Gaseous Emissions from the Fluidized-Bed Incineration of Sewage Sludge*

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Combustion of dried sewage sludge has been studied in a bench-scale fluidized-bed combustor operated under steady-state conditions. The attention has been focused on the emissions of pollutant gases and on the unburned carbon in elutriated fines. Steady-state combustion experiments were carried out at different temperatures by feeding pre-dried sludge particles into an inert bed of ceramsite particles. The 9.4 cm i.d. combustor was operated at superficial gas velocities of 0.3—0.8 m s^{-1} and at different excesses of air. Very small amounts of unburned carbon in the fly ash separated by the cyclone (below 0.8 mass %) and generally very low levels of carbon monoxide in the exit flue gas (usually below 50 ppm) document very good combustion efficiency attained in a bubbling (dense) fluidized bed. Although conversions of the fuel nitrogen, measured under practical conditions, to NO_x and N_2O amounted to only about 10 % and 5 %, respectively, the resulting harmful emissions must be of serious concern. The measured content of NO_x varied between approximately 600 ppm and 1100 ppm, in the case of N_2O it was 150—400 ppm.

The emissions of CO and NO_x are closely interdependent and connected: the lower concentration of CO is in flue gas, the higher level of NO_x measured. Higher operating temperatures, higher partial pressures of oxygen, and longer residence times in the combustor improved the combustion efficiency, according to the amount of unburned CO, C_xH_y, and carbon. Unfortunately, such process conditions inherently lead to unwanted higher conversions of the fuel-bound nitrogen-to-nitrogen oxides.

Sewage sludge is a result of urban wastewater treatment and among municipal and industrial wastes represents an increasing problem due to the expected larger production and the progressive restrictions of the conventional disposal options. Four disposal methods are currently used: recycling in agriculture, landfilling, dumping into sea, and combustion. Agricultural utilization is hindered by the presence of heavy metals and organic micropollutants. Landfilling and dumping into sea are not considered any more environmentally sustainable due to the liquid and gaseous emissions in soil, water, and air. Combustion is in most cases the only practicable; however, costs of sludge incineration make such alternative not competitive when cake concentration is too low (< 30 %). Various issues related to combustion of sewage sludge have been recently summarized and discussed in [1, 2]. In general, waste combustion has grown in interest for the possibilities of safe disposal linked to energy recovery of a wide range of alternative combustible materials, including biomass, peat, municipal, agricultural, and industrial wastes. Fluidized-bed reactors have been indicated as one of the best options, due to their flexibility and the achievement of an efficient and clean combustion and/or gasification process of different combustible materials, used either alone or in combination with fossil fuels [3—6].

Success of the fluidized-bed combustion technology for sewage sludge can also be attributed to the great volume reduction of final ashes and to the efficient destruction of persistent organic substances present in sludge. A peculiar characteristic of sewage sludge includes high contents of moisture, volatile matter, ash, and that of organic nitrogen [7—9].

Main aim of this work was to determine an interrelationship between the concentrations of carbon monoxide and those of NO_x in flue gas exiting a fluidized-bed reactor incinerating pre-dried sewage sludge.

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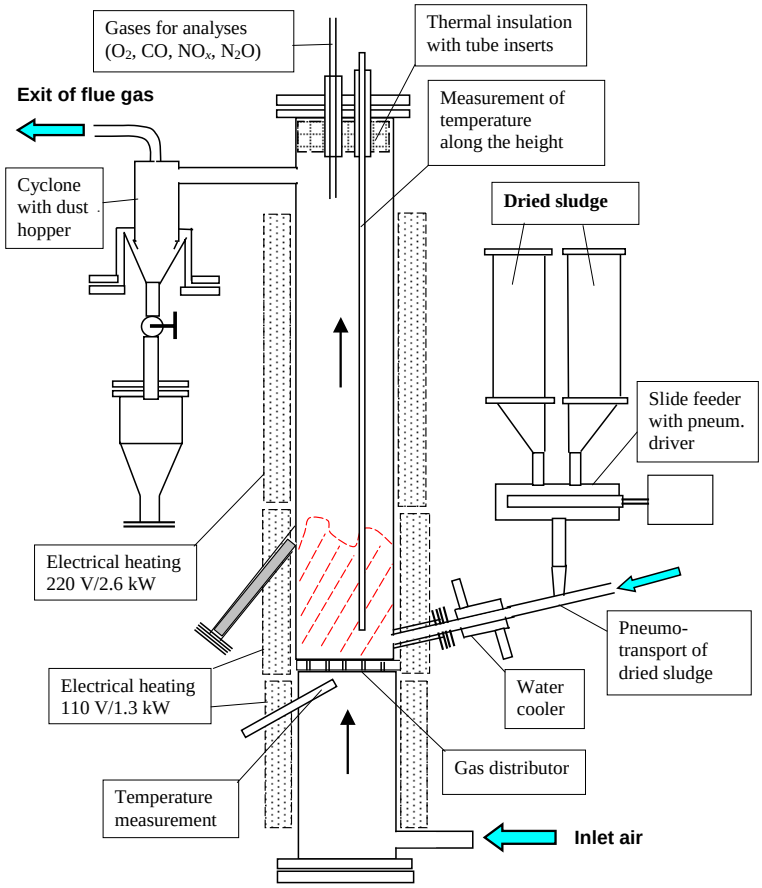

Fig. 1. Experimental set-up for fluidized-bed combustion of dried sewage sludge.

EXPERIMENTAL

A batch of digested (stabilized) sewage sludge was secured from a large wastewater plant. In its original, mechanically dewatered state, it contained 65.4 mass % of water. The samples were dried at 105 °C, then crushed in a ball mill and sieved to 0.5—1.6 mm large particles. Results of the proximate (fuel) and ultimate (elemental) analyses of the sludge are given in Table 1. Scheme of the combustor set-up used in this study is shown in Fig. 1. The fluidized-bed reactor can be operated as a batch or through-flow reactor with respect to the solids. Flow rate of the air entering the reactor is controlled by mass flow controllers.

The combustion air introduced into the reactor can be specified as follows: the air relative humidity, $\varphi = 0.8$; the mole fraction of water vapour in air, $y^{\mathrm{a}}_{\mathrm{H_2O}} = 0.020053$ ($Y^{\mathrm{a}} = 0.097678$), the molar mass of wet air, $M^{\mathrm{w}}_{\mathrm{a}} = 28.361$ g mol^{-1}; temperature 20 °C; pressure 101.325 kPa.

Generated flue gas can be cooled and cleaned when needed. Cylindrical reactor tube was made from heat-resistant alloy (< 1100 °C) with an internal diameter of 93.6 mm. Height of the reactor tube above the gas distributor was 980 mm. Air preheated in the bottom part of the reactor is introduced into the bed through an 8 mm thick perforated plate distributor with 3/1-mm-diameter conical holes drilled on the 7 mm × 7 mm rectangular pitch. Similarly as the reactor tube, all other parts of the reactor were constructed of a heat-resistant alloy.

Table 1. Specifications of Dried Stabilized Sewage Sludge

Species	Concentration	
	w_i	ν_i/(mol g^{-1})
C	0.1440	0.01199
H	0.0207	0.02054
N	0.0163	1.163×10^{-3}
S	0.0026	8.11×10^{-5}
Cl	5×10^{-4}	1.41×10^{-5}
O	0.1260	7.875×10^{-3}
Moisture, w_{f}	0.0120	6.661×10^{-4}
Ash	0.6779	
Volatile matter	0.2869	
Fixed carbon	0.0232	
H^{L}/(MJ kg^{-1})	6.73	
d_{p}/mm	0.25—0.50	

Temperature was measured at different spots (including the cyclone) along the height of the reactor by means of thermocouples (Ni—CrNi and Pt—PtRh) placed in shielding tubes. Preheater and reactor tubes

Table 2. General Operation Conditions of the Fluidized-Bed Combustor

Parameter	Value
d_p/mm	0.25—0.50
m_b/g	252
H_f/cm	12
U/(m s^{-1})	0.30
τ_b/s	$\approx 10^{-1}$
τ_r/s	1—2
G/(g h^{-1})	850

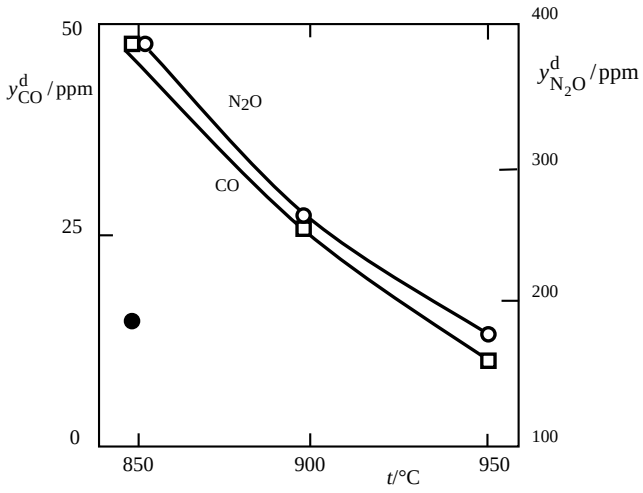

Fig. 2. Influence of the fluidized-bed temperature on the concentration of carbon monoxide (□) and nitrous oxide (○) in exit flue gas, freeboard temperature 840—847 °C, oxygen concentration, $y^d_{O_2} = 0.067$—0.073, excess gas velocity, $U - U_{mf} = 0.270$ m s^{-1}. The plotted values have been recalculated for $y^d_{O_2} = 0.06$. Comparison with nitrous oxide concentration (●) obtained by *Werther et al.* [12] at similar combustion conditions.

were heated by three independent electrical external coils. This makes it possible to control the respective temperatures of the inlet air, the dense fluidized bed, and the freeboard region independently.

Double-acting, air-driven slide feeder with two containers was used to feed the particulate, dried sewage sludge into the high-temperature fluidized bed of inert ceramsite particles. Sliding plate was 10 mm thick, constructed of polytetrafluoroethylene, and provided with two 10-mm-diameter holes. Frequency of dosage was varied to achieve a desired rate of feeding. For further details about the feeding system, the reader is referred to recently published articles [10, 11]. It should be noted that the solid fuel was introduced to the very bottom of the fluidized bed in a cooled tube. The level of the fuel inlet nozzle was about 15 mm above the gas distributor plate. General operation conditions of the fluidized bed combustor are stated in Table 2.

Reactor was fully instrumented to determine the required oxygen, CO, NO, NO_2, and N_2O concentrations in flue gas. The gas samples were withdrawn at the reactor outlet by quartz and Teflon tubes. Water vapour was removed from the gas stream to be analyzed in a condenser cooled in the ice-water bath. Traces of ammonia and amines were removed by absorption in an aqueous solution of phosphoric acid ($\approx$ 30 mass %). Deeper removal of water vapours was achieved in a flask filled with $CaCl_2$. After cleaning the sampled gas by special Teflon filters, gas stream was continuously analyzed. A chemiluminescent analyzer (model 951A Beckman) was used for the determination of NO and NO_2 concentrations. Oxygen content in a flue gas was measured by a paramagnetic analyzer (Siemens Oxymat 5M). An IR, cross interference compensating, Hartmann & Braun analyzer (Uras – Advance Optima System) was employed to determine the CO and N_2O concentrations in the exiting flue gas.

RESULTS AND DISCUSSION

On entering the hot bed, the sludge particles start to devolatilize immediately. Products of the fuel pyrolysis (devolatilization) are quickly converted by oxidation reactions to mainly CO_2 and H_2O, forming some CO depending on the operating conditions and leaving a very small amount of hydrocarbons unconverted, usually taken as C_xH_y or CH_4. It was estimated that more than 80 % of the sludge carbon was released with the volatiles. Therefore, the carbon (char) concentration in the bed is very low and carbon monoxide can be assumed as a reliable indicator of combustion/oxidation mechanism.

No flames were observed in the freeboard region above the bed. Sludge particles well mixed with the bed solids were rapidly moving within the bed. In general, their temperature was significantly higher than that of the ceramsite bed. Since the volatiles account for a very high proportion of the heating value of the sludge, they are an important factor in the overall combustion process. To ensure the volatile components were burned in a controlled manner within the bed, the sludge was introduced through a tube situated in the bed wall very close to the distributor. Volatile components burn very fast in a diffusion flame surrounding the particle. As apparent, an adequate inflow of diffusing oxygen into a reaction zone is a prerequisite.

The effect of the fluidized bed temperature on CO levels in the exit gas is shown in Fig. 2. As expected, higher bed temperatures lead to lower CO emissions. It should be noted that the attained CO concentrations are generally very low. This indicates that the combustion of dried sludge in the bubbling fluidized bed is a very efficient oxidation process. An additional significant improvement in the oxidation efficiency of the combustor can be achieved by a careful control of the freeboard temperature. Fig. 3 illustrates the

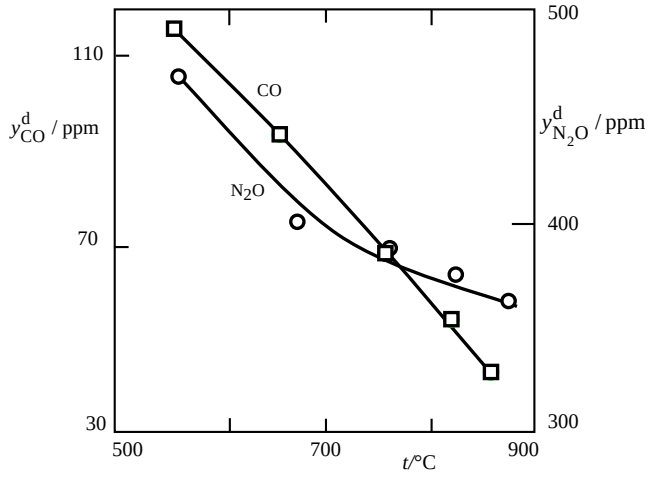

Fig. 3. Effect of the freeboard temperature on the concentration of carbon monoxide (□) and nitrous oxide (○) in exit flue gas, fluidized bed temperature 850—860 °C, excess gas velocity, $U - U_{mf} = 0.270$ m s^{-1}, oxygen concentration, $y^{d}_{O_2} = 0.067$—0.073. The plotted values have been recalculated for $y^{d}_{O_2} = 0.06$.

favourable influence of increased temperature in the freeboard region upon the CO concentrations in the flue gas leaving the reactor. Although intensive combustion occurs within the bed, the afterburning of the volatiles can also take place to a lesser extent in the freeboard region. Carbon monoxide is the main combustible carbon species in the freeboard and its oxidation to CO_2 is the most significant postcombustion reaction occurring in this afterburner space.

Any assessment of the combustion efficiency must include also the unburned fixed carbon present in a bottom ash accumulated in the bed and that in fly ash elutriated from the reactor and captured by means of a cyclone and filters. The ash mass balance showed that about 95 % of the ash present in the sludge was retained in the bed and the cyclone. The amount of fly ash was much less than that of bottom ash by a factor of about 0.1. No unburned carbon was detected in the bottom ash, less than 0.8 mass % of unburned carbon was found in the fly ash, which is an undisputable low figure. The content of the ceramsite dust formed by attrition and contained in the cyclone fines was less than 15 mass %.

Our findings prove that laboratory-scale, fluidized-bed combustors are also capable of attaining high combustion efficiency provided that an effective configuration of the reactor is employed and favourable operating conditions are secured.

It is common knowledge that during combustion increasingly important emissions of nitrous oxide occur in fuel-lean mixtures and at low temperatures and elevated pressures. Thus, low-temperature (750—900 °C) fluidized-bed combustion of fuels containing organically bound nitrogen presents a significant source of N_2O. Its emissions contribute to both the greenhouse effect and the depletion of the stratospheric ozone layer. Nitrous oxide is temperature-sensitive and tends to decompose to N_2 at temperatures higher than 900—950 °C.

The influence of the fluidized-bed temperature and the freeboard temperature on the N_2O content in flue gas was explored for the oxygen concentrations varying from 6.9 vol. % to 7.3 vol. %. All values of the N_2O concentrations were recomputed for the O_2 amount of 6 vol. % in a dry flue gas and in such form are presented in Figs. 2 and 3. As apparent, the higher temperatures are in the combustor, the lower amount of N_2O is present in the exit gas. As demonstrated in Figs. 2 and 3, raising the bed and freeboard temperature proves to be an effective measure in reducing both CO and N_2O concentrations in the flue gas. This indicates that the freeboard region works as an additional reaction space where postcombustion reactions proceed to a significant extent.

Although nitrous oxide is considered to be one of the most important oxides of nitrogen in the atmosphere, relevant data on its emissions from sludge combustion are scarce in the literature. As shown in Fig. 2, the amount of N_2O emissions obtained by *Werther et al.* [12] for a dried sewage sludge under similar conditions was about half of that found in this study.

Nitric oxide (NO) and nitrogen dioxide (NO_2), which is more toxic and irritating than NO, are usually lumped together under the designation of NO_x. Experimental data indicate that only about 9.0—9.5 % of the overall NO_x emissions are constituted by NO_2 at the oxygen concentrations in dry flue gas lower than 6 vol. % and at combustion temperatures above 800 °C. In the light of this, the chemical formula used in computations throughout this work to approximately describe nitrogen oxides is $NO_{1.1}$.

The influence of temperature in the freeboard region on NO_x emissions is exposed in Fig. 4 for the bed temperatures between 850 °C and 865 °C. It can clearly be seen that the higher the freeboard temperature, the higher NO_x concentrations were found in the exit flue gas. Moreover, fairly low conversion efficiency of fuel-bound nitrogen to NO_x was observed, increasing this value with the freeboard temperature from about 6 % to approximately 8.5 %.

Trends for the depletion of CO and the NO_x formation depend on the oxidation kinetics, which is enhanced by temperature. The freeboard gas phase postcombustion/oxidation is a very important process, which must always be considered, particularly in the case of sludge combustion. CO emissions can be lowered to negligible levels when the freeboard temperature is raised, however, at the expense of increasing unwanted NO_x content. The way, the CO concentrations and the NO_x levels are inherently interrelated and connected in flue gas, is visualized in Fig. 5. Also shown in this figure are the experimental results of *Cammarota* and *Chirone* [2]. As can

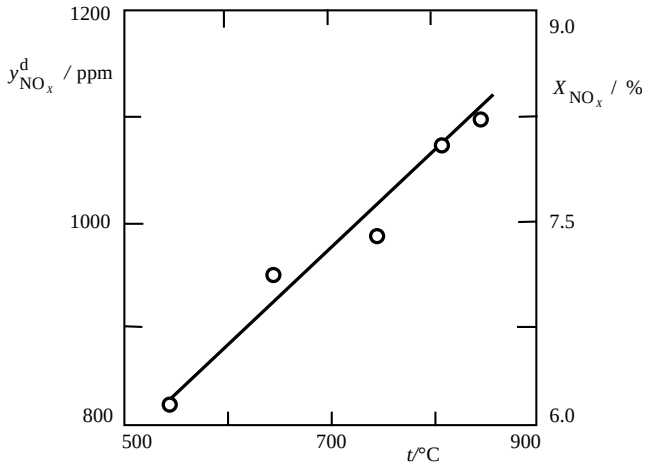

Fig. 4. Effect of the freeboard temperature on the NO_x concentration in exit flue gas. The experimental conditions were the same as those indicated in Fig. 2.

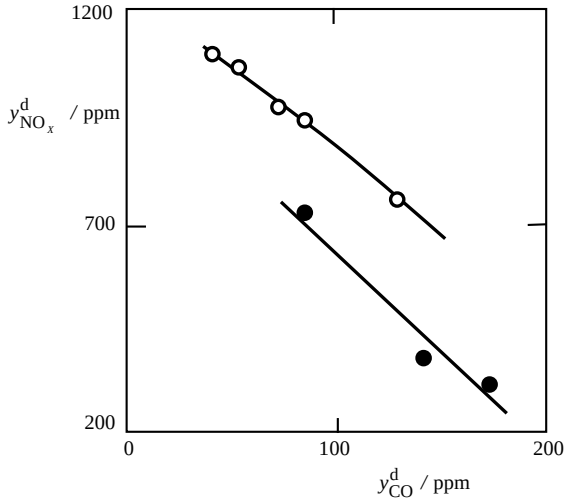

Fig. 5. Correlation between CO and NO_x emissions contents in exit flue gas: (○) this paper; (●) Ref. [2].

be seen, they exhibit a very similar trend as do our data.

Air staging (particularly staged co-combustion), which is known to be an effective means to control the NO_x and N_2O content in a flue gas from coal combustion, is based on the above fact that CO effectively hinders the fuel-bound nitrogen oxidation [8, 9, 13—15]. A marked reduction of NO_x content can also be achieved by flue gas recycling (FGR) or injecting ammonia or urea into the freeboard (selective noncatalytic reduction, SNCR). However, certain adverse effect on the CO emissions accompanying these anti-NO_x measures cannot be ruled out.

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SYMBOLS

d_p	solids particle size determined by sieving	mm
G	feeding rate of fuel	g h^{-1}
H_f	height of fixed bed of ceramsite	cm
H^L	lower heating value of fuel	MJ kg^{-1}
i	element, compound	
k	nitrogen to oxygen mole ratio in dry air, $k = 0.7905/0.2095 = 3.7733$	
M^w_a	molar mass	g mol^{-1}
m_b	mass of ceramsite bed	g
t	Celsius temperature	°C
U	superficial gas velocity	m s^{-1}
U_{mf}	minimum fluidizing velocity	m s^{-1}
Y^a	relative mole fraction of water vapour in air related to oxygen, $Y^a = \frac{(1+k)\, y^a_{H_2O}}{1 - y^a_{H_2O}}$	
w_i	mass fraction of a species i in fuel	
w_f	moisture content in fuel	mol g^{-1}
X_{NO_x}	fractional conversion of the fuel-bound nitrogen to NO_x ($NO_{1.1}$)	
$y^a_{H_2O}$	mole fraction of water vapour in air	
y^d_i	mole fraction of a species i in dry flue gas	
ν_i	stoichiometric coefficients defined as the number of moles of element i per g of fuel	mol g^{-1}
τ_b	mean residence time of gas in fluidized bed	s
τ_r	mean residence time of gas in the reactor	s
φ	relative humidity of air	

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