

ARCHIVES OF ENVIRONMENTAL PROTECTION

vol. 38

no. 3

pp. 81 - 89

2012



PL ISSN 2083-4772

DOI: 10.2478/v10265-012-0027-3

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Zabrze, Poland 2012

POSSIBILITY OF NO_x EMISSION REDUCTION
FROM COMBUSTION PROCESS USING SEWAGE SLUDGE
GASIFICATION GAS AS AN ADDITIONAL FUEL

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Keywords: Sewage sludge, nitrogen oxides, gasification gas, additional fuel.

Abstract: In the paper, a numerical simulation of the co-combustion process of sewage sludge gasification syngas in a hard coal-fired boiler was done. Two different syngases (SS1 and SS2) were taken in consideration. Additional (reburning) fuel was injected into the combustion chamber, which was modeled as a plug flow reactor (PFR). The molar flow rates ratio of reburning fuel is assumed to be 5.0%, 7.5%, 10.0%, 12.5% and 15.0% of the whole exhaust. The simulations were conducted for constant pressure equal to 1atm for temperatures range from 600 to 1400 K. It was assumed that a flue gases which enters the reburning zone contains 300 ppm of NO and that during combustion only NO is formed without other NO_x. Results show that that gas from sewage sludge gasification gives reburning efficiency of up to 90%. Calculation shows also an optimum value of temperature reburning for gas from sewage sludge gasification which is equal to 1200 K. The type of the sewage sludge has no strong influence on the NO reduction.

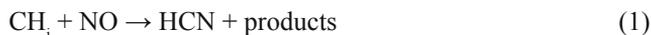
INTRODUCTION

Sewage sludge, originating from the treatment process of waste water, 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 tendency has been reported in Poland in terms of the mass of sewage sludge created in municipal waste treatment plants [7]. In Europe, dry weight per capita production of sewage sludge resulting from primary, secondary and tertiary treatment is an average of 90 g per person per day [6]. Additionally, the implementation of the Urban Waste Water Treatment Directive (1991) (UWWTD) leads to 50% increase in sludge production by year 2005, i.e. 10 million tons annually. The European Union's target is to reduce final waste disposal by 50% by 2050 compared with 2000 [16]. To do this, it has drawn up a strategy of thermal methods of sewage sludge disposal development. During the last twenty years there has been a major change in the ways sludge is disposed [19]. European legislation prohibits the landfill and water deposits of sewage sludge. The latest trends in the field of sludge management, i.e. combustion, pyrolysis, gasification and co-combustion, have generated significant interest of scientists [29].

Gasification has attracted a considerable interest from water utilities as an alternative technology with the advantage of destruction of pathogenic bacteria and volume reduction, and the additional benefits of energy recovery and lower-cost atmospheric emission control. Gasification converts any carbon-containing material into synthesis gas composed primarily of carbon monoxide and hydrogen (referred to as syngas). The efficiency of the gasification process is better than that the incineration, in principle, because produced gas can be used directly in power generation process. Syngas can also be used as a supplemental fuel to reduce the consumption of fossil fuels, and it has the potential to reduce NO_x emissions. In Polish energy sector, syngas can be used as a reburning fuel while reducing NO_x emissions at the same time.

Emission of nitrogen oxides by combustion processes is an important factor in the formation of urban smog, in which ozone and other photo-oxidants constitute a serious health hazard. Nitrogen oxides consist of nitric oxide (NO), nitrogen dioxide (NO_2), and nitrous oxide (N_2O). NO and NO_2 are collectively referred to as NO_x . Combustion systems commonly utilize fossil fuels such as gas, petroleum and coal. Given the fact that Polish power industry is based on hard and brown coal and air pollution has a transboundary character, the significance of the undertaken investigations goes beyond the domestic issues [15]. Coal is generally burned in fixed-bed, fluidized-bed or entrained – flow combustors. Several techniques have been developed to reduce NO_x emissions in coal combustion systems. One of them is fuel staging (reburning). 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 (i.e., 80% of the total chemical energy) is combusted during the first stage (the combustion zone). During the second stage (the reduction zone), 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. 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 (NH_3) and NO species [23].

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. (2–5), and ultimately becomes N_2 via a reverse Zeldovich reaction [13]:



In the last several years, reburning technology in air combustion has been widely studied [11]. The first laboratory [9] and commercial [8] boilers showed that NO_x emissions may be reduced up to 60%. In general, reburning fuels, such as natural gas [2], coke-oven gas [24], synthesis gases [10], alcohols [1], coal pyrolysis and synthetic gases [22, 25] and light fuel oils, should be volatile and highly reactive. These fuels should produce large amounts of CH₁ radicals while decomposing in the reduction zone. The results of recent investigations have shown that even heavy fuel oil, lignite [12], some kinds of biomass [2, 5] and some municipal wastes [18] may be good reburning fuels.

The aim of the work is a numerical simulation of the reburning process of gas from sewage sludge gasification in a coal-fired boiler. All calculations were performed using the Chemkin program managed and distributed exclusively by San Diego-based Reaction Design (USA), and a plug flow reactor (PFR) model was used. The reburning potential of syngas is defined, and the influence of the reaction between the reburning fuel and flue gas from coal combustion on the reduction of NO_x emissions has also been examined.

CALCULATION

In the calculations, two gases from the sewage sludge gasification process (SS1 and SS2) were used as the reburning fuel. Calculation of the gas composition from sewage sludge gasification was performed using a non-stoichiometric approach equilibrium model [30]. This model is based on minimizing the Gibbs free energy in the system [3]. The properties of the syngas depend on the type of gasifier used [17]. It was assumed that the gasification process was carried out in a downdraft fixed-bed gasifier. In this way, the amount of tar (and particulate) at the end of the process is very low because most of it is combusted in the oxidation zone [20, 21]. Thus, tars are ignored in the model, and the only species created during the process are CO, CO₂, H₂O, H₂, CH₄, N₂ and O₂. Additionally it was assumed that the residence time of the reactants in the reactor is long enough to achieve an equilibrium, which has also been confirmed by Zainal *et al.* [31]. A second assumption was that all of the carbon in the sewage sludge is gasified; meaning the formation of charcoal can be neglected. Moreover, as noted by Puig-Arnavat *et al.* [20], the only species present at concentrations higher than 10⁻⁴ mol% after gasification in a downdraft fixed-bed gasifier between 600 and 1500 K are CO, CO₂, CH₄, H₂, N₂, O₂ and H₂O.

The molar composition of reburning fuels is presented in Table 1.

Table 1. Molar composition of analyzed sewage sludges

Compound, vol.%	Reburning fuels	
	Syngas1 (SS1)	Syngas2 (SS2)
H ₂	0.11	0.06
CO	0.24	0.27
CH ₄	0.02	0.09
N ₂	0.46	0.41
O ₂	0.04	0.04
CO ₂	0.09	0.09
H ₂ O	0.04	0.04

During simulations, the combustion of hard coal was assumed to be complete. The mass fraction of the main components of coal was as follows: $c = 0.650$; $h = 0.048$; $s = 0.004$; $o = 0.128$; $n = 0.017$; $w = 0.100$. Reburning fuel was injected into the combustion chamber, which was modeled as a plug flow reactor (PFR). The scheme of the analyzed process is shown in Figure 1.

In such reactors, an important parameter is the reactor length and the reactant flow (which is important so that the reactants do not mix and diffuse). It should be mentioned that plug flow can be used to analyse processes where mass transfer and heat are smaller in comparison to conductive heat transfer [26].

The calculations were modeled using the GRI-Mech 2.11 mechanism. This version is used for analyzing the reburning process. The molar flow rate ratio of the reburning fuel to the whole exhaust was assumed to be 5.0%, 7.5%, 10.0%, 12.5% and 15.0%. The simulations were conducted at a constant pressure equal to 1atm and at temperatures ranging from 600 to 1400 K, with typical residence times of 0.0–0.4 s [4] and local air excess ratio ranging from 1.0 to 2.0. The local air excess ratio in this paper is defined as the weight ratio of air to unreacted fuel used in the entrance of the reduction zone divided by the fuel ratio for stoichiometric combustion [14].

The residence time of the reactants in the reburning zone is a very important factor. In a real boiler, the residence time includes the mixing time between the products coming from the primary zone and the reburning fuel, and the reaction time of those products. It was assumed that all reactants enter the reactor already premixed, so the residence time is taken as the reaction time. It was assumed that the flue gases entering the reburning zone contain 300 ppm NO and that during combustion only NO is formed without other NO_x [27, 28].

RESULTS

The effectiveness of the reburning process depends on the conditions in the reducing zone, which control the interaction between hydrocarbon radicals and NO. The major variables in this zone are the amount of hydrocarbons introduced as reburning fuel, the temperature and the amount of oxygen entering the reburnig zone. The results obtained show how the NO_x reduction efficiency depends on the injection reburning fuel ratio, air excess ratio and the temperature of the reburning process. Figure 2 shows the dependence of the calculated NO_x reduction efficiency as a function of injection reburning fuel ratio.

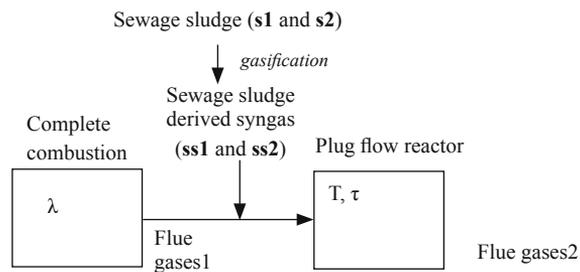


Fig. 1. Scheme of the analyzed system

Analyzing this Figure, it can be said that increment of the ratio of reburning fuel leads to improve the NO_x reduction. The best efficiency (almost 90%) was obtained at the molar rate of reburning fuel of 15%. This efficiency for NO reduction would be considered excellent in a real-world system. Generally, it can be noticed that reburning fuel SS2 is characterized by the higher reduction efficiency than SS1.

In Figure 3, NO_x reduction efficiency as a function of temperature in the reburning zone is shown. NO reduction efficiency of the SS gasification gas is strongly dependent on temperature. Temperature in the reburning zone is a key variable in the process because it determines the degree of conversion of the reburning fuel and thus the formation of hydrocarbon radicals, as well as the rates of important reactions involved in the reburning mechanism. The reduction of NO increases with increasing temperature – resumably through its reaction with CH_i – until it reaches a maximum. At temperatures between 600 to 800 K, the SS-derived syngas does not affect the NO concentration. At higher temperatures, the reburning efficiency of the analysed reburning fuel increases. The strongest NO reduction occurs at temperatures higher than 1000 K; however, the best temperature for the reburning process is 1200 K. In general, as seen in Figure 3, increasing the molar ratio of reburning fuel results in a decrease in the temperature for optimal NO reduction.

Figure 4 shows the dependence of NO reduction efficiency on the air excess ratio for a reburning zone temperature and residence time of 1200 K and 0.4 s. As shown in this figure, an air excess ratio equal to 1.0 reburning fuel has no influence on the reduction of NO. The reduction of NO increases up to a maximum as the stoichiometry becomes more fuel-rich. Such conditions favour the formation of CO and CO₂ via the oxidation mechanism of the reburning fuel instead of the formation of hydrocarbon radicals active in NO reduction.

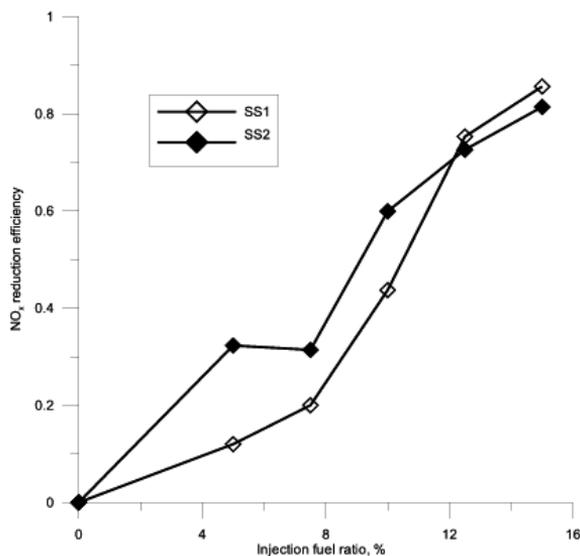


Fig. 2. NO_x reduction efficiency as a function of injection fuel ratio; 1200 K, $\lambda = 1.1$, $\tau = 0.4$ s

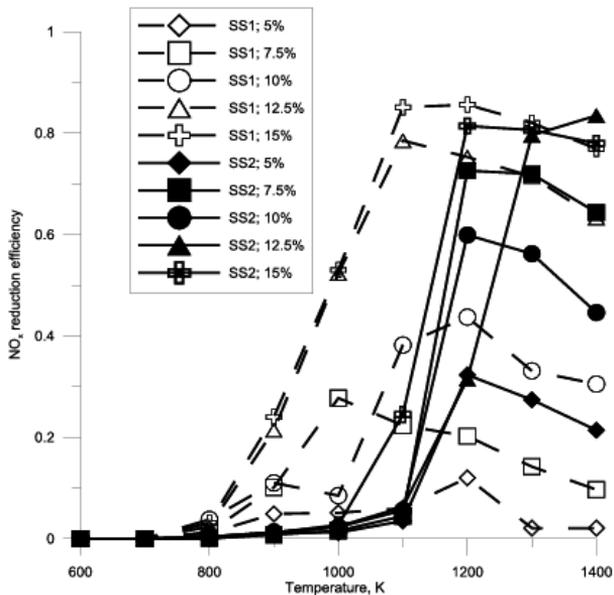


Fig. 3. NO_x reduction efficiency as a function of temperature in the reburning zone for different values of the molar ratio of reburning fuel; $\lambda = 1.1$, $\tau = 0.4$ s

For each molar ratio of reburning fuel, the relative amount of NO initially decreases at the beginning as the air excess ratio begins to increase. As seen in this figure, there is a value of the air excess ratio for which the efficiency of the NO reduction reaches a maximum. Above this value, NO reduction decreases.

The type of SS-derived syngas has no influence on the efficiency of NO reduction (in that case, both syngas types are characterised by more or less the same value of NO reduction). SS2 syngas is characterised by a lower molar concentration of nitrogen and higher values of carbon monoxide and methane. Despite similarities in NO reduction efficiencies of SS1 and SS2 syngas for a molar fraction of reburning fuel of 15%, there is a much wider range of air excess ratios for SS2 gas where NO reduction is highest. This is due to the fact that there is simultaneously a small level of nitrogen in SS and in the syngas. In addition to having a wider working range for the air excess ratio, SS2 gave higher NO reduction efficiency at higher temperatures as compared to SS1. This result can be explained by the fact that SS2 had a higher methane concentration than SS1. Methane needs more time and higher temperatures to decrease NO concentration.

CONCLUSIONS

Alternative methods for the thermal utilization 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

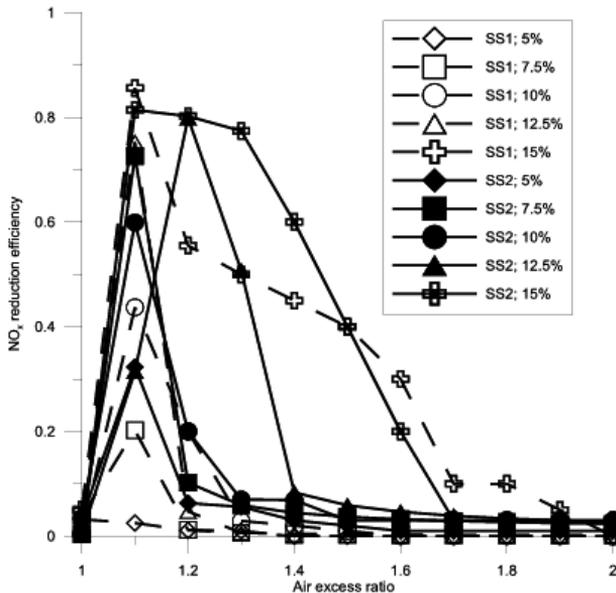


Fig. 4. NO_x reduction efficiency as a function air excess ratio in the reburning zone for different values of the molar ratio of reburning fuel; T = 1200 K; τ = 0.4 s

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.

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%.

Calculation shows also an optimum value of temperature reburning for gas from sewage sludge gasification which is equal to 1200 K.

The type of the sewage sludge has no strong influence on the NO reduction.

ACKNOWLEDGMENTS

The paper has been prepared within the financial support from the National Science Centre under grant no. N N523 737540

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MOŻLIWOŚĆ OBNIŻENIA EMISJI NO_x Z PROCESÓW SPALANIA PRZY UŻYCIU GAZU ZE ZGAZOWANIA OSADÓW ŚCIEKOWYCH JAKO PALIWA DODATKOWEGO

W pracy przedstawiono wyniki symulacji numerycznych procesu współspalania gazu ze zgazowania osadów ściekowych w kotłach węglowych. Do analizy użyto dwóch rodzajów gazu ze zgazowania osadów ściekowych (SS1 i SS2). Dodatkowe paliwo zostało doprowadzone do komory spalania, która została zamodelowana jako reaktor przepływowy (PFR). Udział molowy dodatkowego paliwa wynosił 5.0%, 7.5%, 10.0%, 12.5% i 15.0% całkowitego strumienia spalin. Symulacje zostały przeprowadzone przy ciśnieniu otoczenia oraz w zakresie temperatur 600–1400 K. Założono, iż w chwili początkowej w strefie doprowadzenia dodatkowego paliwa stężenie tlenków azotu wynosi 300 ppm i jedyne tlenki azotu, które się tworzą podczas spalania to NO. Wyniki pokazują, iż zastosowanie gazu ze zgazowania osadów ściekowych jako paliwa dodatkowego (reburningowego) pozwala na ograniczenie emisji tlenków azotu nawet o 90%. Jednocześnie stwierdzono, iż najefektywniejszą redukcję uzyskano przy temperaturze równej 1200 K. Nie stwierdzono istotnego wpływu rodzaju gazu ze zgazowania osadów ściekowych na efektywność obniżenia tlenków azotu.

