ELECTRIC FIELD CONTROL OF NO_x FORMATION IN THE FLAME CHANNEL FLOWS

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ABSTRACT

The present studies aimed to obtain clean fuel combustion and get detailed information about the processes that determine the electric field effect on NO_x formation in flame channel flows. The experimental studies demonstrate that the interaction between the radial electric field and the flame initiates the field-forced drift motion of positive radical ions in a field direction. The energy exchange between ions and gas particles produces interrelated heat and mass transfer to the negatively biased channel walls. Hence, by varying the applied voltage, a field-enhanced reducing of a flame temperature is obtained, thereby lowering thermal NOx formation during the fuel combustion up to 30-80%.

KEY WORDS: electric field, heat/mass transfer, flame quenching, NOx formation.

INTRODUCTION

During combustion, common fuels like natural gas, mineral oil, wood chips, etc. are burned with air to produce heat energy. Nitrogen in a high-temperature reaction zone of a flame flow tends to react with oxygen forming NO and NO_2. A significant amount of NO_x emissions ($\sim 50\%$) is attributed to industrial boilers. About 20,400 tons of NO_x (including NO and NO_2) per year are produced by public power generation, cogeneration and district heating boiler houses in Latvia. Within Europe, emissions of air pollutants (NO_x, SO_2) vary greatly, depending upon many factors such as the degree of industrialization, pollution control practices, road transport intensity and policies on environmental issues. In fact, emissions of air pollutants are harmful to humans because they initiate the production of ozone and acid rain. Therefore, there is a great need for effective reduction of polluting emission levels.

In industrial boilers, NO_x can be formed in two ways- thermal and fuel NO_x. Thermal NO_x is formed during the combustion of gases and light oils at the high temperatures in a flame, when oxygen dissociates and atomic oxygen then reacts with atmospheric nitrogen. The reaction between hydrocarbon radicals and nitrogen contained in the fuel generate fuel NO_x. Fuel NO_x levels are high in coal and mineral oil combustion.

Methods of NO_x control in pollutants are subdivided into post-combustion and combustion control techniques. Clean combustion control techniques include methods of low excess air firing, combustion of low nitrogen fuel oil, different

modifications of burner construction, formation of flue gas recirculation system and the simplest method of reducing NO_x by water injection into the combustion zone of a flame. The alternative low-NO_x combustion technique is based on electric field enhanced processes of heat/mass transfer from flame flow. The electric field effects on the flame flows have received much attention (Payne and Weinberg, 1959; Lawton and Weinberg, 1969) to control the processes of heat/mass transfer and fuel combustion. These studies illustrate some typical features of the electric field-induced processes in the flame flows, which can be used as means of managing and improving some combustion processes. The experimental investigations of the propane combustion demonstrate that the interaction between the electric field and propane flame enhances soot formation in the flame (Vatashin *et al.*, 1995). This increases radiation heat losses from a flame, reduces a flame temperature and levels of NO_x emissions. The investigations in the free propane-air flame flow (Zake, 1995; Zake *et al.*, 1998) demonstrate the effect of the electric field-enhanced heat and mass transfer on the flame composition and temperature profiles. These investigations show that field effect on the flame parameters is more effective under conditions, if the field generates the radial transfer of positive ions of hydrocarbons from the hot reaction zone of a flame outward. By varying the field direction in a flame, the field effects on the flame parameters are quite negligible.

The objective of current laboratory and industrial investigations is to provide combustion control in the flame channel flows using the radial electric field, which is applied between the positively biased central electrode and negatively biased water-cooled channel walls. The radial electric field-enhanced processes of heat and mass transfer to the channel walls are studied with respect to flame composition, flow rate and field strength with the aim to reduce the rate of fuel combustion, peak temperature of the flame channel flows and the levels of the thermal NO_x emissions.

EXPERIMENTAL

The laboratory investigations of the electric field effect on the processes of interrelated heat and mass transfer, flame temperature and composition of emissions are carried out along the water-cooled and steel grid channel flows. The experimental set up includes a gas burner, central wire electrode, water-cooled or steel grid channels (diameter 4.5 cm, length 7.5 cm) and gas feeding system. The wire electrode is axially directed along a cylindrical burner and inserted into a flame channel flow at the distance L=5 mm from the exit of a burner. The electric field is applied between the supporting channel walls and positively biased central electrode. The schematic draw of the experimental set up with water-cooled channel is shown in Fig. 1.

The field-enhanced heat transfer to the channel walls is estimated from the calorimetric measurements of the cooling water flow. The electric field effects on the processes of mass transfer in the flame channel flows are estimated from the measurements of an ion current collected by the channel walls. In addition, in-flame measurements of the electric field effects on the radial and axial distributions of a flame temperature and composition are performed. The field effect on the radial and axial temperature distributions is studied using a Pt/Pt-Rh 13% thermocouple, which can be moved along and across the premixed flame flow. Local investigation of the absorption and radiation spectra of the flame compounds is performed to study the field effect on the flame composition profiles. To obtain the axial and radial distributions of the flame composition and relative mass fractions of the gas compounds, samples of the premixed gas are extracted from the flame flow by using micro-probe technique and then analyzed by absorption spectroscopy in a spectrum range of 2-15mm. The process of NO_x formation in a flame was controlled using the gas analyzer TESTO-3300.

The radial and axial variations of the temperature and composition inside the flame channel flow are studied while varying the applied voltage, velocity and composition of the burner exit flow. The experiments were carried out under the fuel-lean, stoichiometric and fuel-rich conditions of propane-air mixture by varying the equivalence ratio (α) in a range from 0.6 to 1.5. The rate of the propane-air exit flow is varied in a range of 1.0-2.1 m s^{-1}. A bias voltage of a central electrode is varied in a range of 0-(+3.0) kV with subsequent varying of an ion current to the supporting channel walls in a range of 0-3.10^{-4}A. The laboratory investigations were performed for propane-air
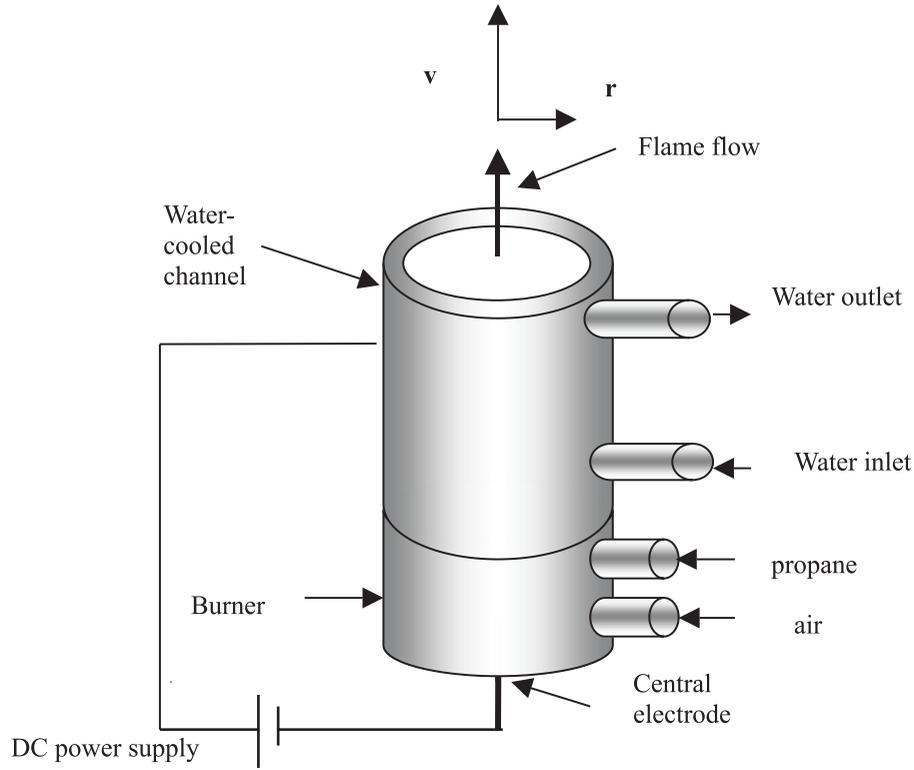
Figure 1. Schematic draw of the experimental set-up with water-cooled channel.

and natural gas flame flows.

Similar industrial investigations of the field effect on the fuel combustion are performed in the boilers of district heating boiler houses, in which natural gas or mineral oil are used as fuels.

RESULTS AND DISCUSSION

Preliminary investigations of the electric field effects on the free flame flow (Zake, 1995) demonstrate that the electric force acting on flame compounds enhances the drift motion of the positive radical ions ($C_2H_4^+$, $C_3H_3^+$, CHO^+) in a field direction. The elastic and inelastic collisions between the accelerated ions and gas compounds disturb the convective motion of a flame channel flow and initiate the field-forced motion of the gas compounds (ion wind) to the channel walls. This produces the local variations in the mass fraction of the flame compounds depending on the ion density, field strength, sheath thickness at the channel walls, flame flow rate and mechanism of a field-induced ion transfer to the channel walls. Under conditions of atmospheric pressure,

plasma and flame flows the dominant mechanism of the field-induced ion transfer to the channel walls can be estimated from appropriate values of $R_E \alpha^2 \chi^2$, where R_E, α^2 and χ^2 can be expressed as (Smy, 1976):

$$R_E = \frac{v_g L}{b_o (k T_{eo}/e)} \tag{1}$$

$$\alpha = (\frac{\varepsilon_o k T_{eo}}{n_{eo} e^2})^{1/2} / L \tag{2}$$

$$X = eU/k T_{eo} \tag{3}$$

Here, L is the length of collecting channel walls; v_g the flame velocity component parallel to the collecting surface; ε_o is the permittivity of free space; b_o is the ionic mobility of the undisturbed flame flow; T_{eo}, n_{eo} are the electron temperature and density in the undisturbed flame flow at the sheath edge; e is the charge of electrons and U is the potential of the negatively biased channel walls.

In the limit of $R_E\alpha^2\chi^2<<1$, the sheath is thin compared with the thermal and boundary layers and the average ion current to the channel walls is determined by the processes of diffusion and convection with saturation characteristic obtained $I_i\approx U^o$ (Smy, 1976). In this limit, the cooling effects at the channel walls influence the ion transfer to the channel walls by reducing the ion density and temperature. In the limit of $R_E\alpha^2\chi^2>>1$ and $R_E\alpha^2<1$, the ion transfer to the channel walls is determined by convection and sheath effects and all ions convected into the sheath are driven to the channel surface. The velocity of the ions into the sheath is the resultant of the linear convective velocity along the flame flow, v_g, and radial drift velocity b_iE. In this limit, the cooling effects for the ion transfer to the channel walls are minimized and sheath-convection of ions from the undisturbed flame flow to the channel walls show reasonably strong voltage dependence with $I_i\approx U^{1/2}$. The average density of the sheath-convection current to the channel walls can be expressed as (Smy, 1976):

$$\langle j_i \rangle = 1.46(\varepsilon_0 b_{i0})^{1/4} U^{1/2}(n_{i0}ev_{g0} / X)^{3/4} \qquad (4)$$

The interrelated field-enhanced heat/mass flows to the channel walls can be approximated by the relations:

$$j_q \approx \frac{kT_f}{e}j_i \qquad \text{and} \qquad j_m \approx \frac{M_i}{e}j_i \qquad (5)$$

The conditions of the present experimental investigations were: $R_E\approx300\text{-}600$, $\alpha\approx10^{-4}$, $X\approx10^4$, with $R_E\alpha^2\chi^2>>1$ and $R^E\alpha^2<1$.

For such conditions, the sheath thickness at the channel walls ($y_s\approx0.02$ m) exceeds the thickness of the electric boundary layer ($y_e\approx0.0016$m), and a

Figure 2. The influence of the equivalenceratio of propane-air mixture on the voltage dependence of ion current.

field penetration into the hot reaction zone of a flame is observed. The results of experimental investigations demonstrate that the voltage dependence of the sheath convection current to the channel walls show a deviation from (4) and can be approximated as $I_i\approx V^n$, where $n\approx0.8\text{-}1.0$ (Fig. 2). Such deviation from (4) is observed for non-uniform flows and can be explained by a decrease in the average electric field strength in the expanding sheath (Smy, 1976).

In the limit of the sheath-convection, the sheath edge penetrates into the hot reaction zone and the electric field drives the hydrocarbon ions from the reaction zone to the channel walls by varying the ion density and flame composition profiles at the initial stage of the flame channel flow formation. Experimental studies of the field-enhanced ion transfer show that the electric field reduces and the mass fraction and absorption of the hydrocarbon

Figure 3. The electric field-induced variations in the ion density (a) and absorption profiles of hydrocarbon radicals (b) in the flame channel flow at L=30 mm.

ions in the central part of a flame while the ion density (ϱ_i) increases near the negatively biased channel walls (Fig. 3-a).

The field-enhanced ion transfer to the channel walls disturbs the mass fraction and absorption profiles of hydrocarbon radicals (C_3H_6, $\nu\approx3000$ cm^{-1}; C_2H_4, $\nu=949,2$ cm^{-1} and C_2H_2, $\nu=729,1$ cm^{-1}) as shown in Fig. 3-b. The most effective field-induced variations in mass fraction profiles are observed at the distance $L<30$ mm from the exit of a burner. At this stage of the flame flow

formation, intensive pyrolysis of propane in the central part of a flame develops and the ion current to the channel walls can be related to the field-enhanced transfer of the radical ions ($C_3H_3^+$, CHO^+, $C_2H_2^+$, etc.). It should be noticed that the sheath-convection of ions to the channel walls is followed by the surface recombination of ions and the neutralized species of hydrocarbon radicals (C_2H_2, C_3H_3) then return into the flame channel flow by increasing the density and absorption of these radicals near the channel
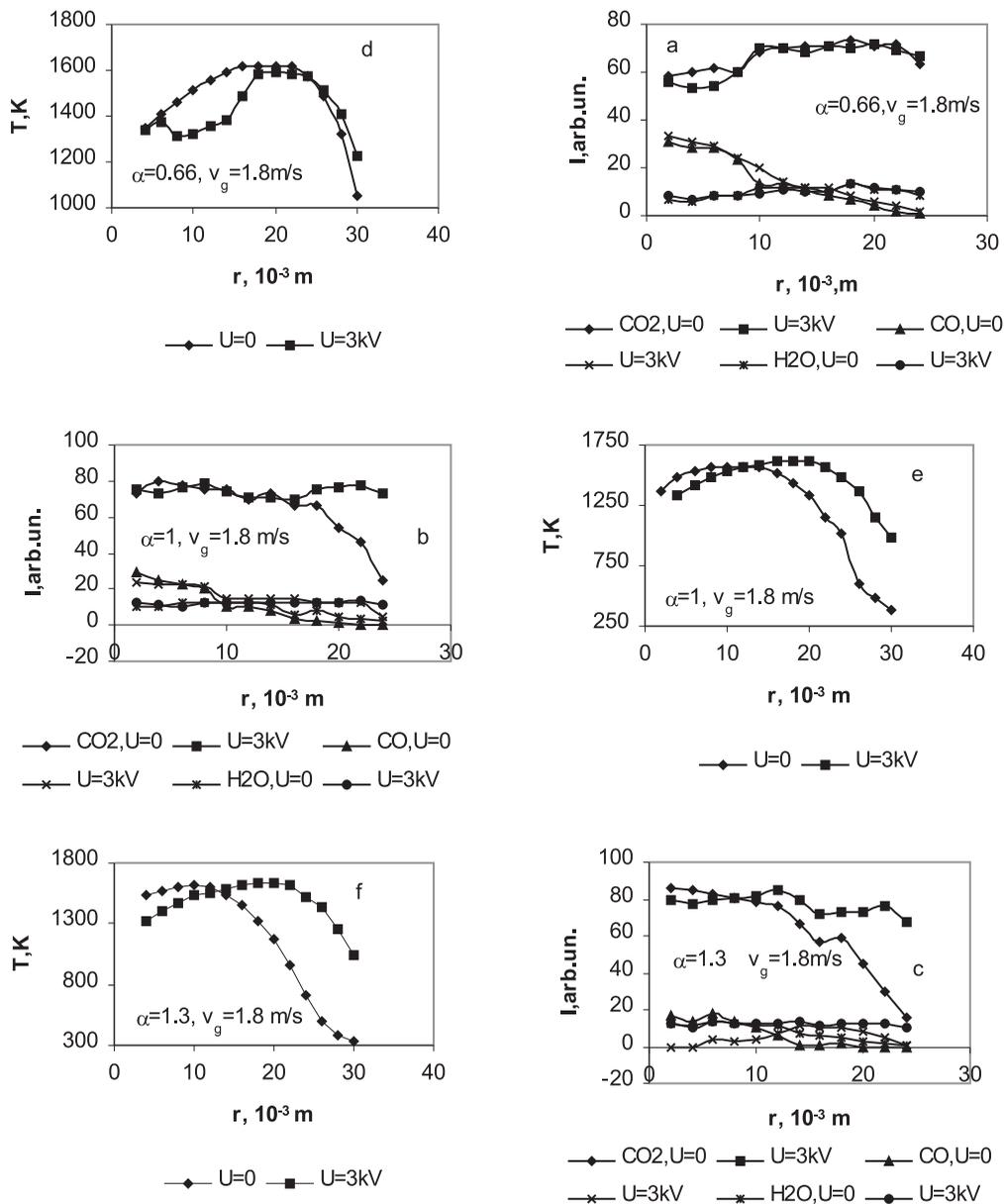
Figure 4. The electric field-induced variations in composition profiles of products (a-c) and temperature profiles (d-f) of the flame flow in the steel grid channel (L=30 mm).

walls. Therefore, in addition to the field-enhanced mass transfer of neutrals (ion wind), the density of hydrocarbon radicals at the channel walls can be increased due to the processes of surface recombination, depending on the ionized species transfer and relative rates of recombination.

The variation in the density profile of the hydrocarbons produces local variations in the rate of fuel combustion by varying the composition and temperature profiles of flame channel flows. Figure 4 presents the dominant electric field effects on fuel combustion in a steel grid channel. As can be seen from Fig. 4, the electric field reduces the mass fraction and absorption of products (CO, $\nu = 2141$ cm^{-1}, CO_2, $\nu = 2349.3$ cm^{-1} and H_2O, $\nu = 3651.7$ cm^{-1}) in the central part of a flame ($R < 15$ mm). In addition, it reduces the enthalpy produced due to the fuel combustion and flame temperature in this part of the flame. An increase in rate of fuel combustion, mass fraction of products and flame temperature is observed near to the channel walls, at $R > 15$ mm. Hence, the electric field shifts the reaction zone from the central part of a flame to the channel walls. The relation between the shape of the field-induced variations in mass-fractions of products and temperature profiles in the fuel-rich, stoichiometric and fuel-lean flame channel flows ($v_g = $const) shows that the greatest reduction in a temperature of the hot reaction zone ($R \approx 5\text{-}15$ mm) is achieved for fuel rich conditions (Fig. 4-d). The greatest increase in temperature, mass fraction and absorption of CO_2 and H_2O near the channel walls ($R \approx 15\text{-}20$

mm) is observed for fuel lean conditions, at $\alpha \approx 1.2\text{-}1.3$ (Fig. 4c, f). The variations in the composition profiles of the flame channel flows demonstrate that the sheath-convection of ions to the channel walls is followed by the field-enhanced penetration of the gas compounds through the steel grid channel walls into the surroundings ($R > 20$mm). This result in outer diffusion flame formation with increase in temperature, mass fraction and band absorption of products outside the channel walls (Fig. 4a-f). The voltage dependence of the band absorption of products outside the channel walls is illustrated in Fig. 5-a. The calorimetric measurements in the water-cooled channel show that the field-enhanced transfer of the reaction zone to the channel walls enhances the chemical enthalpy flux to the walls. Consequently, the field-enhanced enthalpy flux inside the water-cooled channel is followed by an increase in heat loss from the reaction zone of the flame (Fig. 5-b).

As can be seen from Fig. 5-b, in analogy with the increase in temperature and mass fraction of products at the channel walls (Fig. 4), the more intensive heat loss from a flame is observed for fuel lean conditions. In addition, for the fuel lean conditions the ion transfer to the channel walls increases (Fig. 2) and the maximum value of ion current is reached at $\alpha \approx 1.3$. The field-enhanced transfer of hydrocarbons from the reaction zone outward and the local variations in equivalence ratio can explain the most intensive heat/mass transfer for the fuel lean conditions of a propane-air mixture. As soon as the density of hydrocar-
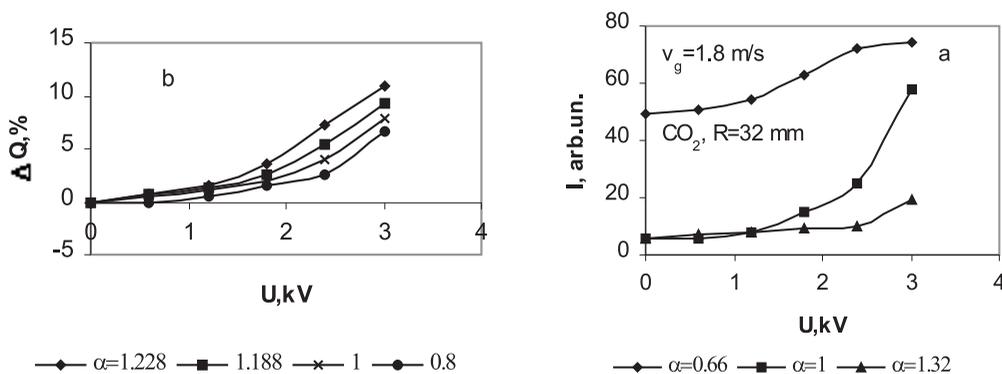
Figure 5. The influence of the equivalence ratio of propane-air mixture on the electric field-enhanced variations in the density and band absorption of CO_2 ($\nu = 2349.3$ cm-1) in the outer diffusion flame flow ($L = 30$mm) (a) and field-enhanced heat transfer from the flame to the channel walls (b).

bons near the channel walls increases, the reaction zone becomes more stoichiometric and burns more intense, as it follows from Fig. 4-c, f. As a result, the ion density in the reaction zone increases (Fig. 3-a) with resulting increase in heat/mass transfer to the channel walls (Equation 5) and a more intensive quenching of the reaction zone can be obtained. The field-enhanced variations in the temperature profiles at the exit of water-cooled channel are illustrated in Fig. 6.
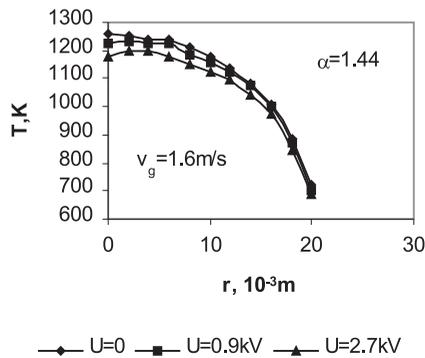
Figure 6. The electric field-enhanced variations in the temperature profiles of lean propane-air flame channel flow at the channel exit (L=70 mm).
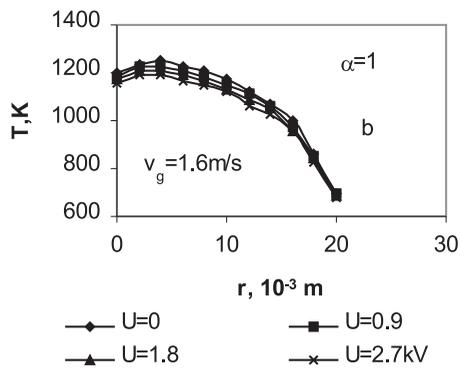
Finally, the results of the present experimental investigation clearly show that the electric field-enhanced variations in the flame temperature profiles and field-enhanced quenching of a reaction zone of a flame channel flow is greatly influenced by the relation between the electric and inertial forces. In terms of dimensionless parameters relation between the electric and inertial forces for electrogasdynamic flows (Vatashin *et al.*, 1983) can be expressed as:

$$S = \frac{en_i}{\rho v_g^2} lE \qquad (6)$$

Here l represents the axial length scale.

From equation (6), it follows that by increasing the rate of a flame channel flow (at $\alpha \approx$const, U$\approx$const) the value of S can be reduced and this in turn reduces the field effect on a flame quenching, as it is illustrated in Fig. 7. Therefore, the results of experimental investigations confirm that the relation (6) can be used to estimate the field effect on a flame. The electric field effect on a flame flow can be increased by increasing the field strength and ion density in a flame, while it can be decreased by increasing the flow rate.
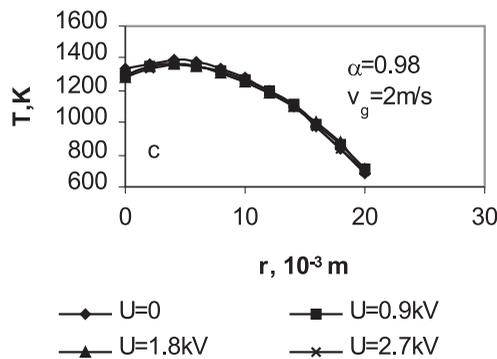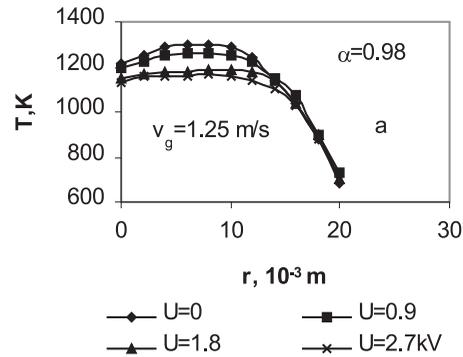
Figure 7. The variations of the field effect on the quenching of the flame channel flow by increasing the flow rate.

The presented results (Figs. 3-7) clearly demonstrate the electric field effect on the shape of the flame composition and temperature profiles and the processes of the heat/mass transfer and fuel combustion in the flame channel flows. In fact, the field-enhanced heat/mass transfer can result in effective quenching of the hot reaction zone of a flame channel flow. Consequently, the field effect on a flame can be used to control the fuel combustion and temperature-sensitive levels of NO_x emissions in products.

The reactions, which lead to the formation of NO in a flame channel flow, can be expressed by the Zeldovich two-step mechanism (Glassman, 1977):

$$N_2 + O \rightarrow NO + N,$$
$$k = 1.4 \cdot 10^{14} \exp(-78.5000/RT) \tag{7}$$

$$N + O_2 \rightarrow NO + O,$$
$$k = 6.4 \cdot 10^{9} \exp(-6280/RT) \tag{8}$$

The NO can be oxidized to NO_2 due to the evolution of the reactions:

$$NO + HO_2 = NO_2 + OH \text{ and}$$
$$NO + O = NO_2 \tag{9}$$
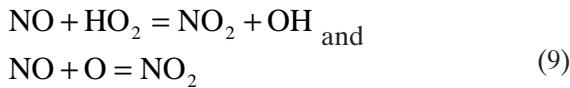
The dominant two-step process, which results at the formation of atomic oxygen in the flame, is associated with the dissociation of hydrocarbons and combustion of atomic hydrogen and can be expressed as:

$$C_2H_2 \rightarrow C_2 + 2H \text{ and}$$
$$H + O_2 \rightarrow OH + O \tag{10}$$
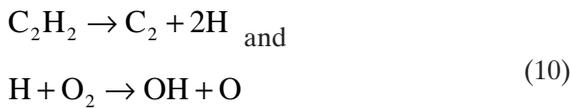
Because NO constitutes most of the NO_x produced in the natural gas and propane combustion, the reactions of NO formation limit the levels of total NO_x formation. Due to the high activation energy (E_a), the reaction (7) obviously controls the process of NO formation; hence reaction (7) is a rate-limiting step. The temperature levels of the hot reaction zone and mass fraction of atomic oxygen determine the Arrhenius-like rate of NO formation in a flame:

$$dNO/dt \approx [N_2][O]\exp(-E_a/kT) \tag{11}$$

From reactions (7-9), it follows that the process of the formation of thermal NO_x from atmospheric nitrogen arises primarily from the very temperature sensitive Zeldovich mechanism of NO formation. Therefore, the most intensive thermal NO_x formation can be obtained in a hot reaction zone of a flame channel flow. Hence, to obtain the effective reduction of NO_x levels in the pollutants, it is necessary to lower the temperature levels and mass fraction of atomic oxygen in a flame. Consequently, the field-enhanced variations in the temperature and mass fraction profiles, which result in a quenching of reaction zone of a flame (Figs. 6, 7), can produce the variations of NO_x levels in the emissions.

The experimental investigations of the electric field effect on NO_x formation confirm that the field-enhanced sheath-convection of positive ions to the water-cooled channel walls reduces the mass fraction of NO_x in a flame up to 80% (Fig. 8-a). In analogy with the processes in a free flame flow (Zake *et al.*, 1998), the field effect on the flame quenching and mass fraction of NO_x reduces to the minimum, if the central electrode is biased negatively.
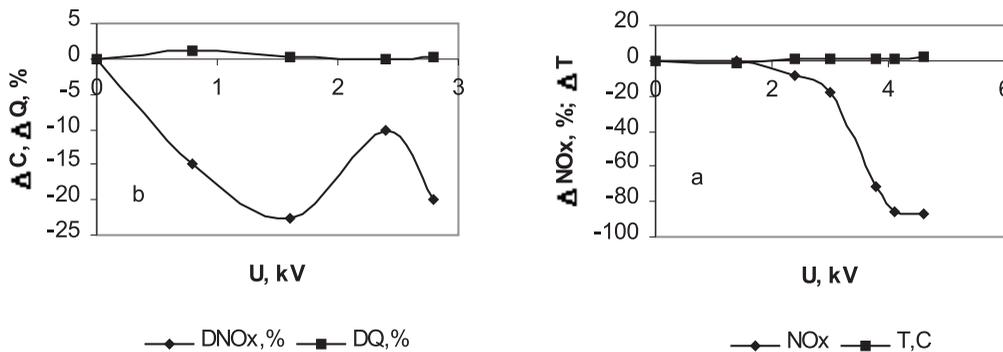
Figure 8. The DC electric field-induced variations of the levels of NO_x mass fraction in the calorimetric channel (Junker); (a) - the electric field is applied in a space between the burner and water-cooled channel walls; (b) - the electric field is applied in a narrow space between the central electrode and cylindrical surface of the burner.
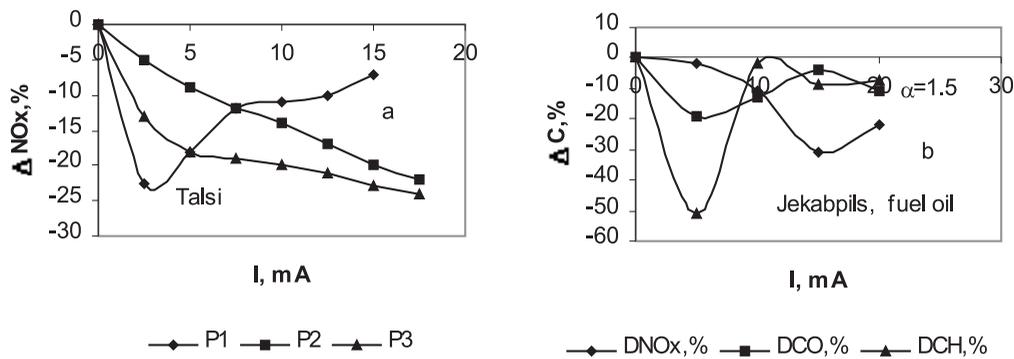
Figure 9. The DC field-induced variations of NO$_x$ mass fraction for different fuels: (a)- combustion of mineral oil in the district heating boiler house of Talsi for different pressures in a boiler (P1=40·10^4 n m^{-2}, P2=75·10^4 n m^{-2}, P3=100·10^4 n m^{-2}); (b)- combustion of fuel oil in the district heating boiler house of Jekabpils.
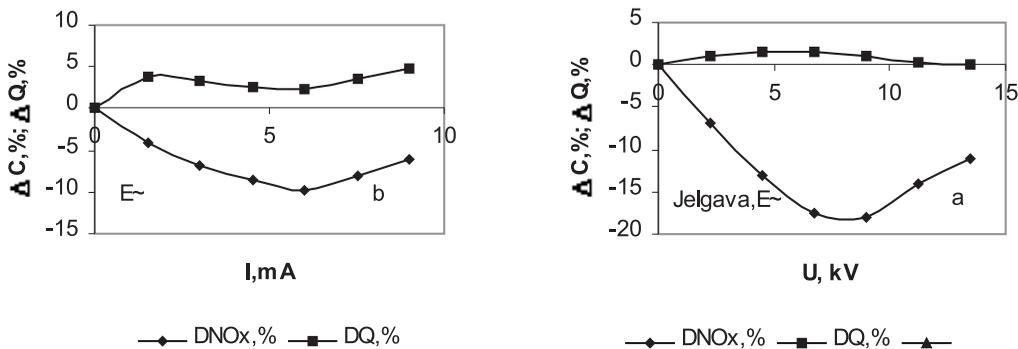
Figure 10. The alternative electric field-enhanced variations in the levels of NO$_x$ emissions during combustion of fuel oil: (a)- boiler house of Jelgava; (b)- boiler house of Jekabpils.

From Fig. 8-b, it follows that less effective reducing of the levels of NO$_x$ emissions is observed if the electric field is applied in a narrow space between the central wire electrode and cylindrical surface of the burner. Note, that by increasing the voltage of the central electrode, the mass fraction of NO$_x$ in the emissions continuously decreases to the minimum value (ΔNO$_x$≈25−35%) and then slightly increases as the electric field strength in the flame is increased and corona discharge develops (Fig. 8-b). The energy of the free electrons in the corona discharge increases by increasing the applied voltage and local intensity of the electric field (E): ε_e≈$e^2E^2/m_e\nu_e$. As pointed out by Slovecki (1980), in the limit of ε_e→6eV, the inelastic collisions between the free electrons and oxygen can result in a field-enhanced excitation of oxygen molecules and dissociation from the excited states: O$_2$+e=2O+e . Consequently, the field-enhanced dissociation of oxygen molecules is followed by an increase in a mass fraction of atomic oxygen and in a rate of NO formation (11) in a reaction zone of a flame. Hence, as soon as the corona discharge in a reaction zone of a flame flow develops, a slight increase in a mass fraction of NO$_x$ can be obtained, as it is observed in a range of U=1,5-2,5 kV (Fig. 8-b). By increasing the field strength, the inelastic collisions between electrons and NO initiates the field-enhanced dissociation of NO in the corona discharge, which is observed in the limit of ε_e → 9eV The dissociation of NO by electron impact is followed by an intensive reduction of NO$_x$ levels, as it is observed at U>2.5 kV and E>2.10^5 V m^{-1} (Fig. 8-b).

A similar shape of the electric field-enhanced variations in NO_x mass fraction is obtained from the results of the industrial investigations both for DC (direct current) and AC (alternative current) electric fields applied to the reaction zone of a flame (Figs. 9 and Fig. 10). Note that the field-enhanced reducing of NO_x levels (up to 30%) correlate with slight increase in the efficiency of heat generation (Fig. 10-a, b), mass fraction of CO and unburned hydrocarbons in the flame (Fig. 9-b). This correlation confirms the field-enhanced heat transfer from the flame to the water-cooled walls of the boiler with resulting quenching of the reaction zone. Reducing the rate of fuel combustion and lowering of the levels of NO_x emissions in the reaction zone of the flame.

Therefore, the laboratory and industrial investigations confirm that the electric field-enhanced effects of ion drift motion can be used to produce local variations in the rate of fuel combustion and to reduce the levels of NO_x emissions in a flame. In addition, the dissociation of O_2 and NO from excited states can be important, if the corona discharge develop in a flame.

CONCLUSIONS

A brief account of the DC electric field-induced variations in the processes of heat/mass transfer and fuel combustion in the flame channel flows is given. The laboratory and industrial investigations demonstrate the following:

The field-enhanced processes of sheath-convection of ions to the channel walls produces local variations in temperature and mass fraction profiles in the flame channel flows and shift the reaction zone from the central part of a flame to the channel walls.

The local variations in temperature and mass fraction profiles of the reaction zone produces a field-enhanced heat transfer to the channel walls with resulting quenching of the reaction zone.

The field-enhanced quenching of the reaction zone at low energies of the free electrons in the flame ($\varepsilon_e < 6eV$)reduces the rate of the thermal NO formation and levels of NO_x emissions. Then a decrease in a mass fraction of NO_x emissions up to 80% is detected.

For strong electric fields there develops a corona discharge in the flame is developed. Under conditions of corona discharge, the variations of the mass fraction of NO_x in the flame flow are strongly influenced by the field-enhanced dissociation of the gas compounds by the electron impact and are determined by the energy of the free electrons.

In the range of $9 > \varepsilon_e > 6$ eV, the corona discharge in the reaction zone enhances the dissociation of oxygen molecules by electron impact and field-enhanced increase in a rate of NO formation is followed.

At $\varepsilon_e > 9$ eV , the inelastic collisions between the electrons and NO initiate a field-enhanced dissociation of NO. Then, an intensive reducing of the levels of NO_x in the emissions then follows and cleaner combustion of fuels can be obtained.

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