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SYNGAS AS A REBURNING FUEL FOR NATURAL GAS COMBUSTION

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The paper aims to confirm the syngas application as a reburning fuel to reduce e.g. NO emission during natural gas combustion. The main aim of this modelling work was to predict pollutants generated in the exhaust gases and to indicate the influence of the syngas on the natural gas combustion process. The effect of residence time of fuel-air mixture was also been performed. Calculations were made with CHEMIKN-PRO for reburning process using syngas. The boundary conditions of the reburning process were based on experimental investigations. The addition of 5, 10, 15 and 19% of reburning fuel into natural gas combustion was studied. The effects of 0.001 to 10 s of residence time and the addition of 5, 10, and 15% of syngas on combustion products were determined. The performed numerical tests confirmed that co-combustion of the natural gas with syngas (obtained from sewage sludge gasification) in the reburning process is an efficient method of NO_x reduction by c.a. 50%. Syngas produced from sewage sludge can be utilised as a reburning fuel.

Keywords: natural gas combustion, sewage sludge syngas, reburning, numerical modelling, air pollutants

1. INTRODUCTION

Thermal utilisation of sewage sludge is an energy production method as well as a chemical waste utilisation (Directive 2008/98/EC; Gross et al., 2008; Kosturkiewicz et al., 2011; Magdziarz et al., 2011). There are several thermal technologies of municipal sewage sludge utilisation enabling to obtain useful forms of energy such as pyrolysis, gasification, combustion, and co-combustion (Manara and Zabaniotou, 2012). Gasification is a thermochemical process in which carbonaceous components of a fuel are converted to combustible gas, so-called syngas (Nipattummakul et al., 2010; Topical report, 1999; Werle, 2013). This process reduces the volume of sewage sludge and converts it into a clean combustible gas being an easy form suitable for disposal, e.g. as reburning fuel. Reburning is frequently applied as a low emission combustion process leading to reduced concentration of environmental pollutants. This is one of the so-called "primary methods" of low-emission combustion which are applied inside a combustion chamber. During the combustion of natural gas air pollutants, e.g. nitrogen oxides, carbon oxides, and some amounts of hydrocarbons are formed. Nitrogen oxides (NO_x) are harmful substances generated during fuel combustion. Typical combustion gases contain two kinds of oxides: NO and NO_2. Other kinds of nitrogen oxides are N_2O, N_2O_3 and N_2O_5, but they do not play any essential role in the total amount of NO_x. In typical exhaust gases obtained from natural gas

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combustion, the volumetric share of NO amounts to about 95% of NO_x or even more, the rest being NO_2. NO_x contributes to acid rain while N_2O is a greenhouse gas. Nitrogen oxides belong to the most harmful substances, and this is why research was undertaken to study how to decrease NO_x emission. The concentration of fuel NO in the global emission of nitrogen oxides grows with a decrease of heat release from the combustion chamber. The knowledge of the NO_x formation mechanism can identify thermal and chemical conditions of furnaces and will enable control of combustion processes, thus leading to preventing or reducing emissions of these harmful substances (Wilk, 2002). Carbon oxide is a very noxious substance for living organisms, especially when its concentration exceeds permissible air emission standards. Therefore, when considering the reburning process with syngas, it is important to study the interaction between natural gas and syngas and emission resulting from their combustion. Syngas may contain sulphuric compounds which will be converted into sulphur dioxide, one of fundamental atmospheric pollutants. SO_2 is assumed to be the only representative of sulphur oxides in combustion gases (at least 98% of whole amount of sulphur oxides). During combustion of natural gas and syngas some specific pollutants, such as Cl_2, HCl, salts, dioxins and furanes can be present in exhaust gases. The emission of particulates is a significant problem in the combustion of sewage sludge.

Numerical analysis of combustion processes using a CHEMIKN software allows to carry out calculations not only for conventional fuels but also for renewable energy sources. Therefore, numerical calculations were performed for the reburning process of natural gas combustion and syngas generated during sewage sludge gasification process used as reburning fuel. The obtained results will widen our knowledge of the sludge thermal treatment.

2. LOW-EMISSION METHOD OF NO_X REDUCTION- REBURNING PROCESS

Nitrogen oxide (NO_x) emissions are generated primarily from transportation, electric utility, and other industrial sources particalarly those involving combustion processes. NO_x is reported to contribute to a variety of environmental problems, including acid rain, high ground-level ozone concentrations, or smog formation. Therefore, there is an increasing need for the development and application of cost-effective technologies for controlling these emissions. Techniques for reducing NO_x emissions can be classified into two fundamentally different categories: combustion controls and post-combustion controls. Combustion controls, "primary methods", reduce NO_x formation during the combustion process, while post combustion, "secondary methods", controls reducing NO_x after it has been formed. Combustion controls include low NO_x burners (LNBs), reburning, overfire air (OFA), flue gas recirculation (FGR), and operational modification (Topical report, 1999; Wilk, 2002). Reburning is an in-furnace NO_x control technique that uses fuel to reduce nitric oxide. Reburning involves the staged addition of fuel into two combustion zones: the primary combustion zone and the reburn zone. In the primary combustion zone (where some fuel is injected above the main heat release zone, creating a fuel-rich zone) the amount of NO_x is reduced, the heat release rate is lower (lower production of thermal NO_x), and generally, the excess air fed to the burners is reduced (a lower oxygen concentration results in lower NO_x). In this zone it is important to obtain a complete combustion of the gas and thus to produce NO_x from the fuel-nitrogen, and trigger thermal NO_x. In the reburn zone additional fuel is added to create a reducing condition assisting in converting NO_x produced in the primary zone to molecular nitrogen N_2 and water. Above the reburn zone is a burnout zone in which secondary air is added to complete combustion. Each zone has a unique stoichiometric air ratio as determined by the flows of primary fuels, burner air, reburn fuel, and secondary air (Adams and Harding, 1998; Maly and Zamansky, 1999; Topical report, 1999; Wilk, 2002). The most popular reburning fuel is natural gas, used in reburning of coal combustion in boilers, but alternative fuels offer potential cost and performance advantages. These alternative fuels include biomass, pulverised coal, coal water slurry (CWS), carbonised refuse derived fuel (CRDF) and Orimulsion (Harding and Adams, 2000; Maly et al., 1999), mixed fuel containing scrap tire and Fe_2O_3 (Su et al., 2010), and syngas produced by the gasification of sewage sludge (Gross et al., 2008; Werle, 2012).

Current scientific research is based not only on experimental studies performed in laboratories with modern testing equipment, but also on computer simulations. Computer calculations often allow analysing the phenomena which are difficult to observe or impossible to implement in reality. Because of the wide possibilities of its application, numerical modelling has become an essential element of experimental research.

3. NUMERICAL MODELLING

Numerical analysis of the combustion of natural gas for the formation of nitrogen oxides was conducted using the latest version of CHEMKIN - PRO. CHEMKIN is the product of Reaction Design, which evolved from its origin as a Sandia National Laboratory combustion code Chemkin II. Currently CHEMKIN is commercial-quality software which enables the simulation of complex chemical reactions for modelling and surface phase chemistry. It is uniquely qualified to lead the Clean Technology approach to the design and improvement of combustors, engines and chemical reactors. CHEMKIN-PRO is specifically designed for large chemical simulation applications requiring complex mechanisms.

The heat chamber of the furnace was assumed as a "Perfectly Stirred Reactor". The results of model calculations using the PSR model are obviously burdened with a small error, due to the condition of ideal mixing of the reactants. However, the investigation of disturbances of e.g. air flow carried out by the authors showed that calculated results of the calculations are in agreement with the model and often overlap. The perfectly stirred reactor is commonly used by researchers not only in the reburning process modelling, but also in other low-emission methods of calculating air recirculation, or air staging, which is evidenced in a large body of published studies (Adamczuk and Radomiak, 2010; Fan et al., 1998; Faravelli et al., 2001; Ljungdahl and Larfeldt, 2001; Luan et al., 2009; Magdziarz et al., 2011; Szecowka et al., 2003; Wang et al., 2005; Werle, 2012).

The main aim of the present work was the modelling of the exhaust gas composition from reburning process of natural gas with syngas as reburning fuel. A scheme of the numerical modelled combustion chamber is presented in Figure 1.

The conditions of reburning process as well as the dimensions of combustion chamber were determined in earlier experiments, which allowed to define the boundary conditions in the modelling procedure (Adamczuk and Radomiak, 2010; Szecowka et al, 2003). The combustion mechanism took into account the formation and decomposition reactions of nitrogen and sulphur oxides. According to the experimental experience the syngas was assumed at a distance of approximately 1/3 the length of the main burner chamber, supplied by natural gas, resulting from combustion conditions. Table 1 presents the composition of natural gas used for the calculations obtained by means of chromatography analysis. The composition of natural gas was identified with GC Agilent Technologies 7890A.

Table 1. Natural gas composition

CH_4, %	C_2H_6, %	CO_2, %	O_2, %	N_2, %
95.9	1.5	1.1	0.2	1.3

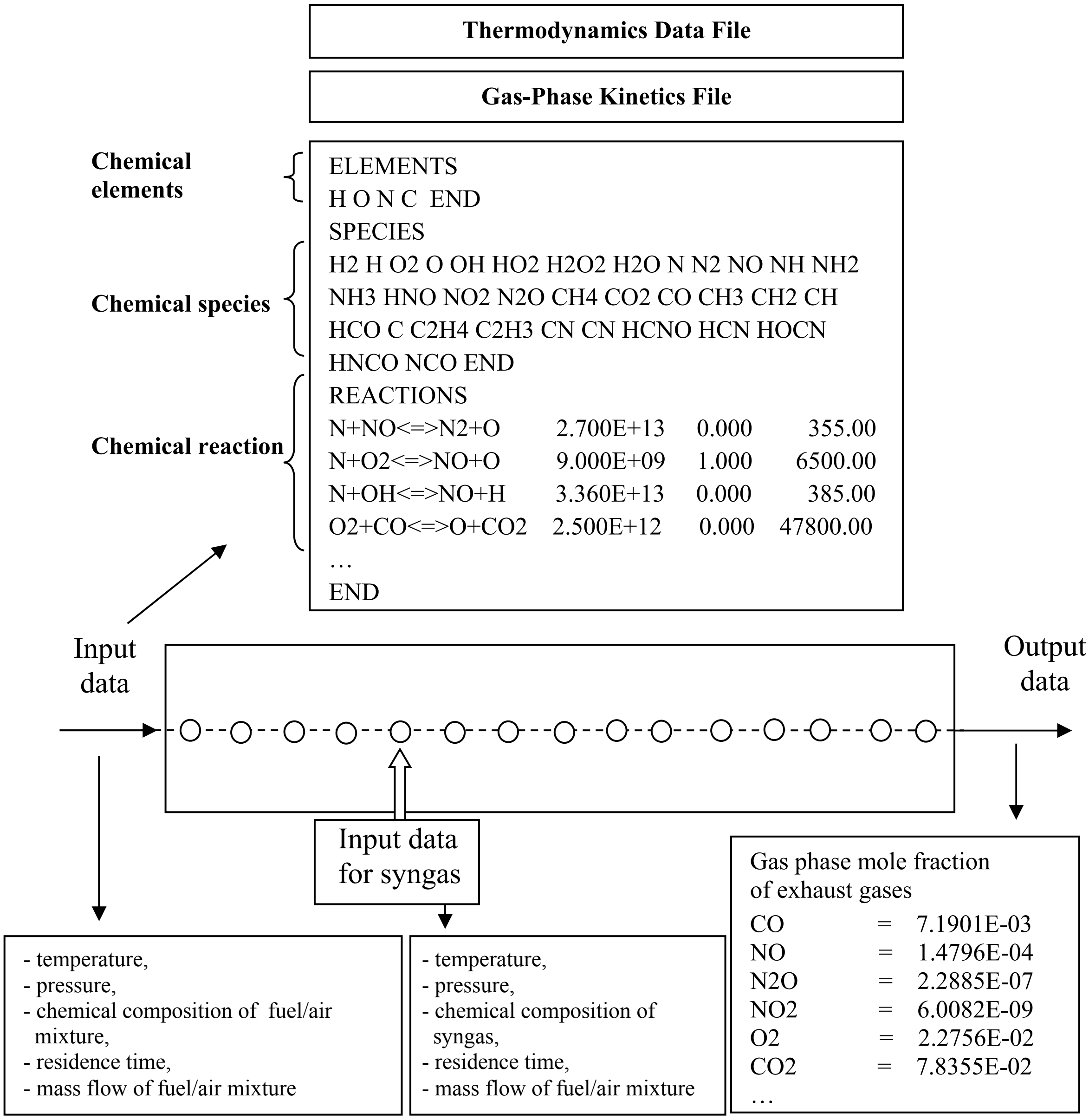

Fig. 1. A scheme of numerical modelled combustion chamber

The syngas chemical composition was taken from the literature (Gross et al., 2008). Table 2 presents the composition of the wet product gas leaving gasifier. Dried sewage sludge gasification was carried out using fluidised bed technology. Carbon (21.8%), hydrogen (3.6%), oxygen (18.7%), nitrogen (3.9%), sulphur (0.85%), and ash (35.9%) were the main components of the sewage sludge.

Table 2. Assumed syngas composition (Gross et al., 2008)

CO	H_2, %	CH_4, %	CO_2, %	N_2, %	O_2, %	SO_2, %	H_2O, %
24.0	11.0	2.0	9.0	46.0	0.7	0.3	7.0

The chemical file "chem.inp" and the thermodynamic file "therm.inp" were obtained from Leeds University website (www.chem.leeds.ac.uk/Combustion/Combustion). The chemical model of natural gas combustion includes 132 chemical reactions and 35 elements and compounds: 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. Therefore, numerical calculations were carried out for the complicated reburning process of natural gas combustion with sewage sludge syngas as reburning fuel. The assumed syngas composition included SO_2. That is why the model was extended to reactions involving SO_2 present in fuel. The model ultimately consisted of 163 chemical reactions and 47 compounds, e.g. SO, SO_2, SO_3, SN, SH, COS, H_2S, HSO, HSOH, H_2SO, HOS, and HOSHO.

Boundary conditions were based on a preliminary laboratory experiment conducted in a quartz chamber with a cylindrical cross section and diameter of 0.12 m and length 3.2 m. Experiments on natural gas combustion using natural gas as a reburning fuel were carried out by Adamczuk and Radomiak (2010); Szecowka et al. (2003). Based on data obtained in the experiment, e.g. excess air ratio, gas volume flows, temperature profile, the input data file was defined. The in-put data included the mass flow of fuel-air mixture, the molar fraction of reactants based on the composition of the fuel and air, residence time in the zone of maximum temperature, temperature profile in the chamber, the temperature of the media input, the combustion temperature, and pressure – 1.01 bar. The amount of gasification gas into the combustion chamber was equal to 5, 10, 15 and 19% in relation to the chemical energy of the fuel. The input data were as follows:

- in the primary zone:

- gas flow, $\dot{V}_{gas}$ = 0.00028 m3/s (T = 293 K),
- air excess ratio, λ = 1.05,
- gas temperature at the beginning of the process, T = 293 K,
- calculated combustion temperature, T_c = 1800 K.

- in the reburning zone:

- reburning gas flow, $\dot{V}_{reburning\ gas}$ = 0; 0.000103; 0.00022; 0.00035; 0.00047 m3/s,
- reburning gas fraction, rb = 0; 5; 10; 15; 19% ,
- residence time, τ = 1 ms,
- air excess ratio, λ = 0.85 - 0.93.

Additionally, calculations taking into account the effect of residence time for the in-put data presented above as well as the addition of syngas into reburning process were done. The residence time used for calculations was from 0.001 up to 10 s.

4. RESULTS AND DISCUSSION

The calculated concentrations of combustion products such as NO, CO, NO_2, N_2O, CH_4 and OH radical inside the combustion chamber are shown in Figures 2-9. The figures present calculation results taking into account the percentages of reburning fuel injected into natural gas during combustion.

The calculated NO and CO concentrations after addition of 0, 5, 10, 15 and 19% of the reburning fuel (syngas) are plotted in Figure 2. Unfortunately, the reburning process using syngas from sewage sludge gasification causes a three-fold increase in CO concentration: from 860 to 26600 ppm of CO for 19% addition of the reburning gas (Fig. 2). The calculated NO molar fraction was estimated in the range of 168 to 91 ppm. The additions of reburning fuel-syngas in the range of 0 to 15%, significantly decrease NO concentration, e.g. 15% of syngas decreases NO concentration by about 44%. On the other hand, the addition of 19% of syngas causes the increase of NO by about 21%, but even the addition of 19% of syngas results in 31% decrease of NO concentration compared to natural gas combustion without reburning process. During natural gas combustion NO concentration in the exhaust gases was higher than that if the reburning process took place despite the fact that the syngas cantained much nitrogen. This is due to the specifics of the reburning process leading to conversion of NO into N_2, which takes place because of the presence of CN, OH, and CH_i radicals generated from the reductive decomposition of the fuel, in this case, the syngas obtained from sewage sludge gasification. The use of the CHEMKIN software modelling procedure enabled a detailed analysis of the reburning process, with particular emphasis on the role of CH_i, and OH radicals in the conversion of NO to N_2. Figure 3 presents the role of OH and CH radicals, showing that in co-combustion of natural gas and syngas an increasing participation of OH and CH radicals is evident.

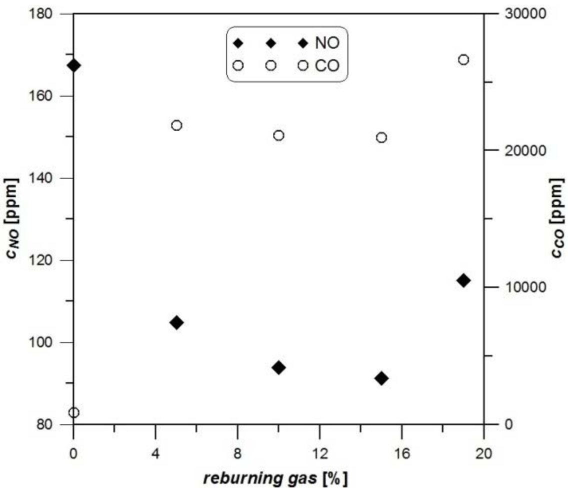

Fig. 2. Concentration of NO and CO as a function of reburning gas addition

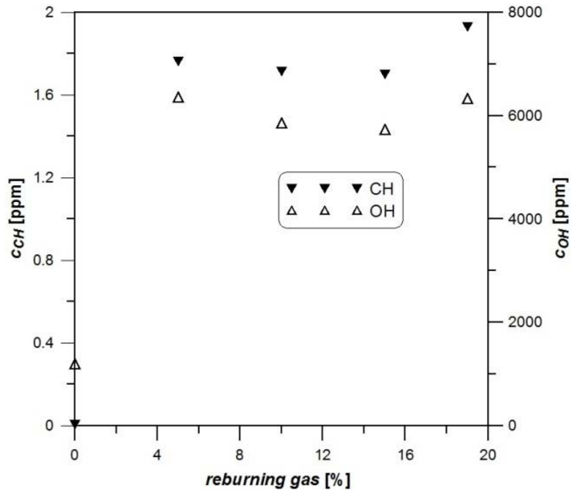

Fig. 3. Concentration of OH and CH radicals as a result of reburning gas addition

Despite the fact that NO_2 and N_2O do not significantly affect the total NO_x concentration, the estimation of NO_2 and N_2O was performed. It is evident that the addition of syngas decreases their levels. Especially NO_2 concentration significantly decreased after the addition of reburning fuel in the entire studied range of syngas addition. In the case of N_2O, the reduction of N_2O is only slightly perceptible (Fig. 4).

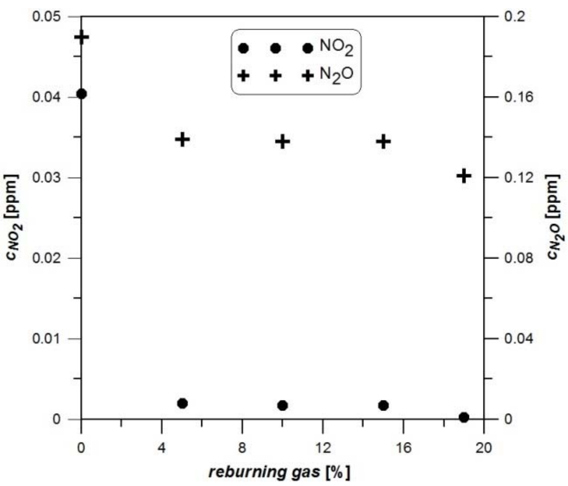

Fig. 4. Concentration of NO_2 and N_2O as a function of reburning gas addition

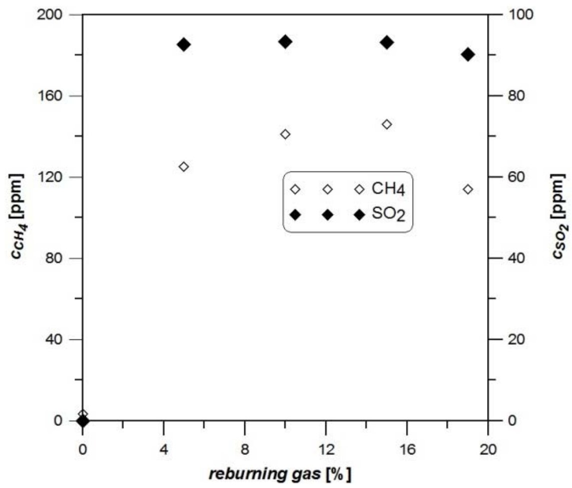

Fig. 5. Concentration of CH_4 and SO_2 as a function of reburning gas addition

Unfortunately, the estimated concentrations of CH_4 and SO_2 increase significantly with the addition of syngas as compared to natural gas combustion process. However, the addition of different amount of syngas hardly changed concentration of CH_4 and SO_2 in the exhaust gases. The concentrations of CH_4 and SO_2 resulting from the reburning fuel composition are shown in Fig. 5.

The effect of residence time, from 0.001 up to 10 s, and the addition of syngas into reburning process for combustion product (NO, CO, CH, OH, CH_4, SO_2, NO and N_2O) concentrations was also studied.

Figure 6 presents the influence of residence time of gaseous mixture in the combustion chamber on NO and CO concentration studied for 5, 10 and 15% syngas added in reburning process. For all studied cases the highest concentration of NO and CO is found for τ = 0.001 s. However, NO concentration decreases slightly with an increasing residence time for 5 and 10% of syngas added, but after 1 s it does not change much. At 15% of the added syngas, NO concentration decreases for all studied values of the residence time. In the case of CO estimated for the added 5 and 10% of syngas, a decrease in CO concentration is significant up to τ = 1 s, then only a slight decrease can be observed. For 15% of syngas CO concentration has a similar tendency as for the two cases mentioned above, but the obtained CO concentration is higher than for 5 and 10% of syngas calculated for 1 to 10 s of the residence time values.

Figure 7 presents CH and OH radical concentrations estimated for the studied parameters. Both, CH and OH radical concentrations fall with an increasing residence time of reacting mixture and the trends of the obtained results are similar in all studied cases. The highest fall is observed for τ = 0.001 to 1 s and then only a slight decrease is found for CH and OH radicals, while higher CH and OH radical concentrations are obtained for 10% than for 5 and 15% of the added syngas.

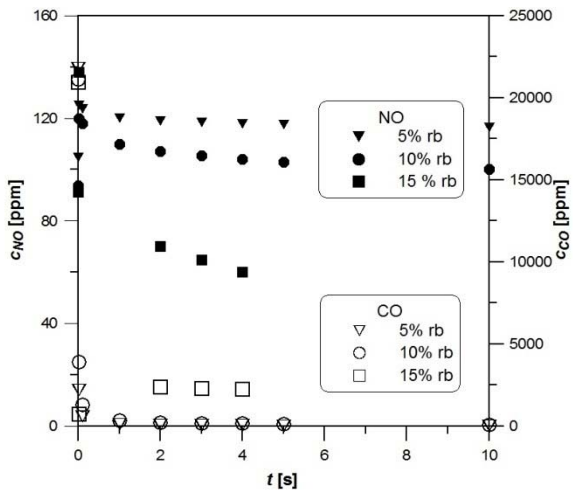

Fig. 6. Concentration of NO and CO as a function of residence time and reburning gas addition

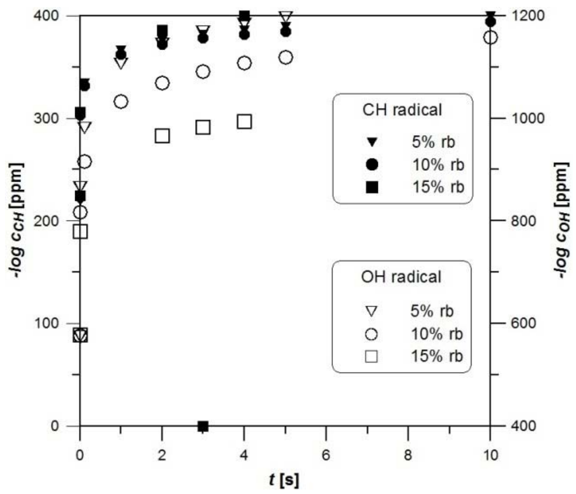

Fig. 7. Concentration of CH and OH radicals as a function of residence time and reburning gas addition

Increasing concentration of NO_2 with an increasing τ up to 5 s for three values of reburning fuel is demonstrated in Fig. 8. The calculated NO_2 concentration estimated for 10 s is a trend continuation. In contrast to NO_2, N_2O concentration decreases under the same conditions.

The effect of residence time and addition of syngas on the concentration of CH_4 is evident. CH_4 concentration decreases with an increasing residence time. The highest results are obtained for 15% of syngas added compared to 5 and 10% of syngas. It results from the increasing amount of the added syngas containing higher amount of CH_4 (Fig. 9).

However, SO_2 concentration does not change with an increasing residence time, while it depends on the amount of added syngas. The more syngas is added in the reburning process, the more SO_2 is formed. Concluding, the highest NO, CO, CH, OH, and N_2O compositions were calculated for 0.001 s in the studied range of the residence time, then a slight decrease was observed. In contrast to above mentioned compounds, the concentration of NO_2 increased with an increasing residence time. The effect of the residence time on CH_4 and SO_2 was not found.

The authors of the paper with the full responsibility state that the CHEMKIN program is the best, if not the only program that allows the analysis of such complex processes to be preformed. Other commercially available computer programs such as KIVA, FLUENT or CANTERA enable onlz limited analysis of phenomena related to combustion processes.

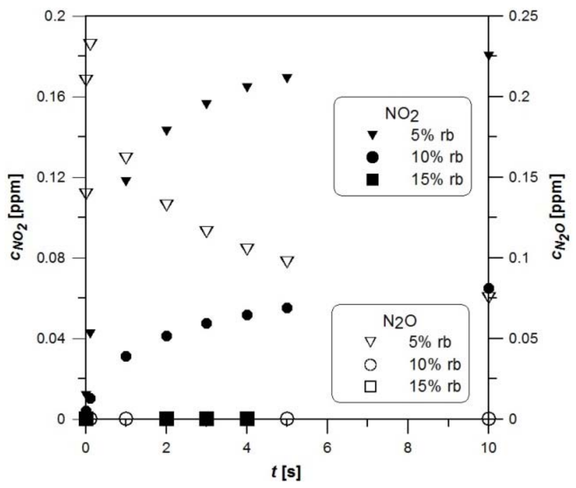

Fig. 8. Concentration of NO_2 and N_2O as a function of residence time and reburning gas addition

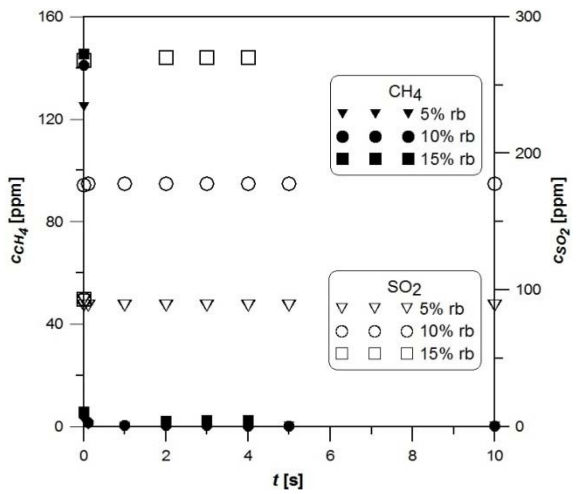

Fig. 9. Concentration of CH_4 and SO_2 as a function of residence time and reburning gas addition

5. CONCLUSIONS

This paper presents modelling of reburning process of natural combustion with syngas obtained from the sewage sludge gasification, which is currently very popular. Due to modern methods seeking to provide solutions to the complexity of problems related to pollution from the natural gas combustion process. Calculations were performed to determine natural gas composition. The concentration of syngas was taken from the literature. The effects of 5, 10, 15, and 19% addition of reburning fuel during natural gas combustion were studied. Numerical modelling enables identification of hazardous gaseous pollutants such as NO, CO, NO_2 and others which form during the reburning combustion process. The influence of an increasing addition of reburning fuel was evidently observed. Concentration of NO, NO_2 and N_2O decreased after the addition of the reburning fuel. In contrast to the observed results for NO_x, the calculated CO, SO_2, CH_4, and OH radical concentrations increased for the studied range of syngas additions. Numerical calculations concerning the effect of residence time and the addition of syngas into the reburning process for combustion product (NO, CO, CH, OH, CH_4, SO_2, NO and N_2O) concentrations enabled to choose the optimal value of the residence time used as an input data. The choice of 1 ms of the residence time for calculations provided the highest concentrations of studied characters. Therefore, when considering the obtained data, the authors suggest that the syngas from sewage sludge gasification process can provide a significant reduction of NO_x.

SYMBOLS

rb	reburning gas fraction, %
T	temperature, K
T_c	calculation combustion temperature, K
$\dot{V}$	gas flow, m^3/s

Greek symbols

λ	air excess ratio
τ	residence time, s

Subscripts

c	calculation method

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