Prediction of the chemical composition of combustion products in metallurgical reheating furnaces by use of numerical methods

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In this paper the results of numerical calculations of chemical composition of flue gases, produced by combustion of natural gas, were presented. Calculations were conducted for metallurgical pusher furnace, working in one of the steelworks located in Poland. The chemistry of combustion process was modeled using the program CHEMKIN-PRO. Simulations of the chemical composition of flue gas, in various zones of the furnace, were carried out based on the measurement data collected from the real object.

Keywords: Numerical modeling - Pollution - Pusher furnace - Combustion

INTRODUCTION

Fuel combustion is a basic way to obtain both primary energy and the main source of emission, such as sulphur oxides, nitrogen oxides, carbon monoxide and particulates. The dominant contribution of total major pollutants emission in Poland falls on the power industry as well as manufacturing industry, including metallurgy. The GUS (Central Statistical Office - in polish GUS) data shows that in 2009 the total emissions of major air pollutants was amounted to 861.000 tons of sulphur dioxide, 820.000 tons of nitrogen oxides and 394.000 tons of particulate matter. The largest emission of the major gaseous pollutants such as SO_2 and NO_x emissions is from combustion processes, in particular in the production and transformation of energy (449.120 tons of SO_2, 271.970 tons of NO_x), in the municipal and housing sector (241.520 tons of SO_2 and 86.520 tons NOx) and in industry (165.490 tons of SO_2 and 97.940 tons of NO_x).
Particularly noteworthy is the emission of carbon dioxide, the main greenhouse gas, and nitrogen oxide emissions associated with the processes taking place in the high-temperature reheating furnaces. Therefore, and due to increasingly stringent standards for environmental protection there is a need to reduce the pollution for example by appropriate optimization of combustion processes [1].

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The numerical methods, due to the complex nature of the issues related to the processes of fuel combustion, became an essential component of research. They are extraordinarily useful tool in the design process of burners and development of special, low emission combustion techniques. One of the many areas in which advanced numerical simulations are used to analyze the flow phenomena is aviation technology and the automobile industry. In recent years, numerical modeling is also successfully used in the analysis of combustion processes, especially in the prediction and reduction of emissions generated in these processes [2÷8]. Numerical modeling is of key importance wherever the implementation of modern methods to optimize operation of heating devices and manufacturing processes, as well as minimize the pollution, are associated with high financial expenditures (partial elimination of the costs incurred for experimental studies). The complexity of the issues of combustion processes (chemical process) and dynamic of flue gas flow puts numerical modeling at a level that often the experimental studies do not match [4, 9, 10].
The most common programs used for modeling of chemical and gas dynamic phenomena include: FLUENT, CHEMKIN, KIVA, COMSOL, FLUE GAS, HSC CHEMISTRY, CANTERA, KINALC, and others [4, 7, 9, 11]. An example of application of CHEMKIN software, described in this article, is numerical analysis of the chemical composition of combustion products generated by the combustion of natural gas in a pusher furnace. In numerical modeling of combustion processes, various types of mechanisms and chemical models are used. The most widespread and most frequently used mechanisms include: the mechanism of Miller - Bowman, GRI-Mech, Burcat's, Konovv's, Berkley's mechanism, and others [12, 13, 14].

EMISSIONS OF POLLUTANTS IN METALLURGICAL INDUSTRY

Polish metallurgy discharges into the atmosphere about 1,9% of total national emissions of NO_x, SO_2, CO and 4,5% of CO_2, of which 98.1% comes from the steel industry [15]. In polish steelworks, the dominant type furnaces are pusher furnaces for the charge heating in hot rolling mills, usually a mixture of coke-oven and blast furnace gas-fired, coke oven gas-fired as well as natural gas-fired [16÷19].
One of the problems associated with the combustion fuels in the high-temperature reheating furnaces is the emission of NO_x. In Table 1, the emission of air pollutants, including NO_x from hot rolling mills in Poland, was shown. These values were compared with the values contained in the reference document [17].
As can be seen from the data presented in Table 1, the emission values obtained in the Polish hot rolling mills are in the range specified in the reference document.
The folowing conditions should be considered in order to not exceed the imposed limitation:

- the use of a methane-rich natural gas,
- low-emission burners,
- the use of a recuperator for heat recovery from the flue gas,
- automatic control of the furnace,
- loading with minimal opening and minimal size of the charge window, as well as limited to a minimum time of opening of the charge windows.

These actions affect the optimization of gas consumption, reduction of excess air and heat losses in the furnace, which results in lower NO_x emissions [17].
The largest share of carbon dioxide emissions have blast furnaces (69%). Subsequently are sintering (13%), steelworks (11%), and finally coke furnaces (7%) [15].
Over the past 20 years, the steel industry has made significant reductions in carbon dioxide emissions, actively engaging in ecological actions. At the moment, many steel plants, based on innovative technical and technological solutions, work at the level close to the lowest achievable emission. Unfortunately, there are still steelmaking plants, which reduce the global steel industry results in the scope of greenhouse gases emission. Therefore, efforts of the steel industry, leading to improvement the environmental state, should focus on the transfer of effective technology in order to accelerate the replacement of obsolete objects. A promising direction, which aims to reduce the CO_2 emissions, is a cooperation of steel works with the scientific-research centers [20, 21]. The results of this cooperation are reflected in research projects, expertise, opinions, as well as numerous publications [9, 21÷26].

MODELLING OF COMBUSTION PROCESSES IN INDUSTRIAL REHEATING FURNACES

The results of own research and the current state of knowledge in the scope of the use of numerical methods in combustion processes, show that the major concern of researchers is to develop a model that would in the best extent represent the real conditions. The problem is connected with the correct formulation of the initial conditions. i.e. providing the data describing the process analysed [4, 27, 28]. Depending on the model scale, different processes are considered or ignored, which affects the type of the equation describing the phenomenon. Due to insufficient amount of input data in the model, different sets of simplifying assumptions are introduced. This is how the new models are created and each of them describes the considered process in a different way. Furthermore, a major difficulty, in creating an accurate model, are the problems associated with discrepancies in a reaction rate constant. Therefore, in order to confirm the correctness of the model adopted in the calculations, verifying the results of the calculations with the experimental studies is necessary. Otherwise, the results of model calculations are only hypothetical [28]. Due to the above, a concept of 'perfect' model' does not

Parameter	The values contained in the reference document	The values obtained in national hot rolling-mills
Particular matter from pusher furnaces	200 – 1400 g/Mg	0,7 – 21 g/Mg / (1,4 – 10 mg/Nm^3)
NO_x	80 – 360 g/Mg (200 – 700 mg/Nm^3)	20 – 464 g/Mg / (100–377 mg/Nm^3)
NO_x from conventional burners	2 – 600 g/ Mg	36 – 464 g/Mg
NO_x from conventional burners	2 – 600 g/ Mg	36 – 464 g/Mg
burners with low-emission of NO_x	150 – 500 g/ Mg	new installations about 95 g/Mg
NO_x from regenerative burners	1000 – 4000 g/ Mg (250 – 900 mg/Nm^3)	
SO_2	0,3 – 600 [0,02 – 900] g/Mg (0,6 – 1300 mg/Nm^3 400 – 800 mg/Nm^3)	0,2 – 19 g/Mg / (7,0 – 8,2 mg/Nm^3)
CO	5 – 850 g/Mg (100 – 170 mg/Nm^3)	5,2 - 65 g/Mg/ (23 – 34 mg/Nm^3)

Tab. 1 - Comparison of emissions into the air from reheating furnaces and furnaces for heat treatment in hot rolling mills working in Poland as well as in countries of European Union [17].

Tab. 1- Confronto tra le emissioni in atmosfera dei forni di riscaldo e dei forni di trattamento termico nei laminatoi a caldo, in Polonia e l'Unione europea [17].

exist. Also, there is no the optimal number of chemical reactions and compounds describing the considered process. For methane, the most widespread gas fuel, a mechanism of Miller - Bowman (MB) is the most commonly used scheme of the reaction of CH_4 oxidation [14]. It is known that not all reactions are equally important. Depending on the conditions under which methane oxidation takes place, namely: pressure, temperature, as well as quantity and composition of the oxidant, some reactions are dominant and the others could be omitted because their influence on the process is small. Literary sources available in this issue provide different mechanisms in which the amount of reactions occurs more than 2,500 and chemical compounds to more than 500. However, due to computational limitations of computer programs, simplified models are used. An example is the Fluent program, one of the most commonly used (widespread) computer code in the calculations of the combustion process. Bringing the chemistry of combustion process of methane to six compounds, namely, CH_4, O_2, CO_2, CO, H_2O, H_2, N_2, NO, NO_2 and 9 chemical reaction, leads to large discrepancies of calculation results and measurements [29].

Such discrepancies do not exist in the case of using Chemkin software. This program, due to the extensive chemical database is a base for the mechanism of formation and reduction of harmful substances in the combustion chambers. An example of appliance of the Chemkin package in complex chemical calculations of combustion process taking place in pusher furnaces, are studies performed by Niska, J., and others [29].

Laboratory experiment

The researches were conducted in a cylindrical experimental chamber, which was the model of a pusher furnace, built of 12 concrete rings (BOS 145-II) with an internal diameter of 0.34 m and total length of 3.12 m (Fig. 2 and 3). The chamber was insulated with 0.05 m layer of mineral wool, and protected with armor of aluminum sheet. The main burner was positioned in the initial segment of chamber, and the composition of burned gas was as follow: CH_4 = 96.7%, C_2H_6 = 1.785%, N_2 = 1.2%, CO_2 = 0.315 %. At the length of chamber, measuring holes were done, in order to measure the concentration of selected combustion products. Flue gas composition was measured using the analyzer TESTO 360. The temperatures were measured by aspiration thermocouple PtRh-Pt. The amount of provided media were measured by flow rotameters.

During the experiment the following values were determined:

- Excess air factor λ = 1.07,
- Gas flux = 2.835 m^3 / h,
- Air flow = 29.00 m^3 / h.

The mathematical model

Numerical simulations of the chemical composition of flue gas were performed using the CHEMKIN-PRO program. For the calculation a model examining the processes of combustion occurring in the so-called 'Perfectly Stirred Reactor' (PSR) was adopted. Chemical kinetics for expanded mechanism of methane combustion was based on the mechanisms of GRI-Mech 3.0 (Gas Research Institute – Mechanism) and the Miller-Bowman. The final modified chemical mechanism included 132 reactions and 36 chemical compounds as well as chemical elements, i.e.: H_2, H, O, O_2, OH, HO_2, H_2O_2, H_2O, N, N_2, NO, NH, NH_2, NH_3, HNO, NO_2, N_2O, CH_4, CO_2, CO, CH_3, CH_2, CH, HCO, C, C_2H_4, C_2H_3, C_2H_5, C_2H_6, CN, HCNO, HCN, HOCN, HNCO, NCO.

The initial stage of the calculation procedure was the boundary conditions formulation, and consequently the 'input file' preparation, based on data of experimental research.

Simplified diagram of the research course is shown in Fig. 4.

The calculation of the experimental chamber

Input data of modeling procedures of an experimental chamber is presented in Table 2.

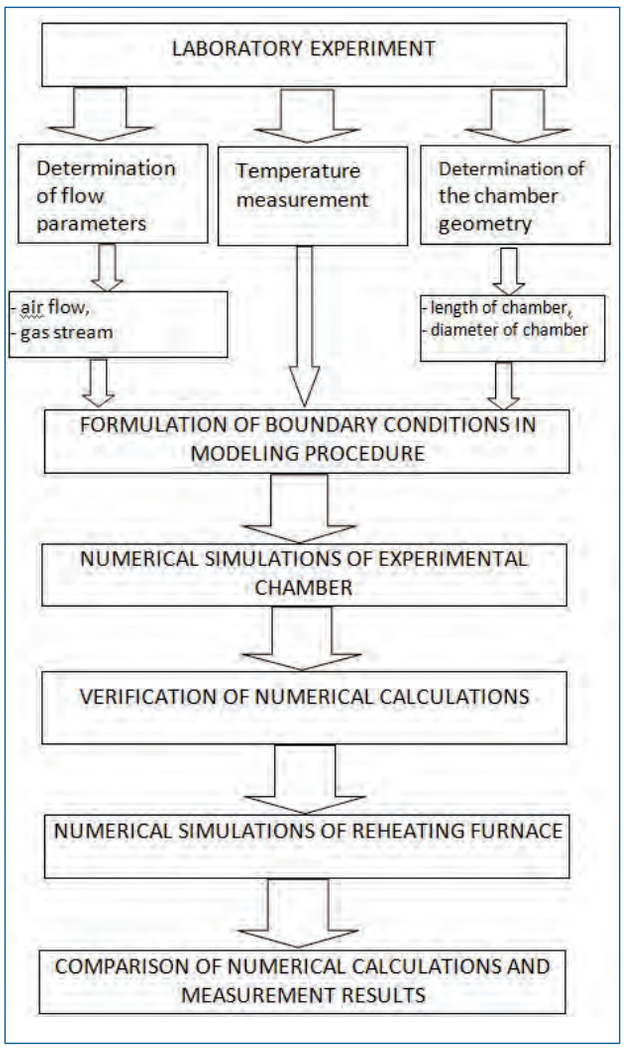

Fig.1 - The scope of conducted research.

Fig. 1 - Schema della ricerca condotta.

Model research of the pusher furnace

The results of experimental and modeling research conducted in the experimental chamber was extended to model on the real 'object', i.e. a pusher furnace working in steelworks located in Polish territory. The furnace was

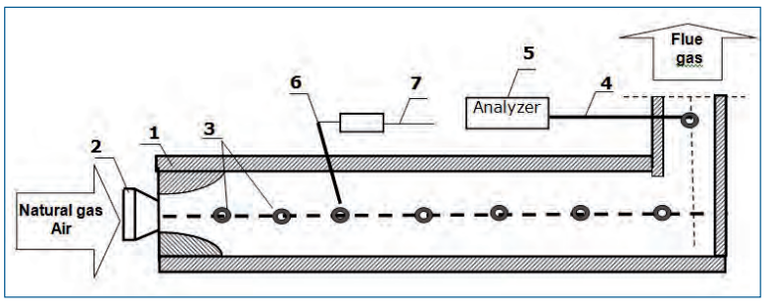

Fig. 2 - The scheme of experimental stand: 1 –chamber, 2 – turbulent burner, 3 - measuring holes, 4 - probe, 5 - flue gas analyzer TESTO 360 , 6 - PtRh-Pt thermocouple, 7 – measuring card.

Fig. 2 - Schema dell'impianto sperimentale: 1 -camera di combustione, 2 - bruciatore a turbolenza, 3 - fori di misurazione, 4 - sonda, 5 - analizzatori TESTO 360, 6 - termocoppia PtRh-Pt, 7 - scheda di misurazione.

divided into six contractual regulating zones (Fig. 5).
The formulation of boundary conditions in the modeling process requires the assembly of the following input data:

- geometrical dimensions of the furnace chamber, dimensions of the regulatory zone of the furnace (Table 3)
- gas and air volume fluxes provided to individual zones (Table 4),
- the average composition of natural gas supplied to the steelworks: CH_4 = 94%, C_2H_6 = 1,5%, CO_2 = 0,2%, O_2 = 0,1%, N_2 = 4,2%,
- the temperature in each zone (Table 4),
- temperature of air supplied for combustion: 350° C,
- pressure in the furnace - 7 Pa.

Analyzed pusher furnace is divided into six regulatory zones (contractual). Due to the fact that in this type of furnaces, three main areas have occurred, i.e. preheating, heating and balancing zone, in this paper the regulatory zones have been limited to four, namely:
- zones I and II (preheating zone)
- the combined zones III and IV (heating zone)
- the combined zones V and VI (balancing zone).

The differences of temperature and the excess air ratio, occurring between V and VI zone, are due to the fact that in the area VI, the load of the charge and opening the charging door take place (table 4).
Furthermore, an additional argument in favor of a combination of zones III, IV as well as V and VI is their location.
Due to the complexity of the analyzed issues in the calculation, some simplifications were adopted namely:

- he number of burners were limited to one, assuming that the fluxes of air and fuel supplied to chamber were equal the fluxes supplied into the entire zone,
- one type of burner was adopted,
- in numerical calculations residence time of reactants, only at the end of each zone, was determined and implemented (according with the direction of gas flow).

The chemical composition of combustion products were calculated, assuming the amount of gas moving in a

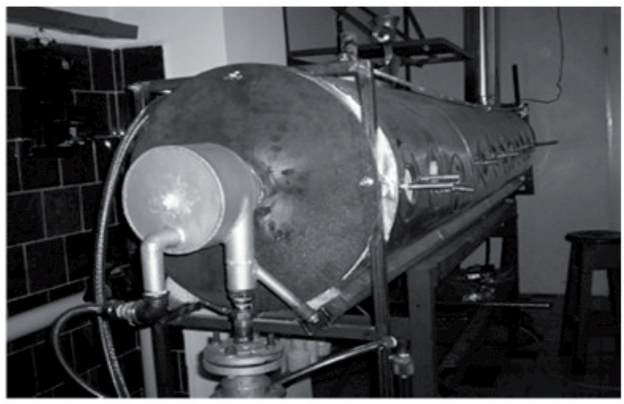

Fig. 3 - View of the chamber from the burner side.

Fig. 3 -.Vista della camera dal lato bruciatore.

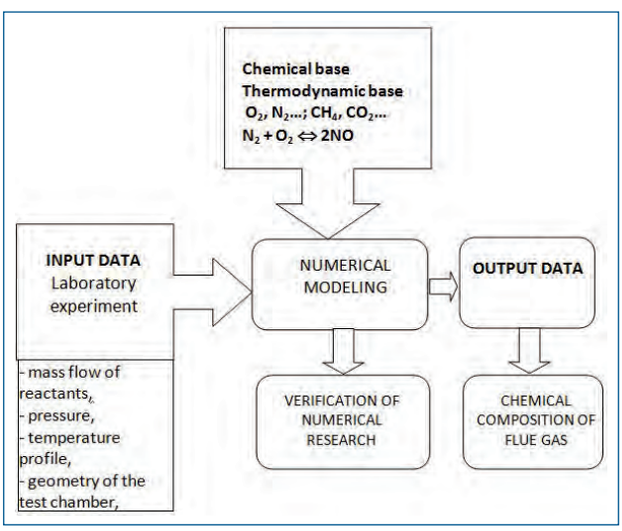

Fig. 4 - Background modeling.

Fig. 4 - Schema della modellazione.

countercurrent direction to the feedstock, namely:

- Zone I – flue gas of zone I,
- Zone II - flue gas of zone I and II,
- Zone III + IV - flue gas of zone I, II, III, IV,
- Zone V & VI - flue of zone I, II, III, IV, V, VI (at the outlet of the furnace).

The input data file included data collected in Table 5.

RESULTS

The results of calculations of flue gas composition in the experimental chamber.

The distribution of selected products of combustion, i.e. nitrogen oxides on the length of the chamber, was based on the temperature profile obtained by experiment (Fig. 6).
The comparative analysis of the results experimentally obtained with using numerical methods shows that the model (PSR) adopted in the calculation and the chemical mechanism is correct, because of the high consistency of

Mass flow of air - fuel mixture, kg / s	0.011
Molar fraction reactant	CH_4 = 0.0861 C_2H_6 = 0.0016 CO_2=0.0003 N_2=0.721 O_2=0.191
The temperature of input media, K	293
Calculated combustion temperature, K	1860
Pressure, atm	1.0
Residence time, s	0.01

Tab. 2 - Input data of modeling procedures of an experimental chamber.

Tab. 2 - Dati di ingresso per le procedure di modellazione di una camera sperimentale.

Zone	Length, m	Width, m	Height, m
I	4.80	7.8	1.25
II	4.10	7.8	1.25
III	8.10	7.8	1.25
IV	6.60	7.8	1.25
V	12.60	7.8	2.05
VI	14.10	7.8	2.05

Tab. 3 -. The dimensions of regulating zones adopted in calculations.

Tab. 3 - Dimensioni delle zone di regolazione adottate per i calcoli.

Zone Parameter	I	II	III	IV	V	VI
Type-power of burner kW	WSG – 1600 PWG - 400	PWG - 400	PWG- 630	WSG - 1600	PWG - 630	**WSG - 1600**
Amount of burners	2 15	15	30	10	25	**8**
Gas fluxes, mn^3/h	237.03	130.39	701.89	619.25	473.79	**60.91**
Air fluxes, mn^3/h	2150.62	1553.07	6802.90	6007.26	4801.48	**2484.33**
Temperature °C	1251	1269	1281	1277	1075	**1064**
Excess air, λ	**0.98**	**1.30**	**1.054**	**1.055**	**1.095**	**4.47**

where: PWG- type of burner, planar type of burner; WSG- type of burner, vortex burner

Tab. 4 - Specification of amount and power of the burners, as well as the average values of gas and air fluxes, and temperatures of each furnace zones. Burner type: PWG planar; WSG vortex

Tab. 4 - Tipo, quantità e potenza dei bruciatori e valori medi delle correnti di gas, flussi d'aria e temperature in ogni zona del forno. PWG - bruciatore tipo planare; WSG - bruciatore tipo vortice

results. In the case of numerical calculations lower NO_x concentration (138 ppm at 2.3% oxygen content in flue gas) can be observed. That might be related to the condition of perfectly stirring of reactants, resulting from the assumptions, namely the perfectly stirred reactor.
Satisfactory results of the calculations were the basis to apply the considerations on the real object, i.e. a pusher furnace.

RESULTS

The results of calculations for heating furnace

Figure 7 shows the amount of concentration of selected combustion products (O_2, CO_2, CO and NO) for the specified calculation zones. Figure 7 shows that the analyzed concentrations of combustion products depend on conditions of prevailing temperature and flow (excess air

Zone	Mass flow of the mixture kg/s	Mole fraction CH_4	Mole fraction C_2H_6	Mole fraction CO_2	Mole fraction N_2	Mole fraction O_2
I	0.817401	0.096	0.001772	0.0003127	0.712765	0.189
II	0.582251	0.0749	0.001383	0.000244	0.729742	0.194
III	2.576178	0.09044	0.001669	0.0002946	0.717237	0.19
IV	2.274687	0.09038	0.001668	0.0002944	0.717287	0.19
V	1.814009	0.08685	0.001603	0.0002829	0.720125	0.191
VI	0.902224	0.02314	0.000427	0.0000754	0.771382	0.205

Tab. 5 - Specification of selected input data of modeling procedures.

Tab. 5 - Sintesi dei dati di ingresso selezionati per le procedure di modellazione.

factor) in each zone. The highest concentration of carbon monoxide, at the level of 0.65% at 0.22% oxygen in the flue gas, was observed in zone I. That was a result of the air- underflow in this zone (λ = 0.98). In case of nitrogen oxides, the dominant way of their formation was thermal mechanism, which is the result of temperatures above 1250°C prevailing in the zone I, II, III and IV. The NO_x concentrations were in the range of 90 - 113 ppm, while the lowest concentrations was at the outlet of the chamber. Final concentrations of analyzed combustion products at the outlet of furnace were as follows: O_2 = 6, 33%, CO_2 = 6,62%, CO = 12 ppm, NO = 90 ppm.

Figure 8 shows the comparison of practical measurements and numerical calculations of selected products of combustion at the outlet of the furnace.

The comparative analysis shows that obtained results are qualitatively correct, but quantitative are different, and these differences are small and result largely from the simplifications and assumptions adopted to the calculations, in particular the assumption of perfectly stirring condition.

The most significant differences in the measured and calculated concentrations are observed in the case of CO. The calculated concentration of carbon monoxide is about 74% lower than the concentration obtained by the measurements. This discrepancy can be explained by better afterburning of carbon monoxide as a result of perfect stirring of reagents. A similar trend is observed in industrial environments, where by appropriate treatment of the stirring processes intensification (e.g. disturbance of pulsation) leads to a reduction in the concentration of undesirable combustion products such as CO and NO_x.

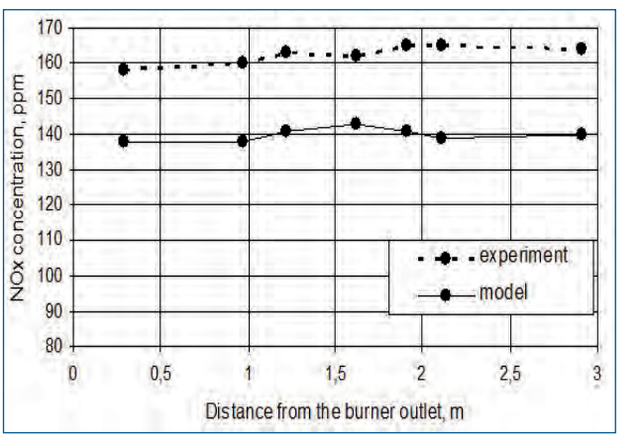

Fig. 6 - Comparison of measurement and numerical calculations results of NO_x concentration of experimental chamber.

Fig. 6 - Confronto tra valori misurati e calcolati della concentrazione di NO_x nella camera sperimentale.

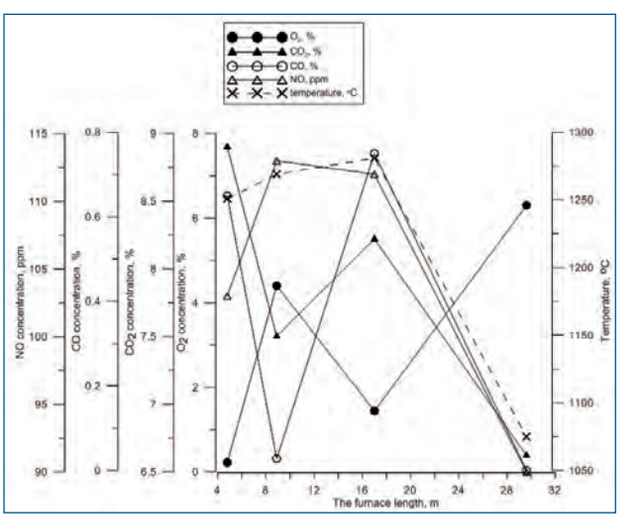

Fig. 7 - The distribution of temperature and concentration of selected combustion products at the furnace length.

Distribuzione della temperatura e della concentrazione di alcuni prodotti di combustione lungo l'asse del forno

FINAL REMARKS

Computer simulations inevitably accompany results of scientific research based on laboratory or industrial

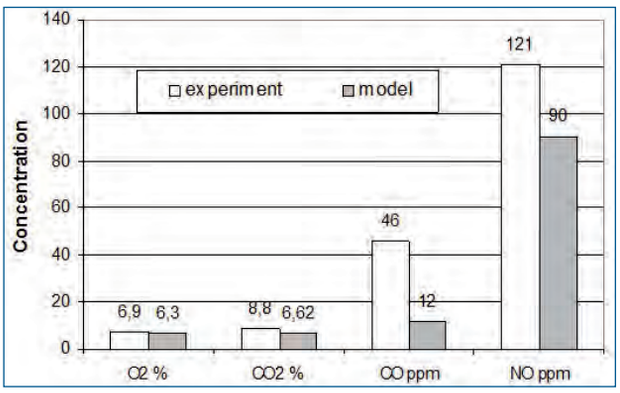

Fig. 8 - Comparison of results of measurements and numerical calculations of selected products of combustion at the outflow of the furnace.

Fig. 8 - Confronto dei risultati delle misurazioni e calcoli numerici all' uscita dal forno, relativi ai prodotti di combustione selezionati.

experiments, which are formulated on the basis of experimental data. Nowadays advanced computer simulations are an extremely useful tool that enables recognition and understanding of complex physical and chemical processes, often impossible to study in the laboratory. An important advantage of computer simulations are small financial outlays in contrast to costly experimental studies.

Commercially available software enables the development of ever more expanded chemical models, and allows to conduct model research not only to predict combustion products, but also to indicate methods for reduction in industrial pollution.

The conducted model research has shown wide possibilities of numerical calculations by the application of CHEMKIN-PRO software. Numerical simulations give the possibility to estimate the chemical composition of natural gas combustion products forming in industrial pusher furnace.

Literature data in the scope of numerical modelling of combustion processes and the results of studies prove the effectiveness of this method and suggest the best way of the harmful products of combustion reduction.

Condition of reagents perfectly stirring, adopted in a studied model research, has been used in analyzing the issues of numerical modelling of combustion processes for a long time. As the example the research conducted by B. Gradon can be mentioned [30]. This points to the advantages of pulsed disorders using in practice in the combustion chambers to increase the intensification of stirring, and thereby to reduce the emissions of combustion products.

Satisfactory results of numerical simulations, and extensive capabilities of software, used for this purpose, suggests undertaking further research. Numerical methods can be used not only to predict the composition of combustion products, but also to reduce their harmfulness with the use of low-emission combustion techniques.

In addition, extension of research range will allow to increase the accuracy of numerical calculations, through the elaboration of the input data including greater number of initial variables obtained through the experiments.

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Previsione della composizione chimica dei prodotti di combustione in forni di riscaldo mediante metodi di modellazione numerica

Parole chiave: Modellazione numerica - Ambiente - Combustione

In questo studio sono stati presentati i risultati dei calcoli numerici della composizione chimica dei fumi prodotti dalla combustione di gas naturale. I calcoli sono stati condotti su un forno a spingitoio operante in un'acciaieria situata in Polonia. La modellazione della chimica del processo di combustione è stata effettuata utilizzando il programma CHEMKIN-PRO. Simulazioni della composizione chimica dei fumi in varie zone del forno sono state effettuate sulla base dei dati di misurazione rilevati dall'oggetto reale.