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INVESTIGATIONS OF SULFUR OXIDES FORMATION AND DECOMPOSITION IN A GAS TURBINE COMBUSTOR WITH STEAM INJECTION

Introduction: This paper presents the results of theoretical investigations of sulfur oxides formation and decomposition in a gas turbine combustor with steam injection into the primary combustion zone. Corresponding mathematical model of a gas turbine combustor based on solutions of the differential equations of mass, impulse and energy conservation for the multi-component, turbulent, chemically reacting system is developed. The results of the model verification confirm its validity for the wide spectrum of operating modes. Specific features of working process of the combustor with eco-friendly and energy steam supply are determined. An eight-reaction kinetic mechanism of sulfur compounds formation and decomposition is used for 3D calculation of the full-scale design of gas turbine combustor. Theoretical investigations of sulfur oxides formation and decomposition in gas turbine combustors with steam injection at sulfur mass content of 1 and 3% in hydrocarbon fuel are performed. The obtained results and recommendations can be used for modeling the operational modes of gas turbine combustors with steam injection, as well as geometry optimization, and the engineering design of prospective power generation units.

Keywords: gas turbine, combustor, sulfur oxide, modeling, emission.

1. Introduction

An important direction of raising the efficiency of hydrocarbon fuel application, reduction of its consumption, and increasing the operation and environmental parameters of up-to-date stationary and transport power systems is usage of modern combustion technology [11, 12, 19].

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The main requirements for modern gas turbine engines (GTE) apart from economic efficiency, reliability and long service life are also a significantly low level of pollutants emission and high stability of combustors.
One of the most prospective methods for decreasing pollutants emission is steam injection into the primary combustion zone, which reduces the flame temperature. At the same time, decreasing the temperature causes destabilization of the burning process, especially for partial load operation modes. That is why additional plasma support is desired and recommended for sustaining the working processes in combustors with steam injection [3, 13-15, 21].
Sulfur compounds, which may form at combusting organic fuels, are toxic and corrosion-active. Thus, the task of predicting of their formation and decomposition in gas turbine combustors with steam injection is urgent especially at operating on liquid hydrocarbon fuels. There is quite a huge amount of sulfur in liquid fuels where its mass content may vary from 0.5 to 3%. Emission of sulfur oxides SO_x (SO_2 + SO_3) during combustion is defined by oxidation of sulfur in fuel. A part of gaseous oxides SO_x may condense on solid particles and metal surfaces with formation of sulfur acid or react with sulfates formation under specific conditions in the presence of water. Thus, sulfur compounds may lead to corrosion of elements of a combustor, a turbine or heat exchange equipment [6].
The main objectives of the present work are theoretical investigation of the main special features of sulfur oxides formation and decomposition in a gas turbine combustor with steam injection.

2. Approach and mehtods

The aim of mathematical modeling of turbulent combustion in gas turbine combustors is determination of hydrodynamics and chemical kinetics features as well as interaction between a turbulent flow and chemical reactions.
Combustor mathematical model is based on solving an equation system which describes convection and diffusion transfer of concentrations for each of the reacting mixture components.
An offered mathematical model of a gas turbine combustor with steam injection is based on the following equations [1, 2, 4, 8-10, 18, 22-24]:
Continuity equation. A mass conservation equation or a continuity equation for compressible and incompressible media shall be represented as follows:

$$\frac{\partial \rho}{\partial t} + \nabla(\rho \vec{\upsilon}) = S_m \ , \tag{1}$$

where:
ρ – flow mass density,
$\vec{\upsilon}$ – local flow velocity vector,
S_m – source term which defines the mass led to the flow by any way.

Momentum conservation equation. A momentum conservation equation in a fixed system of reference may be formed as follows:

$$\frac{\partial}{\partial t}(\rho\vec{\upsilon})+\nabla(\rho\vec{\upsilon}\vec{\upsilon})=\nabla p+\nabla\cdot(\bar{\bar{\tau}})+\rho\vec{g}+\vec{F} \quad , \tag{2}$$

where:
p – static pressure,
$\rho\vec{g}$ – gravitation force,
$\vec{F}$ – external forces,
$\bar{\bar{\tau}}$ – pressure tensor.

Energy conservation equation. In a general form an energy conservation equation is written as follows:

$$\frac{\partial}{\partial t}(\rho E)+\nabla\cdot(\vec{\upsilon}(\rho E+p))=\nabla\cdot\left(-\vec{J}_q+(\bar{\bar{\tau}}_{eff}\cdot\vec{\upsilon})\right)+S_h\,, \tag{3}$$

where:
E – total energy,
$\vec{J}_q$ – mixture heat flow density,
$\bar{\bar{e}}_{eff}\cdot\vec{v}$ – viscous heating,
S_h – sourse term which includes heat of chemical reaction and any other volumetric heat sources.

Mass conservation equation for chemical components of the mixture. If there is need to consider the equations for chemical substances, then it is possible to obtain the concentration of each component Y_i by solving the equation for its convection diffusion transfer. In general this equation has the following form:

$$\frac{\partial}{\partial t}(\rho Y_i)+\nabla(\rho\vec{\upsilon}Y_i)=-\nabla\cdot\vec{J}_i+R_i+S_i\,, \tag{4}$$

where:
R_i – rate of forming the *i*-th component as a result of a chemical reaction,

S_i – rate of additional forming of the *i*-th component from a disperse phase or other sources,

$\vec{J}_i$ – mass diffusion of the *i*-th component.

The equations (1)-(4) are usually closed by adding proper differential equations of the turbulence model, semi-empirical dependences for a pressure tensor, heat and diffusion flows, and the equation of ideal gases mixture state. The dependence of thermodynamic, thermal and physical properties of gases on the temperature is considered via approximation formulas.

The selection of a hydrodynamic turbulence model significantly influences the estimated properties of a gas turbine combustor mathematical model. The equations of a RNG $k-\varepsilon$ turbulence model are obtained from non-steady Navier-Stokes' equations with using a mathematical method [10, 19, 20]. They include an additional equation for calculating turbulent viscosity and the constant values differ from the ones which are used in a standard $k-\varepsilon$ model.

The issue of applying the developed model of a gas turbine combustion for studying physical and chemical processes in various fuel combusting devices is very urgent. In order to check adequacy of mathematical and physical models their verification was done using experimental data of diffusion combustors with steam injection [20]. Three different kinetic mechanisms (Table 1) were included to the mathematical model during numerical experiments.

Table 1

Methane oxidation mechanism

Mechanism	Reaction	*E*, J/mole
1	$CH_4 + 1{,}5O_2 \rightarrow CO + 2H_2O$	2e+08/1.2e+08
	$CO + 0{,}5O_2 \rightarrow CO_2$	1.7e+08
2	$CH_4 + 1{,}5O_2 \rightarrow CO + 2H_2O$	1.17e+08
	$CO + 0{,}5O_2 \rightarrow CO_2$	7.68e+07
	$CO_2 \rightarrow CO + 0{,}5O_2$	1.31e+08
3	$CH_4 + 1{,}5O_2 \rightarrow CO + 2H_2O$	125520
	$CO + 0{,}5O_2 \rightarrow CO_2$	167360

The calculations were performed for a combustor of a stationary GTE with eco-friendly and energy steam injection for nine modes of operation (Table 2). Relation of the flows eco-friendly and energy steam varied from the mode to the mode.

Table 2

Operation modes of a stationary GTE with steam injection

Mode	Relative power $\overline{N}_e$	Eco-friendly steam flow G_{ec} , kg/s	Energy steam flow G_{en}, kg/s
1	0.25	0	0
2	0.265	0	2.130
3	0.425	0	2.869
4	0.488	0	3.141
5	0.487	0.558	2.566
6	0.589	0.703	2.819
7	0.74	0.828	3.152
8	0.866	0.858	3.563
9	0.868	0.774	3.606

According to the results of numerical and full-scale experiments the diagrams of dependences of output nitrogen oxides concentrations (Fig. 1) were built as the function of the relative engine power $\overline{N}_e$ [20].

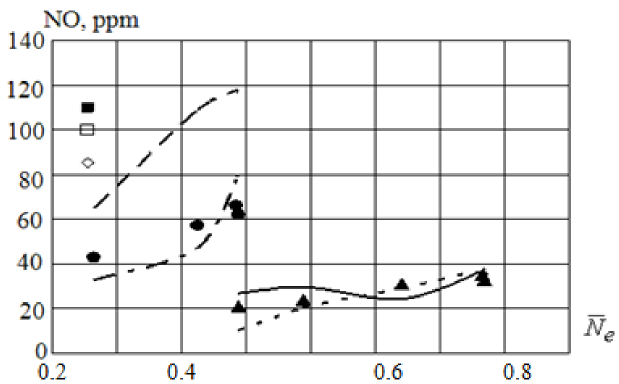


Fig. 1. Comparison of the calculated and experimental emission of nitrogen oxides NO: ■ – without steam supply (experiment);● – with energy steam supply (experiment); ▲ – with supply of energy and eco-friendly steam (experiment); □ – without steam supply (calculation, mechanism 1); ▬ ‐ ‐ ▬ – with energy steam supply (calculation, mechanism 1); ▬▬ – with supply of energy and eco-friendly steam (calculation, mechanism 1); ◊ – without steam supply (calculation, mechanism 2); ‐ ‐ ‐ – with energy steam supply (calculation, mechanism 2); ‐‐‐‐ – with supply of energy and eco-friendly steam (calculation, mechanism 2)

The dependences of the output concentration of carbon oxide CO on the ratio of total steam flow G_{ec}+ G_{en} to fuel flow G_{fuel} (Fig. 2) were also investigated. The dependences of the concentration CO at using kinetic mechanism 1 were not shown here, as their calculated values in the output section of the combustor in this case did not exceed 1 mg/nm^3. Kinetic mechanism 2 determined the difference between the experimental and calculated data only for the modes with energy steam supply when their maximum deviation for the modes with additionally supply of eco-friendly steam did not exceed 10 mg/nm^3. It proves the validity of the used mathematical model.

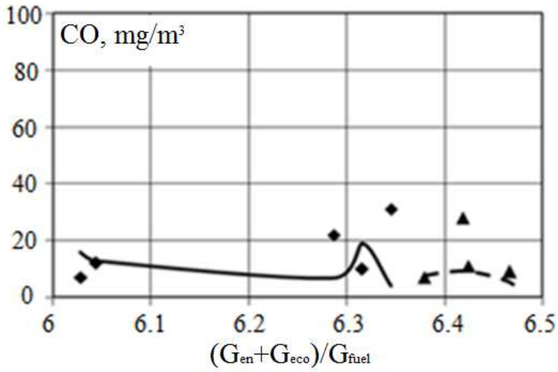
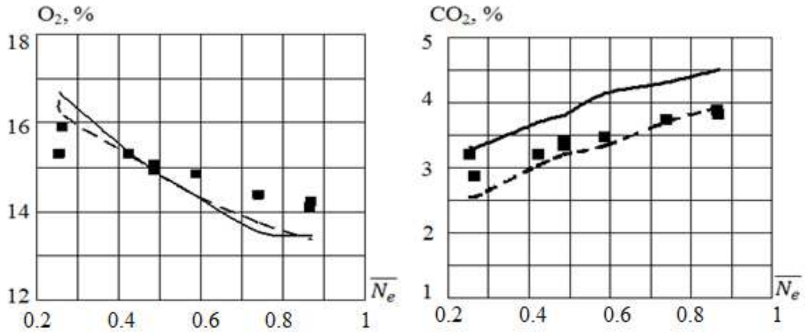


Fig. 2. Comparison of the calculated and experimental values of the concentration of carbon oxide CO on the relative steam flow: ◆ – with energy steam supply (experiment); ▲ – with energy and eco-friendly steam supply (experiment); ▬▬ – with energy steam supply (calculation, mechanism 2); ---- – with energy and eco-friendly steam supply (calculation, mechanism 2)

Dependences of the molecular oxygen and carbon dioxide concentrations on the relative engine power are shown in Fig. 3,**a**,**b**. The character of the calculated curves for the two considered methane oxidation mechanisms almost coincides and average differences with experimental data considering 1.5% of air supply for turbine blades cooling are 3.17% for O_2 and 6% for CO_2 (mechanism 2) and 3.5% and 14.2% for O_2 and CO_2 respectively (mechanism 1).

To provide qualitative comparison of the special features of the kinetic schemes used in the calculation, the zones of the most active CO formation for mode 9 are shown in Fig. 4,**a**,**b**. It is obvious that in case of a three-reaction mechanism this zone is the longest and distributes down the flow in the combustion liner. The same contour is typical for other modes.



a b

Fig. 3. Comparison of the calculated and experimental values of the concentrations O_2 (**a**) and CO_2 (**b**) in the exhaust gases: ■ – experiment; ━━ – calculation, mechanism 1; – – – – calculation, mechanism 2

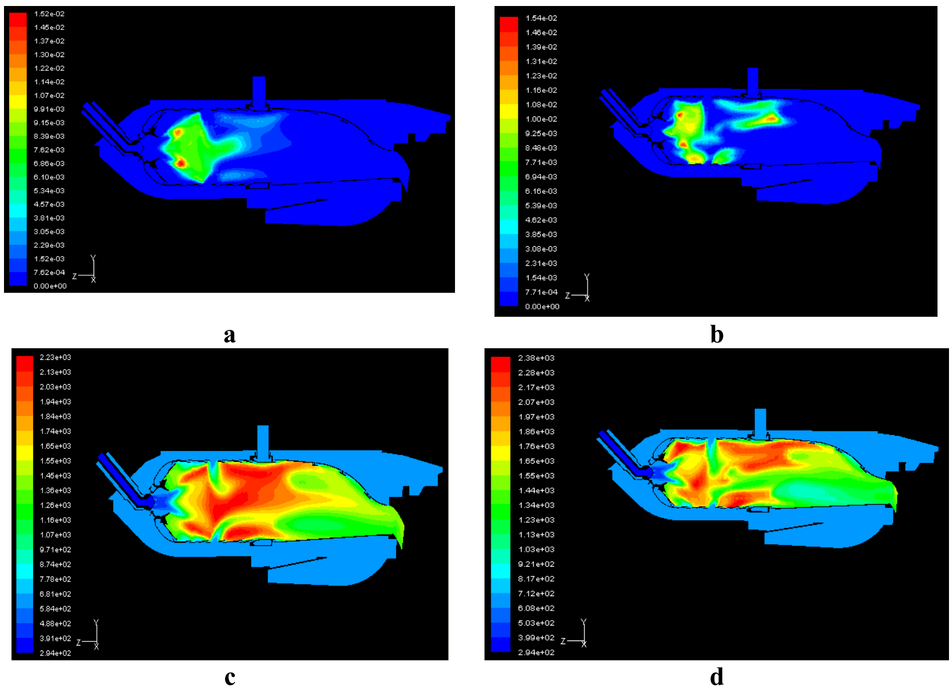


Fig. 4. Calculated distributions of the combustor parameters: **a** – CO concentration, mechanism 1; **b** – CO concentration, mechanism 2; **c** – temperature field, mechanism 1; **d** – temperature field, mechanism 2

Figures 4,**c**,**d** show the temperature distribution in the combustor. The levels of maximum temperatures do not differ from the used kinetic schemes. However, for a three-reaction mechanism the temperature field in the combustion liner has significant nonuniformity, and more corresponds to the real situation in the combustion liner.

At using modern ways of computational fluid dynamics (CFD) determination of sulfur compounds distribution in the combustor sections shall be performed simultaneously with defining the primary parameters of a fuelburn device: pressures, velocities, temperatures, component concentrations. Thus, there shall be the algorithm of transfer equations solution for the following compounds: H_2S, SO_2, SO, SH, SO_3.

The model of sulfur oxides SOx formation is included 3 basic mechanisms:

- Mechanism of formation from sulfur which is in liquid fuel. To simply, we can assume that all sulfur compounds in fuel may be conventionally replaced by hydrogen sulfide H_2S [16].
- Reactions of sulfur compounds in the gaseous phase. For combustion processes of lean air-fuel mixtures the leading components are SO, SO_2, SO_3. At mixture enriching with fuel the formation of considerable amounts of H_2S, SH is possible. In a number of studies, for example in [17], it is supposed that the balance condition of sulfur compounds is in the gaseous phase.
- Absorption of sulfur compounds with solid particles.

In a mathematical model of a combustor the transfer equations for sulfur compounds are written similarly to the transfer equations for the chemical components of the oxidation scheme considering convection, diffusion, formation and decomposition of appropriate compounds.

Thus, additional transport equations have the following form:

$$\frac{\partial}{\partial t}\left(\rho Y_{SO_2}\right)+\nabla\left(\rho\vec{\upsilon}Y_{SO_2}\right)=\nabla(\rho D\nabla Y_{SO_2})+S_{SO_2}\,, \tag{5}$$

$$\frac{\partial}{\partial t}\left(\rho Y_{H_2S}\right)+\nabla\left(\rho\vec{\upsilon}Y_{H_2S_2}\right)=\nabla(\rho D\nabla Y_{H_2S})+S_{H_2S}\,, \tag{6}$$

$$\frac{\partial}{\partial t}\left(\rho Y_{SO_3}\right)+\nabla\left(\rho\vec{\upsilon}Y_{SO_3}\right)=\nabla(\rho D\nabla Y_{SO_3})+S_{SO_3}\,, \tag{7}$$

$$\frac{\partial}{\partial t}\left(\rho Y_{SO_2}\right)+\nabla\left(\rho\vec{\upsilon}Y_{SO}\right)=\nabla(\rho D\nabla Y_{SO})+S_{SO}\,, \tag{8}$$

$$\frac{\partial}{\partial t}\left(\rho Y_{SH}\right)+\nabla\left(\rho\vec{\upsilon}Y_{SH}\right)=\nabla(\rho D\nabla Y_{SH})+S_{SH}\,, \tag{9}$$

where:

Y_{SO_2} , Y_{H_2S} , Y_{SO_3} , Y_{SO} , Y_{SH} – the mass fractions of SO_2, H_2S, SO_3, SO, SH in the gaseous phase,

S_{SO_2} , S_{H_2S} , S_{SO_3} , S_{SO} , S_{SH} – source terms for corresponding compounds.

Simultaneous solution of the differential equations (1)-(9) allows predicting distribution of sulfur compounds in the volume of a gas turbine combustor with steam injection.

The detailed multi-stage schemes of sulfur compounds formation and decomposition in the full-scale designs of gas turbine combustors cannot be nowadays completely implemented into practice, so a number of simplified mechanisms is offered. They allow considering the formation of sulfur compounds at performing three-dimension calculations. For practical applications an eight-reaction kinetic mechanism [7] (Table 3) can be used.

The units of measure for the coefficients in Table 3 are the following: E, J/gmole; A, $m^3/(gmole\cdot s)$ and $m^6/(gmole^2\cdot s)$ - for reactions 13 and 15.

In addition to the reactions of Table 3 it is necessary to consider additional reactions with SO_3 [5].

Table 3

Mechanism of sulfur compounds formation

$k = AT^b \exp(-E/RT)$

	Reaction	A	b	E
1	$H_2S + H \rightarrow SH + H_2$	1.819702e+07	0	7.484300e+03
2	$SH + H_2 \rightarrow H_2S + H$	9.375623e+06	0	6.253660e+04
3	$OH + H_2S \rightarrow H_2O + SH$	1.380385e+02	0	3.742150e+03
4	$H_2O + SH \rightarrow OH + H_2S$	3.104557e+07	0	1.218543e+05
5	$SO + OH \rightarrow H + SO_2$	1.621810e+08	0	2.565926e+03
6	$H + SO_2 \rightarrow SO + OH$	7.691299e+09	0	1.187023e+05
7	$SH + O \rightarrow SO + H$	3.548135e+08	0	2.687316e+03
8	$SO + H \rightarrow SH + O$	2.985385e+09	0	1.694600e+05
9	$O + H_2S \rightarrow SH + OH$	4.365162e+03	0	1.380493e+04
10	$SH + OH \rightarrow O + H_2S$	9.885528e+08	0	1.380493e+04
11	$SO + O_2 \rightarrow SO_2 + O$	4.466832e+05	0	2.703222e+04
12	$SO_2 + O \rightarrow SO + O_2$	1.663412e+06	0	7.613643e+04
13	$H + SH + M \rightarrow H_2S + M$	1.096478e+03	0	0.000000e+00
14	$H_2S + M \rightarrow H + SH + M$	8.669613e+14	0	3.819463e+05
15	$SO + O + M \rightarrow SO_2 + M$	8.709647e+09	-1.8	0.000000e+00
16	$SO_2 + M \rightarrow SO + O + M$	1.905464e+14	0	0.000000e+00

During numerical calculations for studying the mechanisms of sulfur oxides formation and decomposition, the aerodynamic structure of the flow in the combustor was investigated for two cases: 1) without air bleeding for turbine blades cooling; 2) with air bleeding.

Figure 5 shows the diagram of the gas turbine combustor with eco-friendly and energy steam supply:

- in_air – section of air supply to the combustor diffuser from the high-pressure compressor;
- in_par_vxod – section of air or steam supply to the combustor diffuser from the high-pressure compressor or a steam generator. It should be mentioned that if steam is not supplied to the diffuser of the combustor, the total air flow via the combustor diffuser is equal to the air flow via the sections in_air and in_par_vxod. Steam supply via the input diffuser is provided for studying various schemes of steam distribution in the combustor;
- in_ek_par – section for eco-friendly steam supply via the nozzle. Eco-friendly steam is aimed at reducing formation of nitrogen oxides in the primary combustor zone;
- in_enegr_par – section of energy steam supply to the combustion liner. Energy steam is aimed at forcing gas turbine unit power and generated in a heat-recovery boiler installed behind the engine;
- out_1_1…out_10_5 – output sections forming the outlet from the combustion liner;
- out_stock_1…out_stock_3, out_verx – sections of air bleeding for turbine blades cooling.

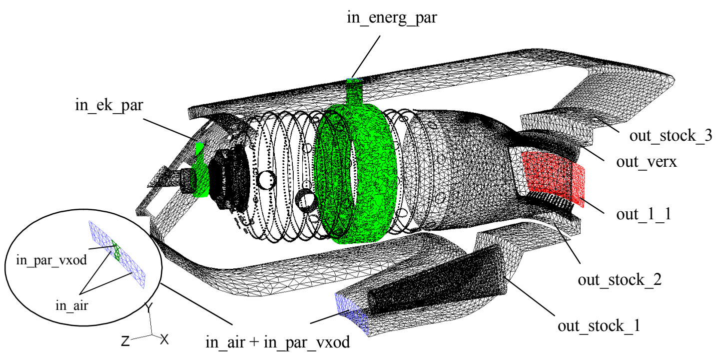


Fig. 5. Diagram of input and output sections of the combustor with eco-friendly and energy steam supply

In the calculations the ratio of the flows of eco-friendly steam and fuel was set as 1.1, and the ratio of the total steam flow to the hydrocarbon fuel flow was 6.5.

For detailed carbon oxide CO and hydrogen H_2 formation modeling the following five-stage reaction mechanism of hydrocarbon combustion was used:

$$2C_{16}H_{29} + 16O_2 \rightarrow 32CO + 29H_2;\ 2H_2 + O_2 \rightarrow 2H_2O;$$
$$2CO + O_2 \rightarrow 2CO_2;\ 2H_2O \rightarrow 2H_2 + O_2;\ 2CO_2 \rightarrow 2CO + O_2.$$

3. Results and discusion

The main parameters distributions in the gas turbine combustor with steam injection are calculated. The contours of the velocity magnitude in the longitudinal section of the combustor are shown in Fig. 6, and the velocity vectors in the head and output parts of the combustion liner are presented in Fig. 7.

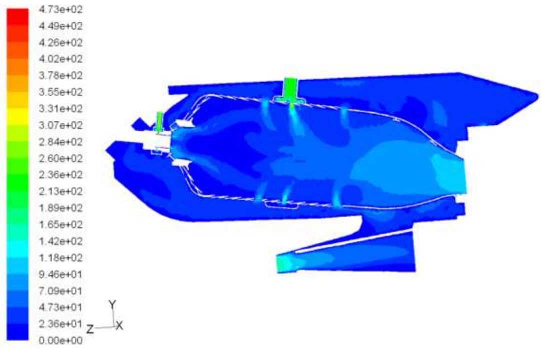


Fig. 6. Contours of the velocity magnitude in the longitudinal combustor section

Specific features of the combustor working process are the following. At fuel injection there is intense heat emission which is attended with increase of the axial component of the working medium velocity and decrease of the tangential component. Gas heating leads to expanding of the flow and local pressure raise. As due to centrifugal action the pressure of the working medium in the periphery part of the combustor is higher than near the axis, the flow expansion takes place in the direction to the combustor axis. The radial velocity component grows, and the dimensions of the reverse flows zone decrease as well as the amount of gases which return to the flame root. While taking away from the primary part of the combustion liner due to friction and turbulent diffusion the intensity of swirling flow sharply drops, the recirculation zone is closed. The sections of the combustion liner are filled with intermixing flows of the combustion products and cooling air which move mainly in the axial direction.

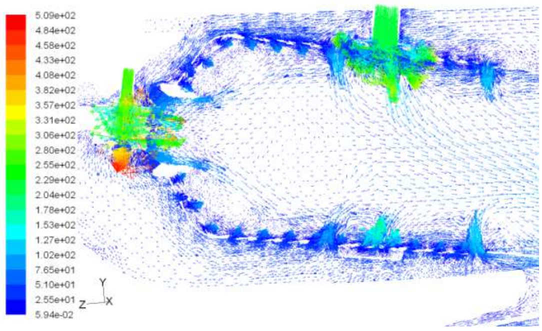


Fig. 7. Distribution of the velocity vectors in the combustor

Comparison of the total pressure losses in the combustor in the cold and hot condition modes of operation is shown in Fig. 8. The increase of the total pressure losses is seen approximately by 0.35% (absolute), which is connected with heat supply in the combustor during fuel burning. The calculated value of total pressure losses is 5.89% for the nominal mode of operation of the combustor with steam injection.

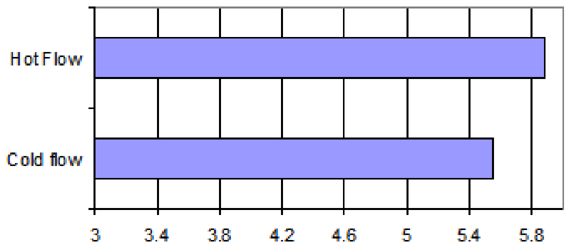


Fig. 8. Comparison of the total pressure losses in the cold and hot condition modes of operation

Modeling of sulfur compounds distribution at hydrocarbon fuel combustion in the gas turbine combustor with steam injection is performed taking into consideration convection, diffusion, formation and decomposition of proper sulfur components.

Figure 9,**a-d**, show the distributions of mass fractions of the main sulfur containing compounds SO_2, H_2S, SH, SO in the longitudinal section of the combustor with steam injection at sulfur mass content of 1% in hydrocarbon fuel.

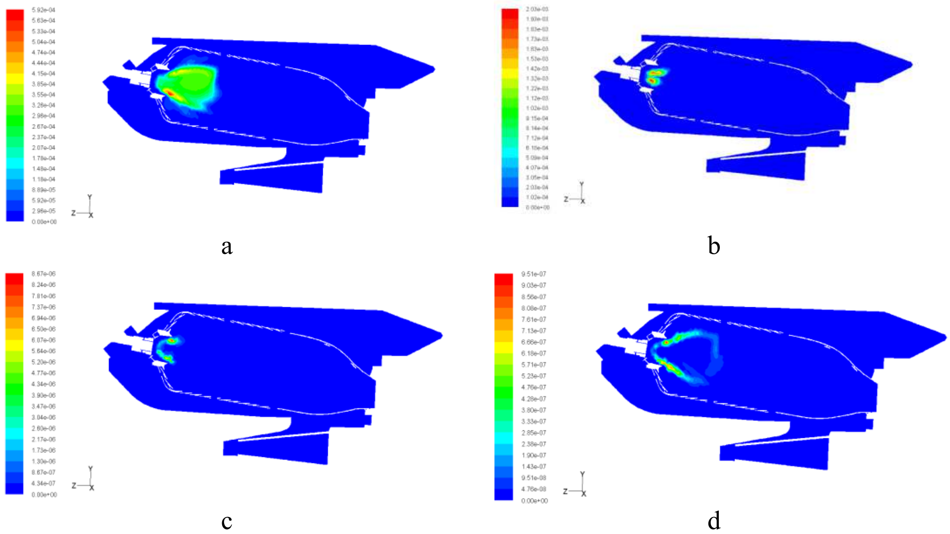
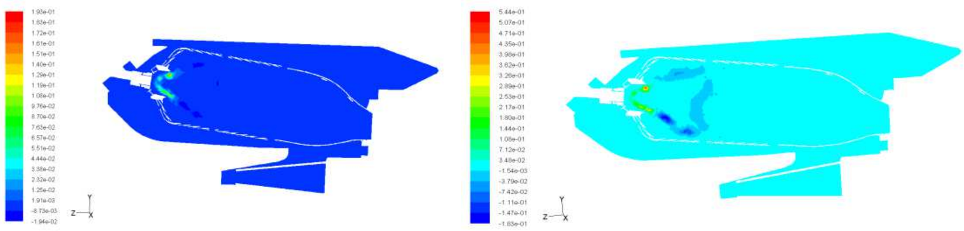


Fig. 9. Contours of mass fractions of SO_2 (**a**), H_2S (**b**), SH (**c**), SO (**d**) in the longitudinal section of the combustor with steam injection at sulfur mass content of 1% in fuel

Figure 10 shows the zones in the gas turbine combustor where there is intense formation of sulfur dioxide SO_2 for sulfur mass content of 1 and 3% in hydrocarbon fuel.

Table 4 gives the values of integral mass and volume fractions of the main sulfur containing compounds in the output section of the gas turbine combustor with steam injection at using hydrocarbon fuel which contains 1 and 3% of sulfur per mass respectively.



a b

Fig. 10. Level of sulfur dioxide formation in the combustor: **a** –sulfur mass content of 1% in fuel; **b** – sulfur mass content of 3% in fuel

Table 4

Content of sulfur compound in the combustion output section

	S = 1%		S = 3%	
	Mass fraction	Mole fraction	Mass fraction	Mole fraction
SO_2	5.4151e-07	2.4417e-07	3.4469e-06	1.5542e-06
H_2S	1.1913e-13	1.0096e-13	5.4498e-14	4.6190e-14
SH	1.2260e-14	1.0707e-14	1.16238e-14	1.0153e-14
SO	4.5389e-11	2.7283e-11	2.2663e-10	1.3622e-10

4. Summary

1. The developed mathematical model of three-dimensional chemically reactive flows enables to forecast exit thermal and ecological characteristics of a gas turbine combustor with steam injection. The results of the model verification confirm its validity for the wide spectrum of operating modes. Specific features of working process of the combustor with eco-friendly and energy steam supply are determined. An eight-reaction kinetic mechanism of sulfur compounds formation and decomposition is used for 3D calculation of the full-scale design of gas turbine combustor.
2. Theoretical investigations of sulfur oxides formation and decomposition in gas turbine combustors with steam injection at sulfur mass content of 1 and 3% in hydrocarbon fuel are performed. The highest concentrations of sulfur dioxide are observed near the frontline of a combustor.
3. Obtained results allowed improve and optimize design of present and prospective gas turbine combustor with steam injection.

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