ESTIMATION OF REBURNING POTENTIAL OF SYNGAS FROM SEWAGE SLUDGE GASIFICATION PROCESS

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The motivation of this work was to define the reburning potential of sewage sludge gasification gas (syngas). Numerical simulation of co-combustion process of syngas in hard coal-fired boiler has been done. All the calculations were performed using the Chemkin program. Plug-Flow Reactor model was used. The calculations were modelled using GRI-Mech 2.11 mechanism. The highest NO conversions are obtained at the temperature of about 1000-1200 K. The highest reduction efficiency was achieved for the molar flow ratio of syngas equal to 15%. The combustion of hard coal with sewage sludge – derived syngas reduces NO emissions and the amount of coal needed to produce electricity and heat. Advanced reburning, which is a more complicated process gives efficiency of up to 80%. The calculations show that the analyzed syngas can yield better results.

Keywords: gasification, sewage sludge, plug flow reactor

1. INTRODUCTION

According to Polish Regulations, sewage sludge belongs to group No. 19, including waste from treatment installations and the equipment for waste management, waste water treatment plants and from the treatment of drinking water and water for industrial purposes. According to the Polish Ecological Policy and the objectives of the National Waste Management Plan 2014 (NWMP) and the National Urban Wastewater Treatment Program (NUWTP), the quantity of sewage treated in Poland systematically increases. In 2010 in Poland, 612.8 thousand tons (dry basis - d.b.) of sewage sludge was produced. Moreover, it is estimated that the quantity of sludge which will be produced in Poland in 2018 will be equal to 706.6 thousand tons (d.b.).

Currently, the predominant method for the disposal of sewage sludge in Poland is its storage and agricultural application (Werle and Wilk, 2010). This is highly unfavorable. The main problems are a the high percentage of stored sewage sludge and a lack of installations for its thermal utilisation. Thermal processes can be used for the conversion of large quantities of sewage sludge (e.g., in large urban areas) into useful energy. Processes for thermal utilisation of sludge can be developed at existing installations (e.g., heating plants, power plants, or cement plants) or in newly built facilities.

There is a large and pressing need for the development of thermal methods for the disposal of sludge. It is worth emphasising that effective from January 1, 2013, the storage of sewage sludge with parameters as presented in Table 1 will be banned in Poland.

Gasification is one of the possible ways of sewage sludge utilisation. This process provides an attractive alternative for the treatment of sewage sludge. It has several advantages over traditional

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combustion. It takes place in a low oxygen environment and limits the formation of dioxins and large quantities of SO_x and NO_x. Furthermore, it requires just a fraction of the stoichiometric amount of oxygen necessary for combustion. As a result, the volume of process gas is low, requiring smaller and less expensive gas cleaning equipment. A lower gas volume also means a higher partial pressure of contaminants in the off-gas, which favours more complete adsorption and particulate capture. Finally, gasification generates a fuel gas that can be integrated with e.g. combined cycle turbines or engines.

Table 1. Polish criteria for the storage of sewage sludge in a non-hazardous waste landfill

	Parameter	Value limit
1	2	3
1	Overall organic carbon (OOC), % d.b.	5.0
2	Loss at calcinations (LOC), % d.b.	8.0
3	The higher heating value, MJ/kg d.b.	Maximum 6.0

Syngas is increasingly seen as a source of environmentally clean fuels and has potential to become a major fuel in the production of essentially pollution – free energy. There are many advantages of using syngas as a fuel: it is inexpensive and can be used for clean combustion resulting in a significant reduction in pollutant emissions. Syngas can be also be employed as a supplemental fuel do reduce the consumption of other fuels, such as pulverised coal and fuel oil. In addition to this, it has potential as a reburning fuel to reduce NO_x emission. Polish energy sector offers a great potential of utilising the gas and simultaneously reducing NO_x emissions.

2. REBURNING

Nitrogen oxides, also known as NO_x, are important and problematic contaminants produced in combustion processes (Dąbrowski and Piecuch, 2011; Piecuch et al., 2009; Szkarowski, 2001; Szkarowski, 2002). The nitrogen chemistry in combustion processes is summarised in Fig. 1.

NO_x are formed along three routes: thermal and prompt formation from N_2 and oxidation of fuel-bound nitrogen. Thermal NO evolves from the recombination of N_2 and O_2, described by the extended Zeldovivch mechanism (Norman et al., 2009):

$$N_2 + O_2 \leftrightarrow NO + N \qquad (1)$$

$$N + O_2 \leftrightarrow NO + O \qquad (2)$$

$$N + OH \leftrightarrow NO + H \qquad (3)$$

Reaction (1) has high activation energy, which controls the reaction rate of thermal-NO_x formation; temperatures above 1400 °C are required for the reaction to occur. The Zeldovivch mechanism is reversible and reduces NO when the equilibrium concentration of NO is low enough, given that the temperature is sufficiently high for the Zeldovivch mechanism to be active. Prompt NO (Norman et al., 2009) is initiated by the reaction between N_2 and hydrocarbon radicals forming a volatile-N (an intermediate gaseous compound) species, which is oxidised to NO or reduced back to N_2. Prompt NO is of minor importance and occurs predominantly under fuel-rich conditions. The conversion of fuel-bound nitrogen contributes to most NO formed during combustion of coal. Fuel-N is split into a volatile- and char–N during the devolatilisation. The volatile–N transforms into either NO or N_2, while char-N reacts through a set of heterogeneous reactions along with the oxidation of char. The split between volatile- and char-N varies with a type of coal and depends on qualities such as nitrogen

content, coal rank and volatility, but is also depends on combustion conditions, such as heating rate and temperature. NO formed can also be reduced to N_2. Similar to the prompt formation, NO can be reduced by hydrocarbons to volatile-N under fuel-rich conditions which is known as reburning.
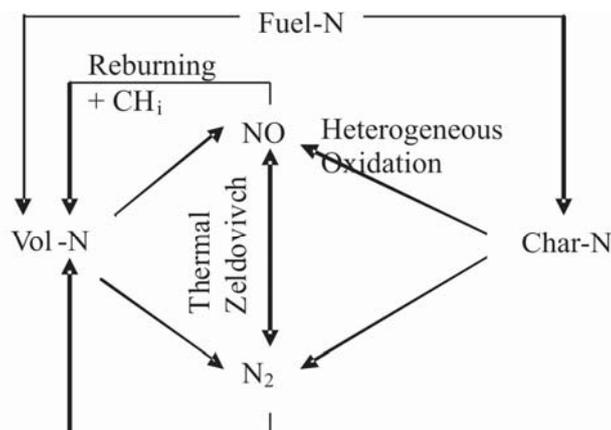
Fig. 1. Overall mechanism of NO formation and reduction

Nowadays, reburning is shown as one of the most efficient and attractive techniques that can be applied to reduce NO_x. In this technique, after the primary combustion zone, a secondary fuel is injected into the boiler in the so-called reburn zone, thereby establishing fuel-rich conditions. The reburn fuel produces hydrocarbon radicals, which can interact with NO and therefore reduce the final NO_x emissions. To complete the process, post combustion air is injected in order to eliminate unburned hydrocarbons.

In the past several years, reburning has been extensively studied. The first researchers who developed this concept calling it 'reburning' were Wendt, Sterling and Matovich (1972). They suggested an injection of additional fuel along the main flame, so that the emission of NO_x might by minimised by forming a reductive atmosphere. Reburning was first demonstrated as a practical NO_x reduction method in Japan where the concept of reburning was first applied to a full-scale boiler (Takahashi et al., 1983). Because of this successful example and high efficiency of reburning technology leading to a of NO_x emission (more than 50%), many investigators have conducted bench-scale and pilot-scale reburning tests. Laboratory (Folsom et al., 1991) and the first commercial (Folsom, 1997a) boilers showed that the NO_x emission may be reduced by up to 60%. It has also been reported (Smoot, 1998) that many European countries conduct reburning projects in coal fired boilers with a goal of less than 200 mg/m^3_n NO_x emissions. Four demonstration projects in the Department of Energy's Clean Coal technology Program are being carried out in the US. One of these uses coal as the reburning fuel, while the others use natural gas. NO_x reductions varied from 30% to over 70%, depending on the boiler type and scale, reburning fuel and simultaneous use of low-NO_x burners. In the US its principal application has been to coal-fired boilers, while in Europe and Japan reburning applications include coal, oil and gas-fired boilers as well as municipal solid waste (MSW) incinerators. The European example of gas reburning is a 600 MW coal-fired boiler at Longannet (Lanigan et al., 1991). This is the largest application of gas reburning in Europe. As far as the kind of the reburning fuels is concerned, they should be volatile and highly reactive. Such fuels are natural gas (Adams and Harding, 1998), coke-oven gas (Smoot et al., 1998), synthesis gases (Frassoldati et al., 2007) and light fuel oils. It should be fuels which while decomposing in the reduction zone produce a large amount of CH_i radicals. The results of recent investigations have shown that also heavy fuel oil, lignite (Hardy, 2003), some kind of biomass (Adams and Harding, 1998; Cariln et al., 2009) and even some municipal waste (Maly, 1999) may be good reburning fuels. A good example of reburning fuels can be the gas obtained from sewage sludge gasification.

The chemistry of reburning is very complex and involves a large number of reactions of formation and consumption of many different species. NO reduction through reburning usually includes interactions of hydrogen cyanide (HCN), ammonia (NH_3) and NO species (Shen et al., 2004)]. These species react in a series of reactions, but finally they end up as NH and N, as shown in Fig. 2.
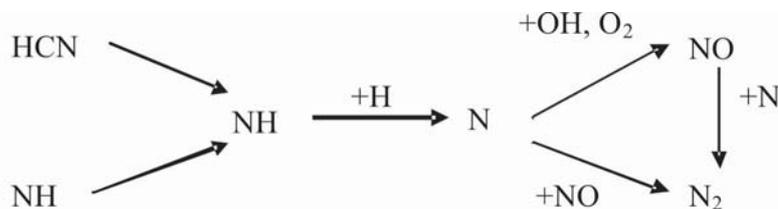
Fig. 2. Reaction-path diagram illustrating step in volatile-N conversion to NO or N_2

Under fuel-rich conditions, the formation of HCN relies strongly on the concentration of hydrocarbon species:

$$CH_i + NO \rightarrow HCN + \ldots \tag{4}$$

which then decays through NCO $\rightarrow$ NH $\rightarrow$ N, as shown in Eqs. (5-8), and ultimately reaches N_2 via reverse Zeldovich reaction (Eq. 8):

$$HCN + O \rightarrow NCO + H \tag{5}$$

$$NCO + H \rightarrow NH + CO \tag{6}$$

$$NH + H \rightarrow N + H_2 \tag{7}$$

$$N + NO \rightarrow N_2 + O \tag{8}$$

Several kinetics models have been reported to describe the reburning process with different levels of details and predictions (Bilbao et al., 1995; Dagaut et al., 1998; Galborg et al., 1998; Klipinen et al., 1992; Miller and Bowman, 1998; Smith et al., 1998;)), providing an overall understanding of the interaction among hydrocarbon and nitrogen species.

Depending on their complexity and description level, these models may be classified as detailed, skeletal and reduced models. Both detailed and skeletal mechanisms are shown to be essential since they are the starting point for the development of reduced mechanism, which can be used in modelling reburning with detailed fluid dynamics, for example, in the presence of turbulent flows. The first reduced mechanism for the reburning process was developed by Glarborg et al. (1992a). This contained a four-step mechanism for methane oxidation plus two rate-of-production terms for HCN and NO. In 1996, Hewson and Bollig (1992) proposed a reduced mechanism, based on a detailed mechanism developed in the same research group that consists of 11 global reactions. Several reduced mechanism have been proposed for the GRI-Mech. Reduced mechanisms have important limitations since their predictions are accurate only for a narrow range of operating conditions. The detailed mechanism GRI-Mech 2.11, that consists of 279 reactions of 48 reactive species, was developed for a wide range of operating conditions.

Numerical simulation of co-combustion process of gas from sewage sludge gasification in coal-fired boiler was the aim of this work. All calculations were performed using Chemkin, the plug flow reactor model were used. Reburning potential of syngas has been defined. The influence of the reaction between reburning fuel and flue gases from coal combustion on reduction of NO_x emission has been examined. The results show a relationship between relative amounts of nitric oxide on the temperature T, the air excess ratio λ and the reaction time τ.

3. CALCULATIONS

In the calculations, two gases from sewage sludge gasification process (ss1 and ss2) were used as the reburning fuel. The molar composition of the reburning fuels is presented in Table 2. Syngases composition was achieved using a non-stoichiometric approach equilibrium model calculation. This model is based on minimising the Gibbs free energy in the system (Werle, 2011; Werle, 2012).

Table 2. Molar composition of the analysed sewage sludge

Compound vol.%	Reburning fuels	
	Syngas1 (ss1)	Syngas2 (ss2)
H_2	11.0	6.0
CO	24.0	27.0
CH_4	2.0	9.0
N_2	46.0	41.0
O_2	4.0	4.0
CO_2	9.0	9.0
H_2O	4.0	4.0

During the simulations, the combustion of hard coal was assumed to be complete. The mass fraction of the main components of coal is equal as follows: $c = 0.650$; $h = 0.048$; $s = 0.004$; $o = 0.128$; $n = 0.017$; $w = 0.100$. The flue gases from the combustion were mixed with the reburning fuel.

The calculations were modelled using GRI-Mech 2.11 mechanism. This version is relevant to analysing reburning process. The flue gases were mixed with the reburning fuel. The molar flow rates ratio of the reburning fuel is assumed to be 5%, 10% and 15% of the whole exhaust. The simulations were conducted for a constant pressure equal to 1 atm. for temperatures range from 600 K to 1400 K and typical residence time 0.0 - 0.4 s. It was assumed that a flue gase which enters into the reburning zone contains 300 ppm of NO and that during combustion only NO is formed without other NO_x.

4. RESULTS

The obtained results show how a relative amount of NO depends on the air excess ratio, temperature of the reburning process, the molar flow rates ratio of reburning fuel and the residence time in reburning zone.

4.1. Influence of the molar flow rates of reburning fuel

Figure 3 (a-c) shows the dependence of a relative amount of NO as functions of air excess ratio for the gas from sewage sludge gasification no 1 (ss1) for different rates of molar ratio of the reburning fuel. While analysing Figure 3 it can be said that the most intensive reduction is observed for the value of air excess ratio equal to 1.1 and the temperature equal to 1200 K. However, it can be noticed that an increment of the molar ratio of reburning fuel (10%) leads to an enlargement of the temperature range in which NO reduction is the most intensive. For example, for the case presented in Fig. 1b, an equally high NO reduction is obtained for the range of the reburning zone temperature equal to 1100-1400 K. Moreover, it seems that a further increment of the molar flow rate (15%) of reburning fuel leads to a further enlargement of the interval of the reburning zone temperature in which NO reduction is the

most effective – in Fig. c the efficiency of NO reduction even at 900K is quite high and equal to 35%. The best efficiency (almost 90%) was obtained at the molar rate of the reburning fuel equal to 15%. However, it is worth emphasising that such a high value of NO reduction in a real system would be considered as an excellent value.

a)

b)

c)
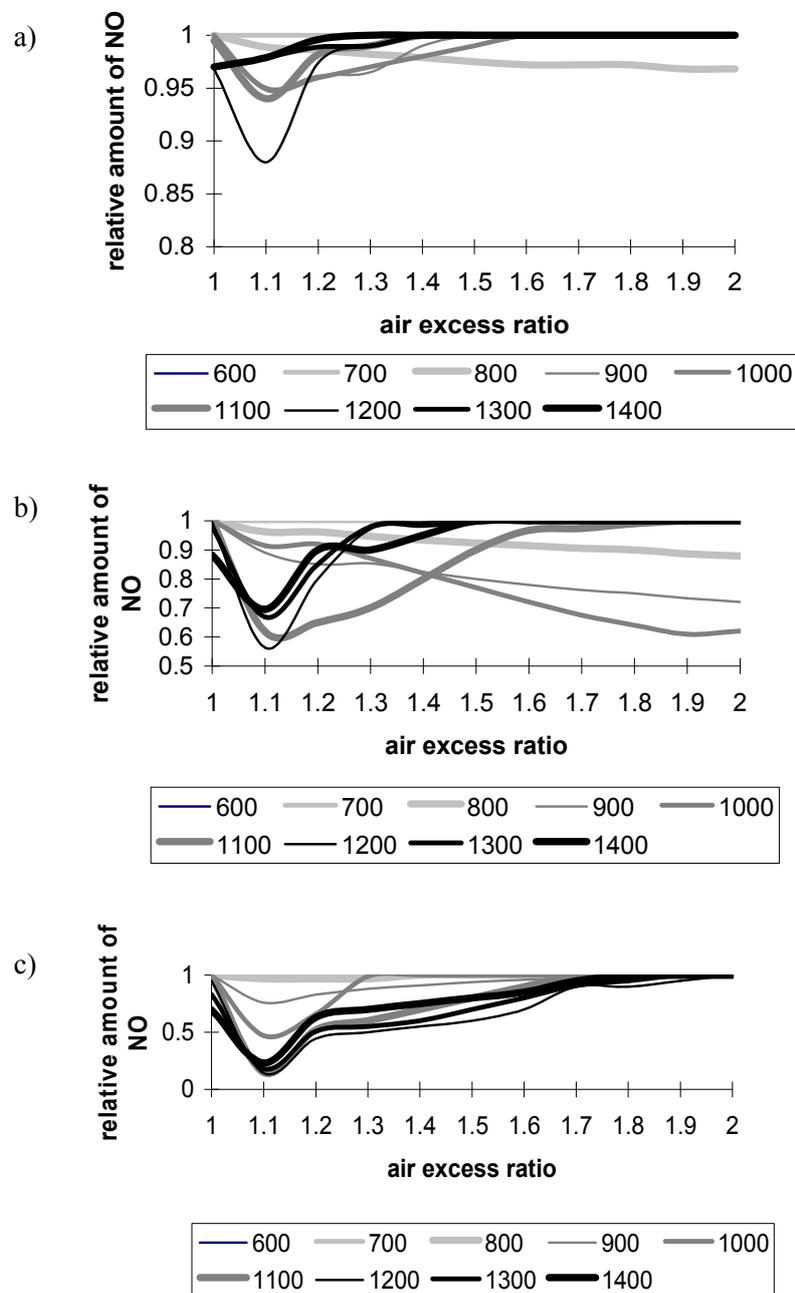
Fig. 3. Relative NO concentration as a function of air excess ratio; a) ss1; 5%; $\tau = 0.4$ s; b) ss1; 10%, $\tau = 0.4$ s; c) ss1; 15%, $\tau = 0.4$ s

4.2. Influence of the air excess ratio

Figure 4 shows dependence of efficiency of NO reduction as a function of air excess ratio for temperature in the reburning zone equal to 1200 K and the residence time in the reburning zone equal to 0.4 s (for ss1). While analysing this figure, it can be seen that at the air excess ratio equal to 1.0 reburning fuel has no influence on NO reduction.
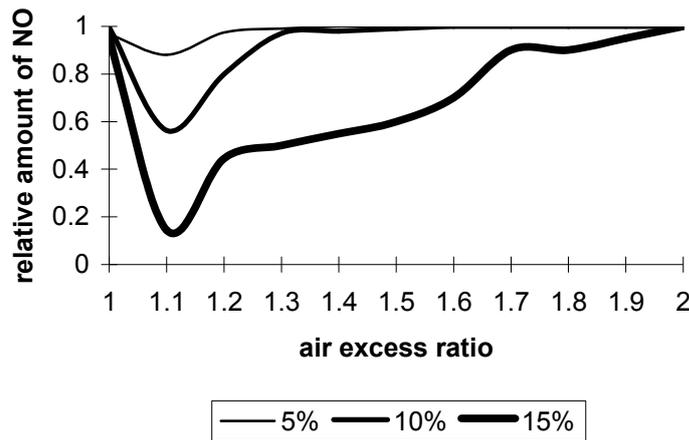
Fig. 4. Relative amount of NO as a function of air excess ratio for different values of molar ratio of reburning fuel (ss1; T = 1200 K; τ = 0.4 s)

It can be observed for each presented curve, that a relative amount of NO decreases at the beginning with the growth of the air excess ratio. As can be observed in this Figure, there is a value of air excess ratio, in which the efficiency of NO reduction reaches its maximum (simultaneously minimum values of the amount of NO). Above the value of air excess ratio in which NO reduction is the highest, this parameter decreases (simultaneously the relative amount of NO increases). Generally it can be concluded that with increasing the molar rate of reburning fuel ratio the efficiency of NO reduction increases.

4.3. Influence of the reburning temperature

In Figure 5 the relative amount of NO as a function of temperature in the reburning zone is shown. NO reduction efficiency of sewage sludge gasification gas is strongly dependent on temperature. For example, at temperatures between 600-800 K sewage sludge derived syngas does not affect NO concentration. At higher temperatures, reburning efficiency of the analysed reburning fuel increases. The strongest NO reduction begins at temperatures higher than 1000 K. The best temperature for reburning is equal to 1200K. It can be concluded that with increasing the rate of molar ratio of the reburning fuel, the optimum temperature for NO reduction is lower.
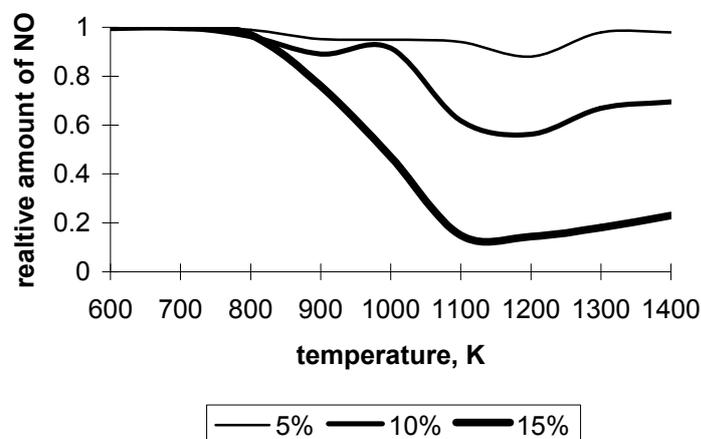
Fig. 5. Relative amount of NO as a function of temperature in the reburning zone for different values of molar ratio of reburning fuel (ss1; λ = 1.1; τ = 0.4 s)

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4.4. Influence of the residence time

Figure 6 shows how relative NO concentration depends on the residence time.
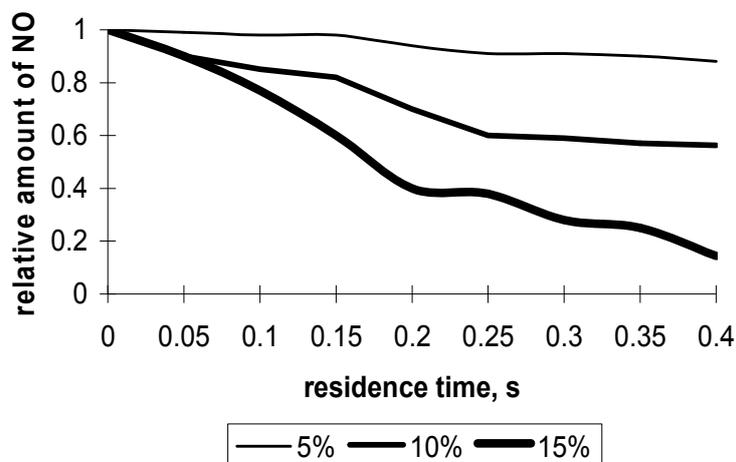
Fig. 6. Relative NO concentration in dependence on residence time (ss1, $T = 1200K$; $\lambda = 1.1$)

The most intensive reduction of NO for the range of residence time equal to 0.05-0.20 s is observed. Generally it can be said, that above this time interval the concentration remains constant. However, an increment of the rate of the molar ratio of the reburning fuel leads to a much longer time in which NO reduction is effective. Finally, the highest NO reduction for syngas from sewage sludge gasification gas occurs at 0.4s for the molar ratio of the reburning fuel equal to 15%.

4.5. Influence of the type of sewage sludge gasification gas

Figure 7 (a-c) shows a comparison of the results for two different types of sewage sludge gasification gas (ss1 and ss2). This figure presents dependence of a relative amount of NO as a function of air excess ratio, temperature in the reburning zone and the residence time. While analysing this figure we can see that the efficiency of NO reduction for using ss2 as the reburning fuel is significantly higher in comparison with ss1. The difference is particularly substantial for the molar fraction of the reburning fuel equal to 5 and 10%. However, it can be observed that at the molar fraction of the reburning fuel equal to 15% the type of sewage sludge derived syngas has no influence on the efficiency of NO reduction (in that case both syngases used in the process are characterised by more or less the same value of NO reduction). The gas from the sewage sludge gasification process denoted as ss2 is characterised by a lower molar concentration of nitrogen and higher values of carbon monoxide and methane. It is also very characteristic that despite the lack of differences in the values of achieved NO reduction efficiency for the molar fraction of the reburning fuel of 15%, gas ss2 is characterised by a much wider range of the air excess ratio in which NO reduction is the highest. It is caused by the mass composition of sewage sludge (and the molar composition of syngas, simultaneously – a low level of nitrogen in sewage sludge and in the syngas). It should be also mentioned that ss2 reaches the highest NO reduction efficiency in higher temperatures in comparison to ss1. It is caused by a higher methane concentration in ss2. Methane needs more time and higher temperatures to decrease NO concentration.
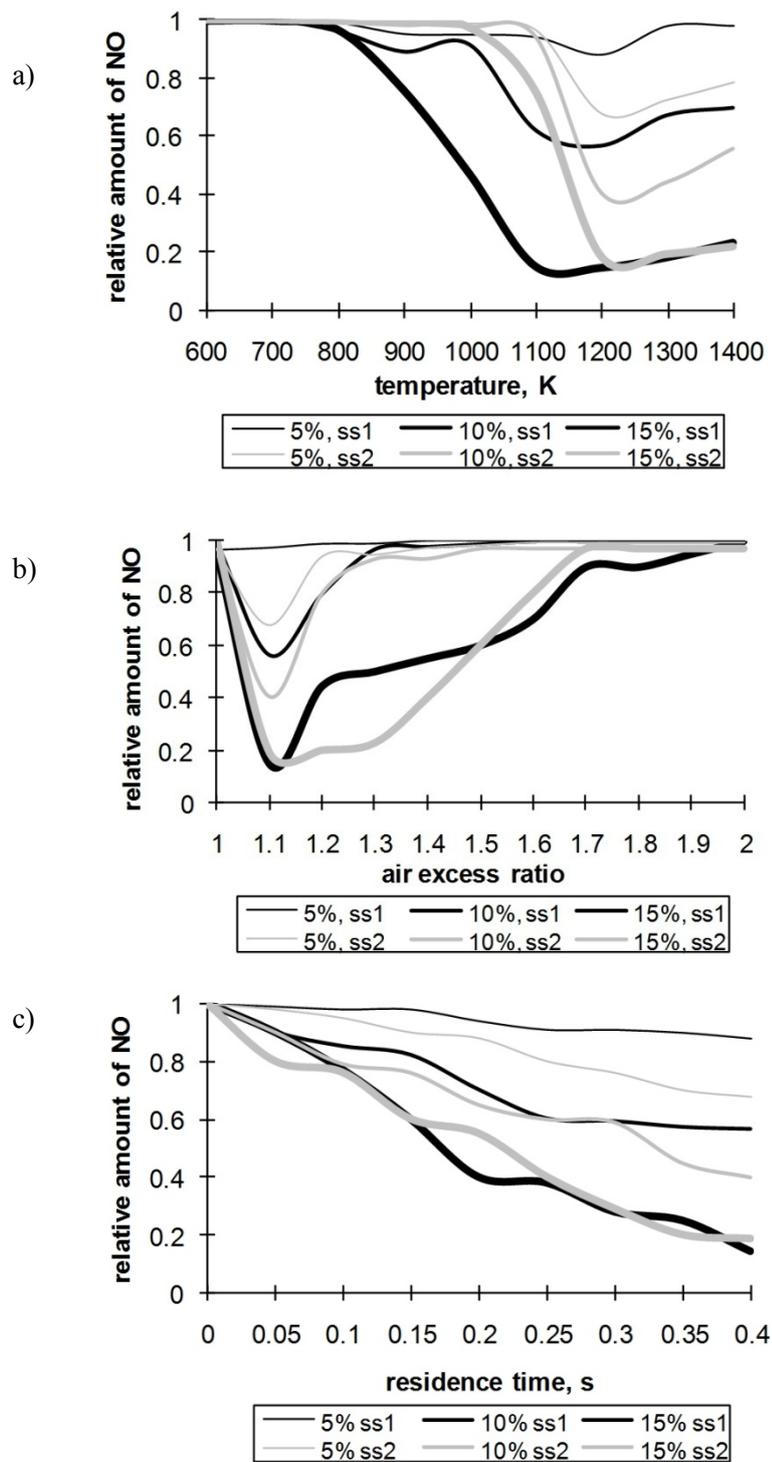
Fig. 7. A comparison of results for two types of sewage sludge gasification gas;
a) function of air excess ratio; b) function of the temperature; c) function of the residence time

5. CONCLUSIONS

- Alternative methods for thermal utilisation of sewage sludge (e.g. gasification process) are an important element in the wider problem of sludge disposal. There are many technologies that use gasification or pyrolysis (or a combination of these two). Their undoubted advantage, in addition to the disposal of sludge, is that it becomes possible to obtain a product that can be effectively used

for the generation of energy. Polish conditions also appear to present a good opportunity to apply this group of waste-disposal technologies.

- Reburning is considered to be one of the most promising and cost-effective NO reduction strategies for coal combustion systems.

- Gas from sewage sludge gasification is a good example of reburning fuel.

- A calculation shows that gas from sewage sludge gasification gives efficiency of up to 90%.

- The optimum value of temperature reburning for the gas from sewage sludge gasification is equal to 1200 K.

- The type of sewage sludge has no marked influence on NO reduction

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