *et al.* [12] As shown in Table 2, there is a good agreement between the two sets of results.

Figure 1 shows a plot of the equilibrium mole fraction of carbon dioxide as a function of temperature, the final and the initial magnetic field strengths. As illustrated, in a certain temperature range, with increase of the final magnetic field strength, a nonlinear decrease in the mole frac-



Fig. 1. Variations of  $CO_2$  mole fraction as a function of temperature at different values of applied magnetic fields.

tion of  $CO_2$  takes place. When the initial magnetic field strength is equal to zero, as the final magnetic strength value rises at a given temperature, the mole fraction of  $CO_2$  significantly decreases. However, by raising the initial values of the magnetic induction, while the final magnetic induction is equal to zero, we do not obtain any significant change in the  $CO_2$  mole fraction. The results of Fig. 1 show an important effect of increasing final magnetic field on  $CO_2$  mole fraction.

Figure 2 illustrates the  $CO_2$  mole fraction variations in the presence of the final magnetic fields of 0 T and 0.04 T at different pressures. For the two cases of 5 and 20 atmospheric pressures, the equilibrium mole fraction of  $CO_2$  experiences negligible variations as the final magnetic field strength increases. But for the pressure of one atmosphere, the final magnetic field significantly affects the mole fraction. Overall, Fig. 2 shows that the effect of a uniform magnetic field at different pressures is only detected at 1 atm and negligible at higher pressure.

Figure 3 demonstrates the equilibrium mole fraction of



**Fig. 2.** Variations of  $CO_2$  mole fraction as a function of temperature in the uniform magnetic fields of 0 and 0.04 Tesla at different pressures.



Fig. 3. Variations of CO mole fraction as a function of temperature at different applied magnetic fields.

carbon monoxide as a function of temperature, the final and the initial magnetic field strengths. The mole fraction of CO increases with growth of the final magnetic field strength within a certain temperature range but it decreases with growth of the initial magnetic field strength. As seen in Fig. 3, at a temperature above 2400 K, the equilibrium mole fraction of CO is observed to increase to a maximum and then decrease slightly. The maximum mole fraction of CO is observed at 3190 K for 0-T magnetic induction. The temperature at which the maximum value of CO



**Fig. 4.** Variations of CO mole fraction as a function of temperature in the uniform magnetic fields of 0 and 0.04 Tesla at different pressures.

mole fraction occurs tends to decrease with growth of the final magnetic induction. Variations of CO mole fraction illustrate a similar effect of the magnetic field gradient. A strong effect when the magnetic field is increasing from initial to final and weak effect for decreasing magnetic field.

Figure 4 includes a plot of the variations in the CO mole fraction for the final magnetic fields of 0 and 0.04 Tesla at different pressures. The mole fraction of CO increases with growth of the final magnetic induction in a certain temperature range at 1 atm pressure. However, it decreases with growth of the final magnetic induction at 5 and 20 atm. pressures. Uniform magnetic field effects are remarkable at 1 atm and slightly marked at 20 atm for the variations of CO mole fraction. However, it is important to specify that the increase in the mole fraction at 1 atm and decrease at 5 and 20 atm is observed for a given temperature.

Figure 5 shows the equilibrium mole fraction of water as a function of temperature, the final and the initial magnetic field strengths. Similar to Fig. 1, the mole fraction of water is observed to have a growth trend with increase in the final magnetic field strength. As the temperature rises, the effect of magnetic field becomes less noticeable. At lower values of the initial and final magnetic fields, the minimum value of  $H_2O$  equilibrium mole fraction shifts to a higher temperature.

Figure 6 depicts the variations in the  $H_2O$  mole fraction in the final magnetic fields of 0 and 0.04 Tesla at the pressures of 1, 5 and 20 atm. Regular behavior of  $H_2O$ mole fraction as a function of temperature was observed



**Fig. 5.** Variations of  $H_2O$  mole fraction as a function of temperature at different applied magnetic fields.



**Fig. 6.** Variations of  $H_2O$  mole fraction as a function of temperature in the uniform magnetic fields of 0 and 0.04 Tesla at different pressures.

in the temperature range considered. Magnetic field induction has a significant effect at temperatures above 1600 K. The mole fraction of water decreases with growth of the final magnetic field but increases with growth of the pressure in a certain range of temperatures. The pressure increase seems to increase the  $H_2O$  mole fraction for a given temperature and application of a uniform magnetic field of 0.04 T is shown to decrease these values.

Figure 7 shows the equilibrium mole fraction of nitro-



Fig. 7. Variations of NO mole fraction as a function of temperature at different applied magnetic fields.



**Fig. 8.** Variations of NO mole fraction as a function of temperature in the uniform magnetic fields of 0 and 0.04 Tesla at different pressures.

gen monoxide as a function of temperature, pressure, the final and the initial magnetic field strengths. Above the temperature of 1650 K, NO was observed to increase to a maximum and then decrease sharply. Similar to the case of CO, the magnitude of the maximum value of the NO mole fraction was observed to increase with growth of the initial magnetic induction and significantly decrease with growth of the final magnetic induction. The maximum value of NO mole fraction was observed at 3612 K for 0.08 T initial magnetic induction. It was observed that the mole fraction of NO shows an increasing trend with increase in the initial magnetic induction strength. Within the temperature range considered, the NO mole fraction reached the maximum value as a result of the variations in the initial magnetic field strength. The maximum NO mole fractions shifted to higher temperatures with growth of the initial magnetic field strength.

Figure 8 illustrates the equilibrium mole fraction of nitrogen monoxide as a function of temperature and uniform magnetic field strength. As it can be seen in Fig. 8, at temperatures greater than 1600 K, the equilibrium mole fraction of NO was observed to increase to a maximum and then decrease. At the temperature of 2587.5 K (the adiabatic temperature with no magnetic field applied), increase of the magnetic induction to 0.04T caused the NO mole fraction to decrease by about 90 %. The maximum value of NO mole fraction keeps increasing with increase in pressure and magnetic induction and also shifts to a higher temperature.

Species	Application the magnetic field to reactants	Application the magnetic field at reaction zone	Increase in the pressure	Combining increase in the pressure and application of magnetic field at reaction zone
СО	Negligible	Increase Max (+2175 %)	Decrease Max (-56.3 %)	Decrease
$CO_2$	Negligible	Decrease Max (-95.6 %)	Increase Max (+20.8 %)	Increase
H <sub>2</sub> O	Decrease Max (-92.2 %)	Decrease Max (-99.1 %)	Increase Max (+7.6 %)	Decrease
NO	Increase Max (+98.3 %)	Decrease Max (–99.6 %)	Decrease Max (-37.4 %)	Decrease

**Table 3.** Variation of mole fraction caused by increase in the pressure and application of the final magnetic field in temperature range of 1500-2500 K.

### 4. Conclusion

In this study, a fundamental understanding of the behavior of equilibrium combustion compositions under the influence of final and initial magnetic fields is achieved. Note that it is possible that the decrease or increase in the values of mole fraction are not the same as the corresponding changes in engineering applications, which is caused by the fact that the time duration of reaction is not considered. However, a fundamental understanding of the trends of the mole fractions under the impact of initial and final magnetic fields can be achieved by using an equilibrium composition analysis. A thermodynamic analysis has been done to examine the impact of initial and final magnetic fields on equilibrium combustion characteristics. The present study focuses on emission reduction by the effects of magnetic fields on equilibrium thermodynamics. By minimization of the Gibbs free energy, the effects of initial and final magnetic fields, at different pressures, on 10 main product species of methane combustion were studied. The results are shown in the Table 3 for 1500-2500 K. also, the main conclusions can be mentioned as follows:

Application of a non-uniform magnetic field to the reaction zone leads to reduction in NO mole fraction; at the considered situation, NO mole fraction has diminution about 99.6 %.

Raising the pressure also brings reduction in NO mole fraction; the calculated data reveal NO mole fraction decreases about 37.4 %.

CO mole fraction increases by non-uniform magnetic field and lessens by pressure increase.

The combination of non-uniform magnetic field and raising the pressure have the beneficial result in decreasing NO and CO mole fractions as well as rise in temperature.

The magnetic field effects are observed on equilibrium

characteristics at very high temperature well above the temperature met in practical situation.

## References

- M. Faraday, J. Edinburgh and Dublin Philosophical Magazine 31, 401 (1847).
- [2] W.-f. Wu, J. Qu, K. Zhang, W.-p. Chen, and B.-w. Li, Combust. Sci. Technol. 188, 472 (2016).
- [3] T. Aoki, Japanese J. Applied Physics 28, 776 (1989).
- [4] D. R. Lide, CRC Handbook of Chemistry and Physics Boca Raton, Fla.: CRC Press (2009).
- [5] N. I. Wakayama, J. Applied Physics 69, 2734 (1991).
- [6] N. I. Wakayama and M. Sugie, Physica B: Condensed Matter 216, 403 (1996).
- [7] P. Gillon, J. Blanchard, and V. Gilard, Russian J. Physical Chemistry B, Focus on Physics 4, 279 (2010).
- [8] V. Gilard, P. Gillon, J.-N. l. Blanchard, and B. Sarh, Combust. Sci. Technol. 180, 1920 (2008).
- [9] J. Baker and M. E. Calvert, Combustion and Flame 133, 345 (2003).
- [10] E. Yamada, M. Shinoda, H. Yamashita, and K. Kitagawa, Combustion and Flame 135, 365 (2003).
- [11] H. Hayashi, Chemical Physics Letters 87, 113 (1982).
- [12] J. Baker and K. Saito, J. Propulsion and Power 16, 263 (2000).
- [13] M. Shinoda, E. Yamada, T. Kajimoto, H. Yamashita, and K. Kitagawa, Proc. Combust. Inst. 30, 277 (2005).
- [14] A. Gupta and J. Baker, J. AIAA 43, 2438 (2005).
- [15] A. Gupta and J. Baker, J. Thermophys. Heat Trans. 21, 520 (2007).
- [16] I. Barmina, R. Valdmanis, M. Zake, H. Kalis, M. Marinaki, and U. Strautins, Latvian J. Physics and Technical Sciences 53, 36 (2016).
- [17] A. Jocher, H. Pitsch, T. Gomez, J. Bonnety, and G. Legros, Phys. Rev. E 95, 063113 (2017).
- [18] A. Jocher, J. Bonnety, T. Gomez, H. Pitsch, and G. Legros, Proc. Combust. Inst. 37, 5637 (2019).
- [19] S. Gordon and B. J. McBride, Nasa sp-273, (1976).