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NITROGEN OXIDES EMISSION REDUCTION USING SEWAGE SLUDGE GASIFICATION GAS REBURNING PROCESS

OBNIŻENIE EMISJI TLENKÓW AZOTU PRZY UŻYCIU PROCESU REBURNINGU GAZEM ZE ZGAZOWANIA OSADÓW ŚCIEKOWYCH

Abstract: Nitrogen oxides can be formed in various combustion systems. Strategies for the control of NO_x emissions in hard coal boilers include the primary (during the combustion) and secondary measures (after combustion). Reburning is the one of the most attractive techniques for reducing NO_x emissions. In the last several years, reburning technology has been widely studied but almost only in power engineering big load boilers. Nevertheless, NO_x emission is an issue that needs to be considered for small capacity boilers as nitrogen oxides resulting from the combustion of any fossil fuels contribute to the formation of acid rain and photochemical smog, which are significant causes of air pollution. Poland is among the largest coal producing country in Europe. Due to this fact, coal fired boilers are very popular in power engineering and also in the municipal sector. Simultaneously, Poland is characterized by the lack of sewage sludge thermal treatment installation. Gasification is considered as a one of the most perspective method of thermal utilization any carbon-containing material. Syngas, which is the main product of gasification, can be used as a supplemental fuel to reduce the consumption of main fuel in boilers, and it has the potential to reduce NO_x emissions. The paper proposes to link those two Polish features so the aim of the work is an experimental investigation of the reburning process of sewage sludge gasification gas in a small capacity domestic coal-fired boiler. The results obtained show how the addition of the reburning fuel influences on NO_x reduction efficiency.

Keywords: gasification, nitrogen oxides emission, reburning process, coal-fired boiler

Introduction

Problem of the sewage sludge management

According to Polish Regulations, sewage sludge (SS) belongs to group No. 19 (the wastes from the treatment installation and the equipment for waste management, waste water treatment plants and from the treatment of drinking water and water for industrial purposes). Sewage sludge, originating from the treatment process of wastewater, is the residue generated during the primary (physical and/or chemical), the secondary (biological) and the tertiary (additional to secondary, often nutrient removal) treatment. An increasing

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tendency has been reported in Poland in terms of the mass of sewage sludge created in municipal waste treatment plants [1].

In 2012, more than 600,000 Mg (on a dry basis, d.b.) of the SS were produced in Poland. Moreover, the National Waste Management Plan for 2014 estimated the quantity of sludge produced in Poland in 2018 will be more than 700,000 Mg (d.b.). Currently in Poland, SS is mainly disposed of by storage or by using it in agricultural applications [2]. However, this is unfavorable because there is no infrastructure for the thermal conversion of SS in Poland. As of January 1st, 2016, the storage of SS with characteristics exceeding those shown in Table 1 will be banned; thus, there is a large need for the development of thermal disposal methods for the treatment of SS. The parameters shown in Table 1 establish the suitability of SS for thermal treatment.

Table 1

Polish criteria for the storage of SS in a non-hazardous waste landfill [2]

	Parameter	The upper value limit
1	Total Organic Carbon (TOC) [% d.b.]	5.0
2	Loss at calcinations (LOC) [% d.b.]	8.0
3	Higher heating value (HHV) [MJ/kg d.b.]	6.0

Gasification as an alternative thermal method of sewage sludge treatment and gaseous fuel production

Gasification is the conversion of solid (or liquid) feedstock into useful and convenient gaseous fuel (or chemical feedstock) that can be burned to release energy or used for production of value-added chemicals [3]. A typical sewage sludge gasification process may include the following steps: (i) drying, (ii) thermal decomposition or pyrolysis, (iii) partial combustion of some gases, vapors and char, (iv) gasification of decomposition products. Gasification requires a gasifying medium like steam, air or oxygen to rearrange the molecular structure of the feedstock in order to convert the solid feedstock into gases or liquids. The main reactions involved during the gasification of organic substances are summarized below [4]:



Gasification is one possible way to use SS and is an attractive alternative compared to other treatment methods. Despite the fact that SS contains phosphorus, nitrogen and, in low concentrations, sulfur, gasification of these components offers several advantages over a traditional combustion process. Gasification takes place in an environment with low levels of oxidizers to prevent the formation of dioxins and large quantities of sulfur and nitrogen oxides [5]. As mentioned previously, sulfur is present in SS, but at low amounts, it is mainly converted to hydrogen sulfide (H_2S) during gasification [6], while the nitrogen is turned into ammonia [5]. It is worth noting that the phosphorus in SS is partitioned into solid (not gaseous) residues [7] and that the volume of produced syngas from SS is low because

gasification requires a fraction of the stoichiometric amount of oxygen necessary for combustion. For all these reasons, gasification requires smaller and less expensive gas cleaning facilities [8]. Syngas is considered a source of environmentally clean fuel and has the potential to become one of the major fuels used in the production of energy. Another possibility is usage this gas as an additional fuel in power engineering to reduce emission of nitrogen oxides from combustion processes (reburning process). The most important advantage of using syngas as such fuel is that it is cheaper than the most popular reburning fuel, natural gas.

Nitrogen oxides chemistry and formation mechanism

Nitrogen oxides are important and complex components in combustion processes. A diagram of nitrogen chemistry found in combustion processes is summarized in Figure 1.

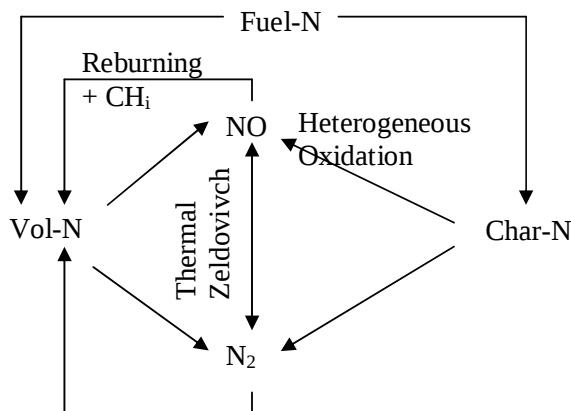


Fig. 1. Overall mechanism of NO formation and reduction in combustion [9]

At present, we may assume there are four different mechanisms for the formation of NO_x:

1. The thermal mechanism
2. The prompt mechanism
3. With formation via N₂O
4. Formation from NO_x fuel

The thermal mechanism for generating NO is a rather well-known and generally accepted mode of generating NO. Generation of NO by this mechanism occurs by oxidation of nitrogen in the air. The rate of these reactions is significant at temperatures greater than 1400°C. These reactions can be described by the extended Zeldovich mechanism [10]:



Considering that a molecule of N₂ is rather stable chemically and does not dissociate easily, reaction (8) requires considerable activation energy. It is an endothermic reaction proceeding with considerable speed at temperatures greater than 1400°C. In comparison

with the exothermic reactions (9) and (10), it is a slow reaction. The Zeldovich mechanism is reversible, and NO is reduced when the equilibrium concentration of NO is low enough and the temperature is high enough for the Zeldovich mechanism to occur [11]. The formation of prompt nitrogen oxides is initiated by the reaction between N_2 and various hydrocarbons (methane, propane, ethylene). Prompt NO is of minor importance and occurs mainly in the combustion of rich mixtures of hydrocarbons and air. Obviously, NO_x emission increases as the nitrogen content in the fuel increases. For gaseous fuels, nitrogen is not chemically bounded with the gas. Therefore, it is generally assumed that during gaseous fuel combustion, no fuel NO is present [12]. The amount of proteinaceous nitrogen decreases with the degree of carbonification of the solid (and liquid) fuel. Fuels with a lower content of nitrogen give lower emissions. Fuel nitrogen is split into a volatile- and char-N during devolatilisation. The volatile-N transforms into NO (or N_2), while char-N reacts through a set of heterogeneous reactions. The formed NO can also be reduced to N_2 . Similar to the formation of prompt NO, NO can be reduced by hydrocarbons to volatile-N under fuel-rich conditions (known as reburning).

Reburning - effective methods of NO_x emission reduction

Today, the reburning process is one of the most attractive techniques for reducing NO_x emissions. In this process, the main part of the fuel (*ie*, 80% of the total chemical energy) is combusted during the first stage (the combustion zone). During the second stage (the reduction zone), only additional fuel is added only to maintain a reductive atmosphere. A reburning fuel produces hydrocarbon radicals, which can interact with NO and therefore reduce the final NO_x emissions. To complete this process, post combustion air is injected to eliminate any unburned hydrocarbons. In the last several years, reburning technology in air combustion has been widely studied [13]. The first researchers to develop this concept and call it 'reburning' were Wendt et al [14]. They suggested the injection of additional fuel along the main flame, so that the emission of NO_x would be minimized by the formation of a reductive atmosphere. This idea was implemented for the first time in Japan, where the concept of reburning was applied to a full-scale boiler [15]. Inspired by these successful experiments and the high efficiency of reburning technology for the reduction of NO_x emissions (by more than 50%), many investigators have conducted bench- and pilot-scale reburning tests. The first laboratory [16] and commercial [17] boilers showed that NO_x emissions may be reduced up to 60%. It was recently been reported by Smoot [18] that many European countries use reburning processes in coal-fired boilers to reduce the NO_x concentration to less than $200 \text{ mg} \cdot \text{m}^{-3}$. Four demonstration plants (mainly using natural gas as a reburning fuel) have been built in the US. Depending on the reburning fuel and the boiler type and scale, NO_x reductions varied from 30% to over 70%. In Europe and Japan, reburning applications include coal-, oil- and gas-fired boilers as well as municipal solid waste (MSW) incinerators. An example of gas reburning in Europe is the 600 MW coal-fired boiler at Longannet et al [19], the largest scale application of gas reburning in Europe. There are also some studies of interest involving reburning in oxy-fuel conditions [13, 20]. The main parameters involved in reburning under oxy-fuel conditions present similar trends to those observed in the literature for reburning in air. In general, reburning fuels, such as natural gas [21], coke-oven gas [18], synthesis gases [22], alcohols [23], coal pyrolysis and synthetic gases [24, 25] and light fuel oils, should be volatile and highly reactive. These fuels should produce large amounts of CH_i radicals while decomposing in

the reduction zone. The results of recent investigations have shown that even heavy fuel oil, lignite [26], some kinds of biomass [21, 27] and some municipal wastes [28] may be good reburning fuels. Nevertheless, to our knowledge, there are no studies that address using gas from the gasification of SS.

The reburning process chemistry is quite complex. It involves the formation and consumption of many different species. Reduction of nitrogen oxides by reburning usually includes the interactions of hydrogen cyanide (HCN), ammonia (NH₃) and NO species [29]. These species interact in a series of reactions and finally end up as NH and N, as shown in Figure 2.

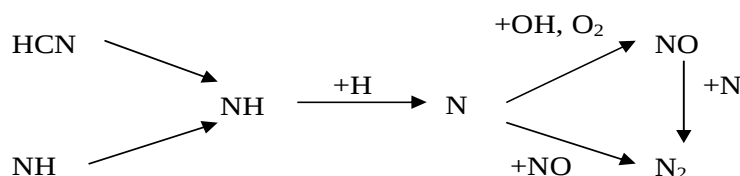


Fig. 2. Reaction-path diagram illustrating the steps involved in the conversion of volatile-N to NO or N₂

For fuel-rich conditions, the formation of hydrogen cyanide depends strongly on the concentration of CH_i species:



Hydrogen cyanide then decays through the pathway $\text{NCO} \rightarrow \text{NH} \rightarrow \text{N}$, as shown in Eqs. (12-15), and ultimately becomes N₂ via a reverse Zeldovich reaction (Eq. 15) [30]:



The paper proposes to link presented features. In one way, Poland is among the largest coal producing country in Europe. Due to this fact, coal fired boilers are very popular in power engineering and also in the municipal sector. Simultaneously, Poland is characterized by the lack of sewage sludge thermal treatment installation. Gasification is considered as an one of the most perspective method of thermal utilization any carbon-containing material and producing fuel which may be used as an additional fuel in coal fired boilers to reduce NO_x emission. So, the aim of the work is an experimental investigation of the reburning process of gas from sewage sludge gasification in a small capacity coal-fired boiler. The results obtained show how the addition of the reburning fuel influence on NO_x reduction efficiency.

Experimental

Sewage sludge gasification equipment facility - reburning fuel production

The granulated sewage sludge sample used in this work was taken from Polish wastewater treatment plant operating in the mechanical-biological-chemical system [31]. Table 2 reports the proximate and ultimate analyses of the sewage sludge.

Table 2

Properties of the sludge gasified	
Parameter	Sewage sludge (SS)
<i>Ultimate analysis [by mass %] (dry basis)</i>	
C	27.72
H	3.81
O	3.59
N	13.53
S	1.81
F	0.003
Cl	0.033
<i>Proximate analysis [by mass %] (as received)</i>	
Moisture	5.30
Ash	44.20
Volatile matter	49.00
LHV [kJ/kg]	10,747

The sewage sludge gasification tests were conducted using a fixed bed gasification facility [32]. A schematic diagram of the system is presented in Figure 3 and photo of the installation on Figure 4.

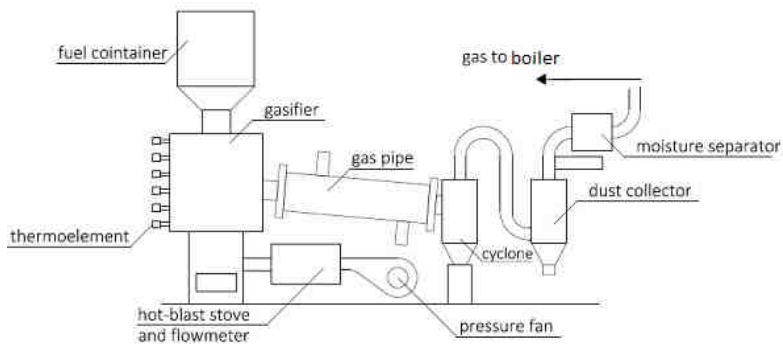


Fig. 3. Fixed bed installation



Fig. 4. Photo of the fixed bed gasifier installation

The main part of the installation was a stainless gasifier 150 mm internal diameter and the total height of 250 mm. For this study, the granular sewage sludge was inserted into the gasifier from the above. Gasification agent was fed from the bottom. The sewage sludge pellets moved in a countercurrent direction to the process gases. There are four specific zones in the gasifier. In the first zone (drying zone) the water from sewage sludge is evaporated. In the second zone (pyrolysis zone) sewage sludge thermal decomposition to volatiles and solid char was carried out. In the one before last zone, carbon was converted, and main combustible components of syngas were produced. In the last zone, the remaining char was combusted. The combustion zone is a source of energy for reactions in the upper zones. There are mainly endothermic reactions. The temperature inside the reactor was measured by six N-type thermoelements. They were located along the vertical axis of the reactor. Additionally, the temperature of the gasification gas at the outlet of the reactor was measured. The flow rate of gasification agent was measured by a flow meter. Syngas was transported from the gasifier by the pipe. The syngas was cleaned by a cyclone, scrubber and drop separator and then added to boiler.

Reburning test - boiler design and set up

A 25 kW coal-fired boiler (Fig. 5) manufactured by a Polish boiler manufacturer was used for the present study. The boiler was equipped with automatic fuel loader and automatic screw feeder. The boiler incorporated a few new design features (*eg* different additional fuel injections) to enable various NO_x reburning tests to be carried out [33] - Figure 6.



Fig. 5. The coal-fired boiler



Fig. 6. Boiler modifications - additional fuel inlets

Several ports on rounded steel element put into the combustion chamber (combustion zone - primary fuel zone) were provided for injecting gaseous reburn fuel. Reburn fuel injected directly into the flue gas stream from the central part of the combustion chamber generates a locally fuel rich zone (reburning zone - additional/secondary fuel zone). The flow rate is controlled using a mass flow controller.

Reburning test - main and additional fuel properties

Polish hard coal serves as the primary fuel. The proximate and ultimate analyses of the coal are given in Table 3. The gaseous reburn fuel is sewage sludge gasification gas. Composition of the reburn fuel is presented in Table 4.

Table 3

Characteristic of main fuel (hard coal)

<i>Proximate analysis [by mass %] (as received)</i>	
Moisture	9.30
Volatile matter	30.60
Ash	4.30
<i>Ultimate analysis [by mass %] (dry basis)</i>	
C	72.36
H	4.34
N	1.35
O (by difference)	21.59
S	0.36
<i>Calorific value</i>	
LHV [kJ/kg] (dry basis)	29,200

Table 4

Characteristic of additional fuel (sewage sludge gasification gas)

[by volume %]					LHV [MJ/m³_n]
CO	H ₂	CH ₄	CO ₂	N ₂	
28.50	5.00	1.00	15.00	50.50	4.49

Results and discussion

Two experiments series were carried out: without reburning (case 1) and with reburning (case 2). Reburning fuel is added to combustion chamber using rounded profile (see Fig. 6 down) with twenty 1 mm holes situated around the axial. The flue gas was continuously sampled using sampling unit. Scrubber, cooling system and conditioning system were used to remove moisture and particulars before gas samples were passed to a Sick Maihak N700 and Rosemount NGA 2000. The Rosemount NGA 2000 analyzer is equipped with a Chemiluminescence detector to measure concentration of NO_x, CO, CO₂ in flue gases. Sick Maihak N700 analyzer to measure the oxygen concentration in flue gas. Data from the analyzers are registered by HP Agilent Data Logger. During the reburn experiment the amount of reburn fuel is defined as a 5% of total flow of chemical fuel energy [34]. Table 5 reports main results. There are presented two cases: without reburning (case 1) and with reburning (case 2). Analyzing presented data considerable reduction of nitric oxides emission (up to 40%) from a domestic/small scale 25 kW coal fired boiler have been achieved by reburning process using sewage sludge gasification gas. Additionally, it

can be seen an increment of CO and CO₂ concentration and decrement of oxygen concentration. The trade-off between NO_x emissions and CO emissions need to be carefully considered when fuel staging is used to control NO_x emissions of domestic/small-scale coal fired boilers.

Table 5

Main results achieved in both analyzed cases

	Case 1	Case 2
Primary fuel flow [kg/h]	4.50	4.00
Additional fuel flow [m ³ /h]	-	2.00
Flue gas [by volume %]		
(6%O ₂)		
CO ₂	10.62	12.50
CO	0.035	0.70
O ₂	10.05	6.00
N ₂	79.30	80.80
NO _x [ppm]		
Range	182.00-244.00	129.00-179.00
Average	213.00	154.00
Boiler capacity [kW]	25.00	25.00

Conclusions

1. Alternative methods for the thermal utilization of sewage sludge (*eg* 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 utilize this group of waste-disposal technologies. Poland is among the largest coal producing country in Europe. Due to this fact, coal fired boilers are very popular in power engineering and also in the municipal sector.
2. Reburning is considered to be one of the most promising and cost-effective NO reduction strategies for coal combustion systems.
3. Gas from sewage sludge gasification is a good example of reburning fuel.
4. Considerable reduction of nitric oxides emission from a domestic/small scale 25 kW coal fired boiler have been achieved by reburning process using sewage sludge gasification gas. The application of the additional fuel inlets above the fuel bed was strongly affected the nitric oxides emissions.

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OBNIŻENIE EMISJI TLENKÓW AZOTU PRZY UŻYCIU PROCESU REBURNINGU GAZEM ZE ZGAZOWANIA OSADÓW ŚCIEKOWYCH

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Abstrakt: Tlenki azotu mogą powstawać w wielu systemach spalania. Strategia ograniczania emisji tlenków azotu w kotłach węglowych obejmuje metody pierwotne (w trakcie spalania) oraz metody wtórne (po spalaniu). Reburning jest jedną z najbardziej atrakcyjnych metod ograniczania emisji tlenków azotu. W ostatnich latach technologia ta była przedmiotem wielu badań, ale głównie w kotłach energetycznych dużej mocy. Niemniej jednak, emisja tlenków azotu dotyczy także kotłów mniejszych, gdyż stanowi ona istotne źródło powstawania kwaśnych deszczów i smogu. Polska należy do grupy krajów europejskich, gdzie węgla wydobywa się (i zużywa) dużo. Z tego powodu kotły węglowe są bardzo popularnymi jednostkami wytwórczymi. Jednocześnie Polska charakteryzuje się brakiem instalacji do termicznego zagospodarowania osadów ściekowych. Zgazowanie jest wymieniane jako jedna z najbardziej perspektywicznych metod przetwarzania substancji zawierającej węgiel, a produkowany w tym procesie gaz może być używany jako paliwo dodatkowe potencjalnie obniżające emisję tlenków azotu. W pracy zaprezentowano techniczne możliwości połączenia tych dwóch typowych dla Polski cech. Celem pracy jest zatem eksperymentalna analiza możliwości obniżenia emisji tlenków azotu z procesu spalania węgla w kotle małej mocy przy zastosowaniu gazu ze zgazowania osadów ściekowych. Wyniki pokazują, jak dodatek paliwa gazowego wpływa na efektywność redukcji NO_x .

Słowa kluczowe: zgazowanie, emisja tlenków azotu, reburning, kocioł węglowy