

MODELING OF NITROGEN OXIDE FORMATION IN A TURBULENT DIFFUSE FLAME

É. P. Volkov and N. Yu. Kudryavtsev

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A mathematical model is represented for a turbulent diffuse methane flame in air with nitrogen oxide formation in the combustion products according to the Ya. B. Zel'dovich reaction mechanism taken into account. The model is tested on experimental data for diffusion and premixed flames.

The requirement that occurs for checking the emission of contaminating substances results in the necessity to investigate processes of nitrogen oxide formation in outgoing gases of steam boilers of thermal electric stations. In this connection, the problem of describing the processes of development and burn-up of a turbulent diffusion flame that is formed during combustion of a methane jet in an air flow is urgent during combustion of the gas fuel in steam boiler furnaces. The process of gas fuel combustion in the majority of power plants with separate jet delivery of fuel and oxidizer in the furnace chamber can be described by using the model of a turbulent diffusion flame. Thus, for instance, the gas fuel in an oil-gas burner of TKZ-VII type is delivered to the furnace by a system of direct-flow jets in the surrounding air coflow. A parabolic approximation of the Navier-Stokes equations is used to describe such flows, which is convenient for numerical realization by using an economical Marsh iteration-free method.

It is established in the theory of diffusion combustion that the reaction zone thickness is, as a rule, substantially less than the characteristic dimensions of the problem [1]. This is associated with the fact that the chemical combustion reactions proceed at high speed only for high temperatures, while the reaction rate is diminished radically with distance from the flame front. Random flame front vibrations occur because of random fluctuations in primarily the reagent concentrations as well as the flux rates, and for a high vibration amplitude this results in a reduction in the mean completeness of the combustion. Therefore, to estimate the efficiency of the combustion process it is necessary to have information about the probability distributions of the fuel and oxidizer concentrations.

The main role of the concentration (or temperature) distribution density in the theory of turbulent combustion is manifest in that the chemical reaction rates that depend nonlinearly on the temperature and concentration must be averaged during solution of the equations describing the behavior of the reacting gas. The statistical characteristics of the flame front vibrations are here directly independent of the chemical reaction rates, however, an analogous assumption concerning substantially nonequilibrium processes, nitrogen oxide formation, say, is not valid.

A single-stage irreversible fuel + oxidizer \rightarrow product reaction is examined in this paper, which agrees with a real process in its total effects and does not take account of the essential intermediate stages. It is assumed here that the effective Prandtl and Schmidt numbers are mutually equal at any point of the mixture volume. A certain conserving quantity, a passive scalar f for which the space-time distribution in the reacting flow does not differ from its distribution in a medium with invariant chemical composition, appears in the flow for such a selection of the combustion model. Then the problem reduces to describing the probability distribution density of the random variable β that is essential for the development and progress of the chemical reactions. Such a random variable can be either the passive scalar f or the fuel concentration m_f or certain other flow parameters.

The most constructive method of overcoming the difficulties associated with modelling chemical reactions in a turbulent medium is the a priori assignment of the probability distribution density in the form of a dependence on a small number of parameters determined from

the solution of equations for low order moments. Consequently, in conformity with the Lockwood and Naguib approach [2], the probability distribution density is selected in the form of a truncated normal distribution with alternation taken into account. The truncated distribution of the random variable β subject to a normal law and defined in the interval $(-\infty, +\infty)$ has the following form

$$P(\beta) = \begin{cases} A\delta(\beta) & \text{for } \beta = 0, \\ p^*(\beta) & \text{for } 0 < \beta < 1, \\ B\delta(\beta - 1) & \text{for } \beta = 1, \\ 0 & \text{for } \beta < 0 \text{ for } \beta > 1, \end{cases}$$

where

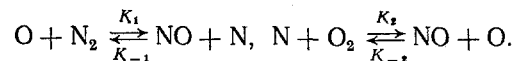
$$p^*(\beta) = \frac{1}{\sigma \sqrt{2\pi}} \exp \left[-0,5 \left(\frac{\beta - \mu}{\sigma} \right)^2 \right],$$

$$A = \int_{-\infty}^0 p^*(\beta) d\beta, \quad B = \int_1^{\infty} p^*(\beta) d\beta, \quad \int_{-\infty}^{\infty} p^*(\beta) d\beta = 1.$$

The distribution density function $P(\beta)$ is constructed after the appropriate conservation equations for the first μ and second σ^2 moments of this distribution have been solved. The quantity f (or m_f) is taken as the first moment in the problem and the averaged square of the concentration fluctuations g as the second. Knowing the function $P(\beta)$ the value, averaged over time, of any variable Φ that is a function of the random variable β can be

found from the formula $\bar{\Phi} = \int_0^1 \Phi(\beta) P(\beta) d\beta$. Afterwards, the average square of the fluctuating component of this variable $\overline{\Phi'^2} = \int_0^1 [\Phi(\beta) - \bar{\Phi}]^2 P(\beta) d\beta$ is easily determined.

The main source for nitrogen oxide formation in the combustion of gas fuel is atmospheric nitrogen. The mechanism of thermal NO_x formation is investigated in detail, it is the Zel'dovich reaction scheme [3]:



Using the assumption about the quasistationary concentration of the atoms N, an expression can be written for the rate of NO formation:

$$W_{\text{NO}} = \frac{dr_{\text{NO}}}{d\tau} = \frac{2K_1 K_2 r_{\text{O}_2} r_{\text{N}_2} r_{\text{O}}}{K_{-1} r_{\text{NO}} + K_2 r_{\text{O}_2}} \left(1 - \frac{K_{-1} K_{-2}}{K_1 K_2} \frac{r_{\text{NO}}^2}{r_{\text{N}_2} r_{\text{O}_2}} \right). \quad (1)$$

On the basis of experimental observations, Ya. B. Zel'dovich concluded that the rate of NO generation is much less than the combustion velocity and that the main part of NO is formed after completion of the combustion. Therefore, the rate of nitrogen oxide formation can be determined under the assumption of equilibrium of the combustion reactions. Consequently, when computing complex multidimensional flows we neglect the interrelation between NO generation and the combustion process and we compute the rate of nitrogen oxide formation from atmospheric nitrogen by the Zel'dovich chain reaction mechanism by using equilibrium concentrations of the chemical components being formed during combustion. We obtain the necessary equilibrium concentrations of the components from auxiliary equilibrium computations by constructing interpolation polynomials dependent on the combustion temperature and the coefficient of excess oxidizer.

To analyze a quasistationary jet turbulent axisymmetric flow with combustion it is necessary to integrate numerically a system of equations, averaged over time, for the continuity, motion, energy, and concentration that is supplemented by appropriate equations of the turbulence model. The nonequilibrium dynamics of turbulence development is described within the framework of the semiempirical two-parameter Rodi-Spalding k- ϵ model [4]. In writing the

equations we neglect terms with density fluctuations since taking such terms into account radically increases the computation time. Within the framework of the gradient hypotheses for estimation of turbulent flows, the system of averaged equations has the form

$$\frac{\partial}{\partial x}(\rho u) + \frac{1}{r} \frac{\partial}{\partial r}(r \rho v) = 0; \quad (2)$$

$$\rho u \frac{\partial u}{\partial x} + \rho v \frac{\partial u}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \mu_{ef} \frac{\partial u}{\partial r} \right); \quad (3)$$

$$\begin{aligned} \rho u \frac{\partial h_0}{\partial x} + \rho v \frac{\partial h_0}{\partial r} &= \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\mu_{ef}}{\sigma_h} \frac{\partial h_0}{\partial r} \right) + \\ &+ \frac{1}{r} \frac{\partial}{\partial r} \left[r \mu_{ef} \left(1 - \frac{1}{\sigma_h} \right) \frac{\partial}{\partial r} \left(\frac{u^2}{2} \right) \right] - Q_{RAD}; \end{aligned} \quad (4)$$

$$\rho u \frac{\partial f}{\partial x} + \rho v \frac{\partial f}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\mu_{ef}}{\sigma_j} \frac{\partial f}{\partial r} \right); \quad (5)$$

$$\rho u \frac{\partial m_f}{\partial x} + \rho v \frac{\partial m_f}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\mu_{ef}}{\sigma_j} \frac{\partial m_f}{\partial r} \right) - R_f; \quad (6)$$

$$\rho u \frac{\partial k}{\partial x} + \rho v \frac{\partial k}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\mu_{ef}}{\sigma_k} \frac{\partial k}{\partial r} \right) + \mu_{ef} \left(\frac{\partial u}{\partial r} \right)^2 - \rho \epsilon; \quad (7)$$

$$\rho u \frac{\partial \epsilon}{\partial x} + \rho v \frac{\partial \epsilon}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\mu_{ef}}{\sigma_\epsilon} \frac{\partial \epsilon}{\partial r} \right) + C_{\epsilon 1} \frac{\epsilon}{k} \mu_{ef} \left(\frac{\partial u}{\partial r} \right)^2 - C_{\epsilon 2} \frac{\rho \epsilon^2}{k}; \quad (8)$$

$$\rho u \frac{\partial g}{\partial x} + \rho v \frac{\partial g}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\mu_{ef}}{\sigma_g} \frac{\partial g}{\partial r} \right) + C_{g1} G - C_{g2} \rho \frac{\epsilon}{k} g; \quad (9)$$

$$\begin{aligned} \rho u \frac{\partial m_{NO}}{\partial x} + \rho v \frac{\partial m_{NO}}{\partial r} &= \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\mu_{ef}}{\sigma_j} \frac{\partial m_{NO}}{\partial r} \right) + \\ &+ \frac{\mu_{NO}}{10^8} W_{NO} - m_{NO} \left(u \frac{\partial \rho}{\partial x} + v \frac{\partial \rho}{\partial r} \right). \end{aligned} \quad (10)$$

The system (2)-(10) is supplemented by standard relationships of the turbulence model

$$\begin{aligned} \mu_{ef} &= \mu_t + C_{\mu\rho} \frac{k^2}{\epsilon}, \quad C_{\mu} = 0,09 - 0,04b, \\ b &= \left| \frac{\Delta y}{2(u_{\max} - u_{\min})} \left(\frac{\partial u_{0,c}}{\partial x} - \left| \frac{\partial u_{0,c}}{\partial x} \right| \right) \right|^{0,2}, \\ G &= \mu_{ef} \left(\frac{\partial f}{\partial r} \right)^2 \quad \text{or} \quad G = \mu_{ef} \left(\frac{\partial m_f}{\partial r} \right)^2, \end{aligned} \quad (11)$$

$C_{\epsilon 1} = 1,43$; $C_{\epsilon 2} = 1,92 - 0,0667b$; $C_{g1} = 2,8$; $C_{g2} = 2,0$; $\sigma_h = \sigma_j = \sigma_g = 0,9$; $\sigma_k = 1,0$; $\sigma_\epsilon = 1,3$, as well as an expression describing the connection of the total specific enthalpy to the temperature in the flow

$$h_0 = m_f H_f + \frac{u^2}{2} + \int_0^T \sum_i m_j C_{p_j}(T) dT.$$

Moreover, it is assumed that the reacting gas mixture is subject to the equation of state for a perfect gas while the pressure in the stream is everywhere constant and equal to atmospheric pressure.

Certain equations of the system (2)-(10) have source terms needed in additional modelling. The source in the energy equation (4) consists of two parts, a completely defined term describing kinetic heating, and the term Q_{RAD} describing heat elimination from the flame due to intrinsic flame radiation. The approximation of an optically thin layer, that is valid for

a diffusion flame because of the small combustion zone thickness [1], is used to represent this last term. It is also assumed that radiation from the flame into the surrounding medium is determined by radiation of the triatomic gases H_2O and CO_2 that are present in the combustion products in quantities close to their equilibrium concentrations. Then the quantity Q_{RAD} can be determined from the formula [5], where r_i^* are the molar fractions

$$Q_{RAD} = 5,67 \left(\frac{T}{100} \right)^4 [407,41 \exp(-1,51 \cdot 10^{-3}T) r_{CO_2}^* + 79,72 \exp(-1,4 \cdot 10^{-3}T) r_{H_2O}^*].$$

Modeling of the source R_f describing the rate of fuel disappearance because of chemical combustion reactions in Eq. (6) for the fuel concentration is realized by combining the Arrhenius mode of writing the generalized combustion reaction rate with the Spalding hypothesis about the mean reaction rate in a turbulent flow. This hypothesis, based on the model of turbulent vortex breakdown, admits that the reaction rate cannot be greater than the expression $C_{RG}^{1/2} \rho(\varepsilon/k)$ [6], consequently the term R_f finally appears as

$$R_f = \min \left\{ 10^{10} \rho^2 m_f m_{ox} \exp \left(- \frac{18400}{T} \right); C_{RG}^{1/2} \rho \frac{\varepsilon}{k} \right\}. \quad (12)$$

In industrial power plants the gas fuel proceeds to the burner at the temperature of the surrounding medium and traverses a certain path to ignition in the furnace. Reverse combustion product currents that can approach the very root of a gas-air jet effluxing from the burner are a source of fuel mixture heating prior to ignition in the majority of cases. The parabolic formulation of the problem does not afford a possibility of taking strict account of such return flows, consequently, balance relationships of the mass, momentum, and energy and the assumption about ideal and instantaneous mixing of the reverse currents of the combustion products upon contact with the external air stream of the gas-airjet are used to take account of ignition in the paper. Having been given the reverse current parameters obtained from tests and the discharge distribution functions in the whole return flow energy, the influence of the reverse currents of the combustion products on the heating of the jet initial section, on its concentration dilution, and on the momentum change in any transverse section of the jet. After conversion of the external air flow parameters these new quantities are given as boundary conditions for the mixing layer, the influence of the reverse currents is thereby transmitted over the whole jet section down to its axis.

The boundary conditions for all the equations of the system (2)-(10) are similar. The condition that the derivative of the desired variable with respect to the radius is zero $\partial\phi/\partial r = 0$ is given on the axis of symmetry. The flow parameters on the outer jet boundary are assumed equal to known analogous parameters in the unperturbed external air stream. The boundary conditions are given analogously on the inner free boundary of the potential jet core. Moreover, initial flow parameter distributions are given for $x = 0$.

The obtained system of parabolic equations in a cylindrical coordinate system is solved by the numerical Patankar-Spalding method by using the GENMIX program [7]. A difference mesh with 40 nodes in the transverse section is used for the numerical integration. The mesh spacing and the integration spacing are substantially nonuniform during passage from section to section.

Equation (10) drops out of the total scheme of the solution. The singularity of this equation is that the kinetic source term induces strong rigor, consequently, it is solved in two stages by the method of splitting. The rigorous ordinary differential equation

$$\frac{dr_{NO}}{dx} = \frac{W_{NO}}{u}$$

is initially solved along a trajectory by the Gear numerical method [8] with automatic step selection, and then (10) with a zero source term is solved on the same mesh, as are the other conservation equations.

A feature of the method for solving the system of equations is also the double solution of the system in each section. Initially the shift is computed without taking account of the combustion chemical reactions whereupon the relationship of the oxidizer and fuel is determined at each point of the transverse section. Then only the coefficient of excess oxidizer α just determined is used in the duplicate computation of the total system of equations for the representation of nitrogen oxide formation.

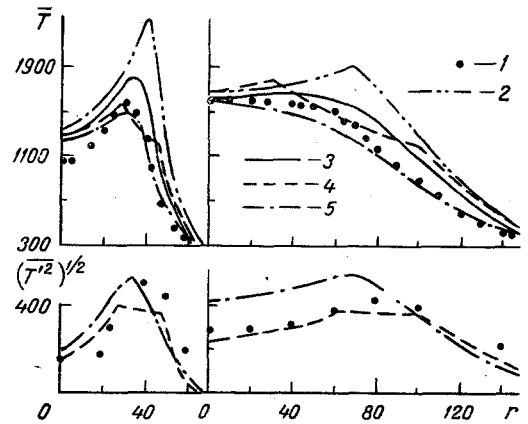


Fig. 1. Transverse averaged temperature and temperature fluctuation profiles for a methane diffusion flame, $Re = 15,000$, $d = 7.74$ mm: a) $x/d = 36.2$; b) $x/d = 113.7$; 1) experimental data [9]; 2) model A; 3) model B; 4) model C; 5) model D. \bar{T} , K; $(\bar{T}')^2)^{1/2}$, °C; r , mm.

RESULTS OF THE COMPUTATIONS

Verification of the computation model is ordinarily performed by comparison with experimental data or with computations of other authors. Taking this circumstance into account, the paper [9] was selected in which there is a detailed experimental investigation of the combustion of a subsonic submerged jet of methane in air. Four different combustion models were also tested during execution of the computations. Let us present a brief description of these models.

Combustion Model A. It is assumed that the combustion reactions pass at an infinite rate while the turbulent temperature and concentration fluctuations can be neglected. In this case there are no equations (6), (9), and (10) in the reduced system.

Combustion Model B. It agrees with the combustion mode used in the GENMIX program [7]. Neglecting the composition and temperature fluctuations is allowed here but the finite combustion reaction rate expressed by using the Spalding hypothesis about turbulent vortex breakdown is taken into account. Equation (9) is here eliminated from the system.

Combustion Model C. It is assumed that the chemical reaction rate is infinite but turbulent composition and temperature fluctuations exist in the stream. Then the parameter G describing concentration fluctuation generation in (9) depends on f according to (11) and equations (6) and (10) are eliminated from the system.

Combustion Model D. The complete system of differential equations (2)-(10) is solved with a finite combustion chemical reaction rate (12) and turbulent fluctuations taken into account. The parameter G in (9) is here evaluated in terms of m_f in conformity with (11).

Results of a computation as compared with experimental data are represented in Fig. 1. It follows from the graphs presented that just the most idealized combustion model A does not satisfactorily describe the real process. The model D can be extracted from the rest. It describes both the mean and the fluctuation characteristics of a flow with combustion most adequately, it is here most complex in formulation and therefore takes account of a greater quantity of factors of the real combustion process.

Several experiments on the combustion of gas jets described in the literature were still relied upon for more detailed testing of the proposed model. Tests on measuring the parameters of a burning gas jet, represented in [10], are analogous to those examined above from [9], but differ by the initial jet parameters and dimensions. Results of experiments [10] together with computed curves obtained by using the combustion models B and D are shown in Fig. 2. As is seen from the graphs, the correspondence between computation and experiment is good.

It was remarked in an analysis of the mathematical model produced that it can be applied to describe not only diffusion combustion of pure gas jets in air but also pre-mixed gas jet

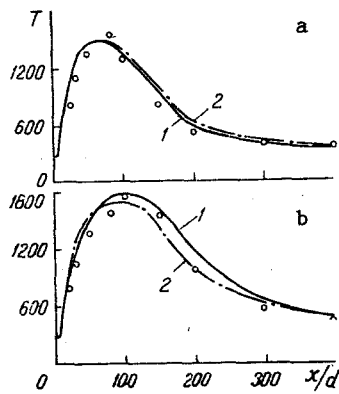


Fig. 2. Axial averaged temperature distributions over the jet length for a methane diffusion flame for different flow modes in comparison with experimental measurements [10], $d = 5$ mm: a) $Re = 2920$; b) $Re = 11,700$; 1) combustion model B; 2) combustion model D. T, K.

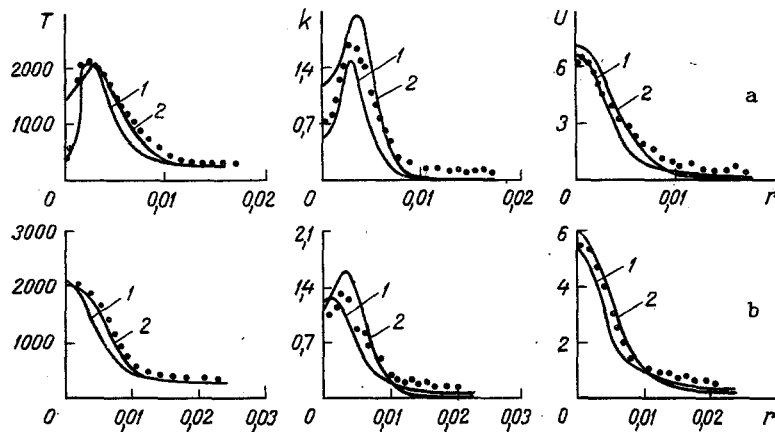


Fig. 3. Transverse temperature, turbulence kinetic energy, and longitudinal velocity profiles for a floating jet turbulent flame of premixed methane and air, $d = 3$ mm, a) $x = 0.03$ m; b) $x = 0.05$ m; points are experiment [11]; 1) a computation with 33 combustion reaction kinetics [11]; 2) a computation using the diffusion combustion model B. T, K; k, m^2/sec^2 ; U, m/sec, r, m.

combustion with air emerging in a submerged air medium. To verify this deduction, results of Sommer [11] experiments on combustion of a floating jet of premixed methane and air with an excess fuel coefficient $1/\alpha = 1.6$ in air was utilized.

The computational-experimental paper [11] is of considerable interest primarily because the author did not limit himself to measuring the fundamental stream parameters therein but performed a numerical computation of jet combustion by using a detailed kinetic mechanism of 33 elementary gas combustion reactions. As follows from Fig. 3, the proposed diffusion flame model B describes the experiment well and in practice as on no account a significantly more complex and tedious computation of the combustion process with complete chemical kinetics.

The careful verification performed for the combustion model produced for a gas jet showed its applicability and reliability in describing the combustion of turbulent diffusion flames as well as confirmed the well-foundedness of modelling the combustion process by a single-stage generalized chemical reaction.

The proposed mathematical model for gas jet combustion was applied to the investigation of a burning methane jet under almost real conditions in steam boiler furnaces. The initial jet radius was selected from requirement of correspondence between the temperature profiles

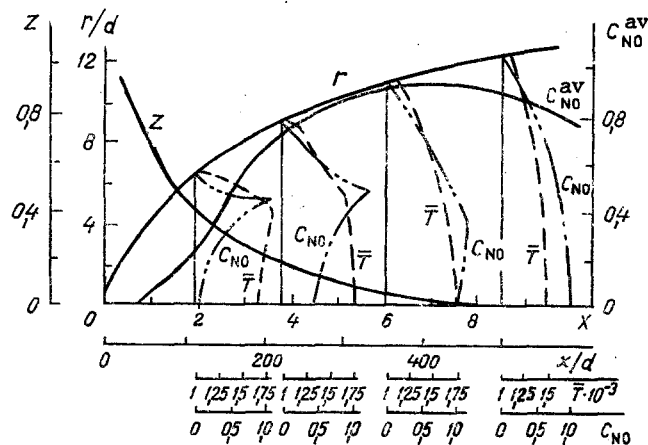


Fig. 4. Change in the averaged temperature \bar{T} , K and nitrogen oxide concentration C_{NO} , g/m³ profiles, dependence of the mean nitrogen oxide concentration over a section C_{NO}^{av} , g/m³ along the jet length, and also fuel burn-up curve Z ; Γ is the jet boundary; computation using the model D. x , m.

in the computational modification and in the real gas burner. The initial gas and air temperatures were 273 and 600 K, respectively, and the initial velocities were 120 and 40 m/sec. The computation was performed with gas jet heating by reverse currents of the combustion products taken into account.

The results of computing the flow modification under investigation with nitrogen oxide formation taken into account are presented in Fig. 4, which illustrates well the mutual location and dynamics of the shift of the maximums of the transverse NO concentration and temperature profiles. It follows from Fig. 4 that the nitrogen oxide temperature and concentration maximums do not agree and that the nitrogen oxide formation zone determined not only by the temperature level but also in conformity with the excess air coefficient at a given point of the section has quite limited dimensions as compared with the width of the jet. The quite definite maximum NO concentration in this zone is gradually smoothed out because of the diffusion transfer of concentration over the whole jet width and displacement of the location of this zone over the section. As the fuel in the jet burns up the nitrogen oxides temperature maximum and the concentration maximum after it are displaced in the jet axis direction and the absolute values of the temperature and concentration maximum start to diminish for a large fraction of the burn-up. Computation of the mean nitrogen oxides concentration over the section (Fig. 4) results in values close to the measured nitrogen oxide concentration levels in furnace chambers of oil-gas steam boilers.

Therefore, the mathematical model produced for a gas flame permits obtaining a sufficiently true pattern of combustion in a turbulent diffusion flame. Even under serious kinetic simplifications associated with the introduction of a single-stage combustion reaction, the nitrogen oxide temperature and concentration levels computed by using this model are close to the real corresponding parameters measured in steam boiler furnaces. The model produced can be used in investigations of gas combustion processes with combustion product recirculation, in studies of modal parameters influencing the nitrogen oxide emission as well as in an appropriate finishing up for investigations of the influence of water injection in the combustion zone on the nitrogen oxide output.

NOTATION

x , r , longitudinal and transverse coordinates; u , v , longitudinal and transverse stream velocity components; ρ , gas mixture density; h_0 , total specific enthalpy; m , mass concentration; k , ϵ , turbulence kinetic energy and rate of its dissipation; $\mu_{ef} = \mu_l + \mu_t$, effective, laminar, and turbulent viscosity; T , temperature, K; $f = (Y - Y_0)/(Y_1 - Y_0)$ a conservative scalar quantity; $Y = m_f - m_{Ox}/S$, Schwab-Zel'dovich variable; s , stoichiometric relationship

between the fuel components; g , averaged square of the concentration fluctuations; σ_h, σ_j , effective Prandtl and Schmidt numbers; $C_\mu, C_{\epsilon 1}, C_{\epsilon 2}, C_{g1}, C_{g2}, \sigma_k, \sigma_\epsilon, \sigma_g$, standard constants of the turbulence model; Q_{RAD}, heat flux due to intrinsic radiation of the burning gas; R_f , rate of fuel burn-up in chemical combustion reactions; W_{NO} , rate of nitrogen oxide formation; C_R , a constant in the Spalding turbulent vortex breakdown model; Δ_y , characteristic thickness of the mixing layer; u_{max}, u_{min} , maximal and minimal values of the longitudinal velocity in a given jet section; β , a random variable; $p^*(\beta), \mu, \sigma^2$, probability density functions of a normal distribution and the first and second moments of this distribution; r_{N_2}, r_{O_2}, \dots , molar-volume component concentrations moles/cm³; μ_{NO} , No molecular weight; H_f , calorific power of the fuel; $C_{pj}(T)$, specific isobaric specific heat of the j -th mixture component; δ , Kronecker delta. Subscripts: f , fuel; ox , oxidizer; $o.c.$, jet axis; 1 and 0 refer, respectively, to the fuel flux and oxidizer flux; upper bar is for average with respect to time, and prime is for fluctuating quantity.

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