

Modelling fo N₂O emissions during fluidized bed incineration of sludge.

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Abstract:

This work aims at studying the chemical mechanisms responsible for the formation and destruction of nitrogen oxides during combustion of sewage sludge in a fluidized bed. To do so, a mathematical model has been built in order to represent the relevant phenomena occurring within the reactor and estimate the nitrous emissions as a function of the operating parameters.

This paper presents the general equations that have been written in order to represent both the processes occurring within the gaseous phase (homogeneous combustion, heat transfer with sand...) and the processes that undergo the particles of sludge introduced within the reactor. More precisely, the particles are represented by a volumetric concentration within the bed, concentration for which a diffusion like equation is written in order to compute its value within each part of the bed (dense bed, disengagement, post combustion...). This pseudo transport model is coupled with a reaction scheme that allows a better understanding of the transformation of sludge particles within a hot fluidized bed. An important modeling effort has been done in order predict the becoming of nitrogen initially present in the sludge and that can be both released (as HCN and NH₃) during the pyrolysis step or burned during the combustion of the remaining char. The final model allows computing the local concentration of NO, N₂O, NO₂, HCN and NH₃ at every location of the bed.

Numerical estimations of the model are compared to experimental results obtained at industrial scale in a fluidized bed combustor of Veolia Company. This comparison shows very good agreement what makes the model a useful tool for design and optimization of such reactors.

Keywords:

Mathematical modeling, combustion, sludge, Fluidized bed.

1. Introduction

The global generation of sewage sludge, which is a by-product of wastewater treatment, is increasing continuously. Incineration of this material is a potential alternative for its disposal. It is commonly processed in fluidized beds either as a unique fuel [1-7] or in co-combustion [8-11]. This incineration is also of great interest since it might contribute to the renewable of our energy mix and to a lowering of carbon emission from fossil fuels. However, because of the high nitrogen content of the sludge, its combustion into fluidized beds leads to the production of nitrogen oxides, and more particularly to the production of nitrous oxide (N₂O) [12-18]. N₂O is a toxic compound and it is a greenhouse gas (with a potential which is 298 times the one of carbon dioxide), consequently it is necessary to control and reduce its release into the atmosphere.

Such a goal can be achieved using mathematical modelling. Indeed, using this technique, it can be understood how N₂O is produced from the nitrogen held with the initial sludge, what are the relevant phenomena that control its final production, and hence, how the combustor could be operated in order to reduce N₂O generation [19-23]. This is the scope of this paper. A mathematical model that has firstly been built to characterize fluidized bed combustion of Municipal Solid Waste (MSW) [24-26] was updated to consider sewage sludge as the fuel. During this first update, the processes like drying and pyrolysis were no more considered as instantaneous and the overall model

was set to a transient one [27]. Finally, in a recent evolution of this model, a detailed reaction scheme allowing for the estimation of the fate of nitrogen during fluidized bed combustion was added. This new development of the model was achieved in collaboration with Veolia Environnement Company. Indeed, this company is highly invested in “in situ” nitrogen pollutant emission reduction in order to reduce the impact of sludge combustion on environment. This final model was compared to industrial data obtained in a bubbling fluidized bed combustor with a nominal capacity of 2 t/h (2.5 MW Thermal input) of sludge with an initial moisture content of 75% (on a raw basis). This new development of the model as well as its application to this combustor is further developed in the present paper.

More precisely, the first part of the paper is devoted to the description of the final mathematical model that put into mathematical formalism the different phenomena occurring in such a fluidized bed combustor fueled with sewage sludge. The main assumptions on which the model relies, as well as the main phenomena taken into account by the model and derived into equations are presented. Then, comparison between those results (obtained by solving the above mentioned model in a Fortran homemade program) and the ones that were obtained using the industrial unit (for which detailed information compatible with industrial property will be given) over a week of operation is going to be presented in the third part of the paper. The variation of some of the operating parameters (fuel flow rate, extra-gas flow rate, primary and secondary air flow rates) will also be discussed. Finally, a conclusion will be drawn.

2. Materials and methods

Before entering further description of the model, some information on the system under consideration has to be given. The first one is relative to the nature of the fluidized bed. As it is depicted on Figure 1, the reactor which is considered here is of the dense bed type, operating under the bubbling mode. The feeding of the combustion air is split into primary air and secondary air. Both of them can be heated up and can be composed of air, enriched air, oxygen, or whatever gaseous species. Primary air is introduced at the bottom of the bed of material constituting the bed (sand, alumina...) through a distributor. Sludge can be introduced in the reactor at any location (height above the distributor), either in the dense bed or within the freeboard. Extra gas can be added in the reactor at the freeboard of the reactor or in the dense bed or at both locations. An extra sand supply ensures that the overall mass of bed material remains constant within the reactor.

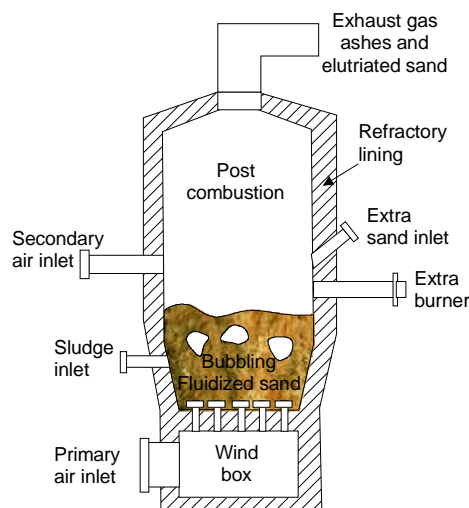


Figure 1: Scheme of the combustor under consideration.

2.1. Hydrodynamics of the bed

The scope of this paper is not to give a complete description of the overall model that has been developed, but to focus on the main concepts of this model. Dealing with bubbling fluidized bed, the main and first aspect that has to be addressed is the description of the hydrodynamics of the bed.

In accordance with previous work [24-27], the description of the dense part of the bed relies on the assumption formulated by Werther [28] that a “film” exists between the bubbles conveying the main part of the fluidization gas within the bed and the emulsion part remaining at incipient fluidization conditions. This assumption led us to the description of the gas held within the dense bed of the reactor as a model composed of three parts:

- A plug flow reactor, describing the bubbles of the bed
- A CSTR describing the gas held within the emulsion
- A 2D region (called from hereafter the “buffer zone”) for which the thickness corresponds to Werther’s film, but where heat and mass transfer, and reaction that processes are described as 2D processes being balanced by the ascending gas at incipient fluidization conditions and a net flux of gas arising from the release of volatiles within the emulsion part of the reactor that requires to be extract by the bubbles to maintain the emulsion at incipient fluidization conditions

This description of the gas held in the dense part of the reactor is completed by the description of the freeboard which is further decomposed into two plug flow reactors:

- A first one representing the disengagement zone where sand might be thrown as the bubbles burst at the top of the dense bed,
- A second one, which is supposed to be completely free of sand (post combustion zone).

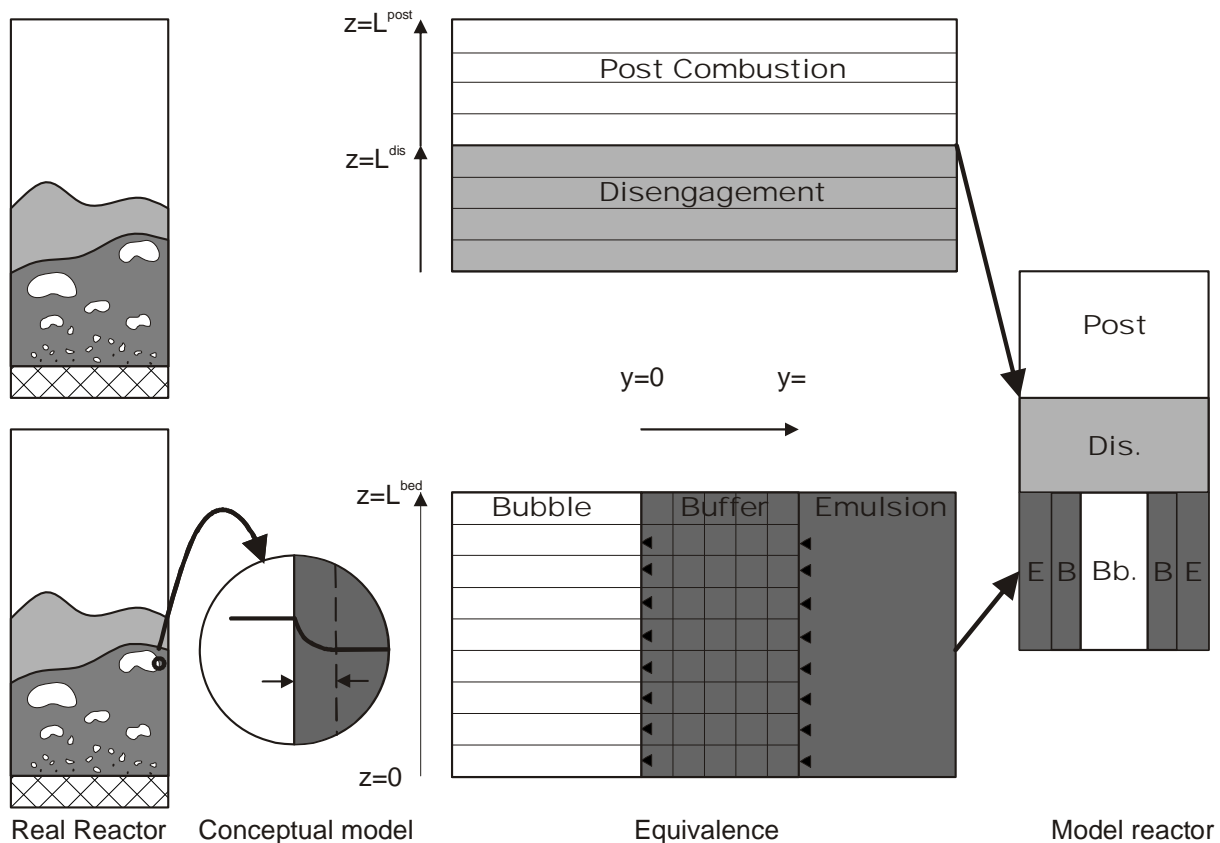


Figure 2: Scheme of the assemblage of models used to describe gaseous processes within the combustor.

- The scheme of this overall “hydrodynamics” model of the gas phase is depicted on Figure 2. This first description of the gas held within the reactor requires to be completed by the description of the particles that are present in the system. These ones are composed of two different categories. The first one represents the bed material which is supposed to be inert (from the chemical point of view). This “sand” might be present in the emulsion zone, in the disengagement zone, and, in the buffer zone. In our modelling effort, this sand is characterized

by an average temperature, computed according to specific heat transfer coefficient values [29] and to a ratio of presence between the dense bed and the disengagement zone. The second category of particles is reactive particles. It represents the particles of sludge that decomposes within the reactor. Each particle (which is further assumed to be spherical) is supposed to be composed at any time and any location of:

- • Free water
- • Bound water
- • Fresh organic material
- • Char (arising from pyrolysis and that might be composed of carbon, hydrogen, oxygen nitrogen and sulfur)
- • Inorganic material (corresponding to the ash remaining after complete combustion of the particle).
- The transport of these particles within the reactor is supposed to take place as “diffusion like” transport within the dense bed, where as it is mainly described by a “convection like” law within the dense bed which takes into account elutriation of the particles as a function of their size and density (which is a function of the initial characteristics of the sludge to burn and of the thermal and chemical “history” of the particle within the reactor). Indeed, it is assumed that drying of the free water and combustion of char do affect only the external radius of the particle (shrinking core model) whereas drying of bound water and pyrolysis do affect solely its density. The simplified description of the overall model is provided into figure 3. It has to be noticed that the coupling between the different phases (gas of the five considered regions, sand, and reactive particles of the dense bed or of the freeboard) are mainly taken into account through “source terms” characterizing heat and mass transfer associated to heterogeneous reaction and heat and mass transfer. For detailed information about the mathematical derivation of this “description”, interested reader should refer to [24; 25; 27]. It has to be noticed also, that each “regions” of the overall model is not considered as a “steady state” region but a transient one. The different equations describing the physical and chemical phenomena occurring within each of them includes a transient term representative of the accumulation of mass, species and energy, respectively.

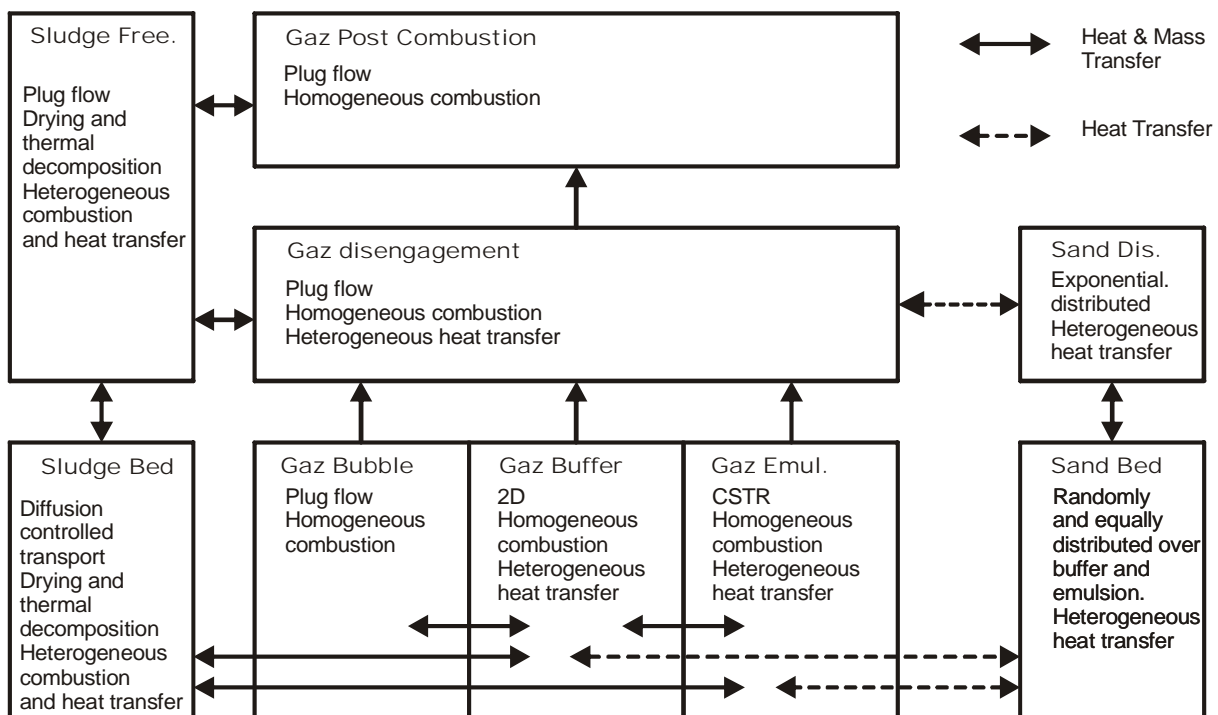


Figure 3: Complete description of the different zones of the model and of their connections.

2.2. Pyrolysis of Sludge

Once the drying step of the particles that enter the reactor is finished their temperature begins to rise again and the particles begin to decompose under the effect of heat and without any need of an additional reactant. This is the pyrolysis step. This process has been extensively described in the literature (see for example [30-38]). It has to be noticed that a lot of model were created to describe this phenomenon and the associated kinetics. For our purposed, the choice has been made to consider the pyrolysis step as a single and unique reaction so that the conservation of elements (C, H, O, N and S) is ensured during this step. In the literature, it is often shown that this pyrolysis occurs into two successive steps, with the creation of an intermediate species. This description does probably represent with much more realism the exact processes that the one assuming a single reaction. However, the literature does not generally provide the full kinetics information on this two steps reaction scheme. No information is really given on the composition of the product of the different reactions involved in the two steps scheme. Hence, it is really difficult to introduce such information within a reactor model. This is moreover the case, that this reaction scheme should ensure that the elements entering the feed port be balanced over the pyrolysis step. The procedure that was used to characterize this process in our modeling effort was thus the following.

- Assumption of a single and unique reaction leading (from the fresh and dry organic material of the sludge) to the production of two fractions: a solid one and a gaseous one (a part of which might be condensed at ambient temperature)
- Use of kinetics data to take into account the rate at which the fresh and dry organic material is decomposed into these two fractions. This information has been extract from the work of Conesa et al. [38] who assume a reaction order of 1.31 with respect to the organic material, activation Energy of 63.438 kJ/mol and a pre-exponential factor of 79.114 SI.
- Dealing with the estimation of the composition of the gaseous and solid fractions of the reaction, a specific pre computational step was performed. More specifically, it has been chosen to use the results obtained by Gomez Barea et al. [32] to determine the composition of the volatiles that would satisfy the experiment performed by this author and that would in turn respect the balances of the elements (C, H, O, N and S). Indeed, since the composition of the permanent gases (in terms of CH₄, H₂, CO and CO₂) was known from the work of Gomez Barea et al., and since specific assumptions were done concerning the element nitrogen, an inverse method was used to determine the composition of the volatiles and of the char of our pyrolysis model. More precisely, the objective function to minimize was that of the composition of the permanent gas, with the constraint of fulfill of the elemental balances, when the particles were subjected to any thermal history. Given the composition of the organic and fresh material of the sludge (see table 1) it was possible, according to the above mentioned pre-computation step, to estimate the composition of the products of the reaction. Finally, this procedure led to:
 - o A solid fraction (char composed of Ash, Carbon and Nitrogen) of 10.50% and a gaseous fraction (including condensable and permanent gases) of 89.50%
 - o A composition of the char (ash free basis) of 94.56% of Carbon and 5.44% of Nitrogen
 - o A composition of the gas which is provided in Table 2 (mass fraction)

Mass content (% , dry, ash free basis)	
C	58.97
H	8.21
O	26.77
N	5.44
S	0.53
Cl	0.08

Table 1: Ultimate analysis of the initial sludge.

Mass Fraction (-)		Mass Fraction (-)	
CH ₄	2.996 10 ⁻²	HCl	9.205 10 ⁻⁴
H ₂	2.854 10 ⁻³	HCN	1.051 10 ⁻²
H ₂ O	0.1288025	H ₂ S	6.303 10 ⁻³
CO	0.129	NH ₃	5.965 10 ⁻²
CO ₂	0.151	C ₂ H ₄	0.272
C ₆ H ₆	0.209		

Table 2: Composition of the gaseous product of the pyrolysis step.

2.3. Fate of Nitrogen

According to previous work [15], it has been assumed that the initial nitrogen content of the fresh and dry organic matter of the sludge was released during the pyrolysis into two fractions: one in the solid phase, and one in the gaseous phase (volatiles). Because we had no information on the part that was kept within the solid residue of pyrolysis, we assumed that the fraction of nitrogen in the solid residue was equal to the one initially present in the dry organic content of the sludge (5.44%, see tables 1 and 3). Also, dealing with the fact that nitrogen could be released as volatiles either as HCN or NH₃, the work of Liu & Gibbs [39] that assumed that the molar ratio of NH₃ to HCN released during pyrolysis was equal to 9 was also used in the frame of this study. These two data were added as constraints in the inverse method that was used to estimate the complete composition of the products of pyrolysis (see section 2.2).

Dealing with the becoming of both fractions, the work that was done by Sung [40] was used to take into account both, nitrogen present in the char and released as HCN and NH₃. Some previous study has also shown that this mechanism was suitable for our purposes [15]

3. Results and discussion

This section of the paper aims at providing the reader information about the industrial unit that considered getting some information on the influence of the process operating parameters on the nitrogenous emissions at the output of the combustor. In a first step, some insights on the industrial unit will be given. Information on the value of the operating parameters that were modified is provided in a second step. Then, the comparison between industrial data and numerical predictions is shown and discussed.

3.1. Industrial unit under consideration

It is of the bubbling bed type. Figure 4 gives a scheme of such a unit. The complete information relative to the geometry of the unit is confidential data and cannot be given to the reader of the paper. Nevertheless, these data have been introduced in the overall model to fit the real case. The main characteristics of the process are:

- Equivalent diameter of the reactor (considered as a cylinder): 3.22m
- Total height of the equivalent reactor: 11.5m
- Nature of the extra gas (for dense bed and reactor): Methane

- Proximate analysis of the sludge being burnt and previously mechanically dewatered (on a raw basis):
 - o Moisture : 75.40%
 - o Organic Material: 20.44%
 - o Inorganic Material: 4.16%
 - Lower Heating value of the sludge being burnt (raw basis): 3.8 MJ.kg-1
 - Ammonia held in the moisture content (mass basis): 0.309% (it has to be noticed that this ammonia part is released as NH₃ in the model during the drying of particles).

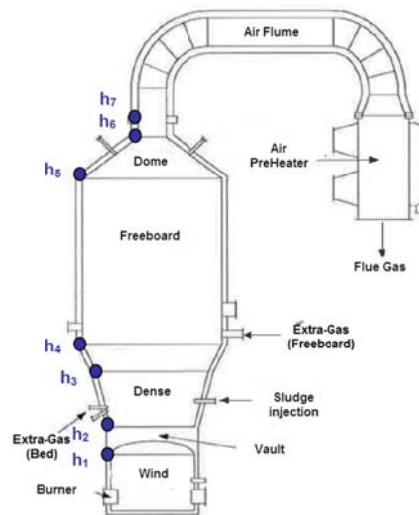


Figure 4: Scheme of the bubbling bed combustor.

The different samplings that were performed on that unit to measure the composition of the gas leaving the furnace was done at the air flume. It must be noticed here that this location for sampling was chosen to analyze the working of the combustor. It must be emphasized here that the composition of the gas that will be presented in the following section, is not the composition of the gas that is released to the atmosphere. Indeed, thanks to the flue gas treatment unit, this composition is adjusted to respect the law in force at the location of the plant.

3.2. Operating parameters

Series of experimental tests were carried out, over a week, with the above mentioned industrial unit. For this experimental investigation the following working parameters were modified:

- Sludge flow rate
- Flow rate of extra gas to the freeboard
- Flow rate of extra gas to the bed
- Flow rate of primary air (fluidizing air)
- Flow rate of secondary air (freeboard air)
- Temperature of the primary air.

To close this section, it has to be noticed that 7 industrial runs were carried out during the campaign.

3.3. Numerical vs industrial

To evaluate the ability of the model to properly describe the fate of nitrogen within the combustor, the experimental and simulated compositions of the gas leaving the reactor are studied as a function of the operating conditions. As the full stationary state of the industrial unit could not be completely reached (thermal inertia of the refractory lining of the bed, slight modifications of the operating

parameters around the values reported in Table 7 to ensure operation within the expected range), the different thermal levels computed by the model did not exactly match the industrial data. Hence, since the conversion of nitrogen within the reactor strongly depends on these thermal levels, the temperature of the sand was arbitrarily fixed, in the model, to the value of the average temperature of the bed measured in the industrial device. Although this could be considered as an “unsatisfactory” use of the model, it can however show the ability of the model to correctly predict the nitrogen conversion once the temperature of the sand is properly estimated.

The composition of the gas leaving the industrial reactor was measured continuously using an IFTR analyzer and a paramagnetic oxygen analyzer, respectively. The gas was sampled within the Air Flume of the reactor. It must be noticed here that this sample point is located before the flue gas treatment process, and that, during the different runs of the campaign, the SNCR used in the industrial unit to reduce the NO emissions was switched off. The duration of this mode operation was kept compatible with the regulation law in force at the industrial plant.

Figure 5 shows the evolution of the outlet temperature of the gas as a function of the different test runs. The numerical results were compared with two available industrial data: the temperature at the top of the combustor and the air flume temperature. It can be seen on Figure 5 that the model fits rather well the thermal levels measured on the industrial unit. The small discrepancies that appear are associated to the thermal inertia of the system. Small variations might also occur between the recorded values of the operating parameters and the values actually applied to the process.

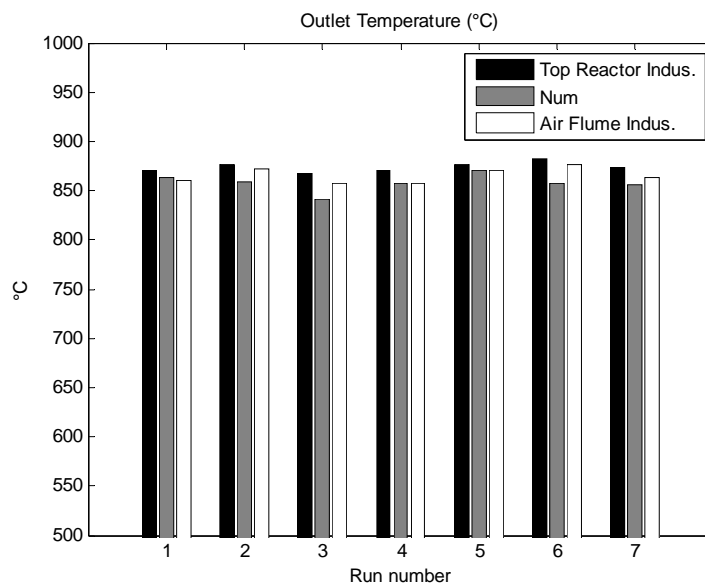


Figure 5: Value of the temperature at the outlet of the combustor during the test runs. (Industrial at the top of the bed and at the air flume, Numerical at the outlet of the equivalent reactor).

Figure 6 compares the predicted composition of the gas and of the measured one in terms of volumetric fraction of oxygen (on a dry basis), water and carbon dioxide (both of them on a raw basis). This comparison is also shown for the HCl concentration on the flue gas (It is reminded to the reader that this HCl content was measured before the gas enters the flue gas treatment unit). Thanks to this figure, it can be concluded that the model perfectly predicts the conversion of carbon, oxygen and hydrogen originating from the sludge (moisture and organic content) and from the combustive air, respectively. The very little discrepancies between simulated and measured data might be associated to fluctuations of the composition of the sludge.

On Figure 7, the estimation of the outlet concentration in N_2O and NO is compared to the value measured in the industrial device (it is reminded here that this data was collected while the SNCR of the unit was switched off). Dealing with NO estimation, it can be concluded that the computed results match well with industrial data (except for the test runs n° 4 and 5). Dealing with the estimation of the N_2O concentration in the flue gas, the agreement between simulated and industrial

data is less satisfactory. The observed discrepancies (with both species NO and N₂O) might be associated to small variation in the composition of the sludge. An inexact prediction of the temperature of the flume temperature (which might be associated to inexact value of the working parameters (see above)) may also impact the results. An illustration of this statement is provided on Figure 8 which shows the evolution of the relative error on the value of the concentration of the nitrogen containing species as a function of the relative error on the temperature of the Air Flume. This figure shows that the error on the nitrogen containing species might be correlated to the error on the temperature

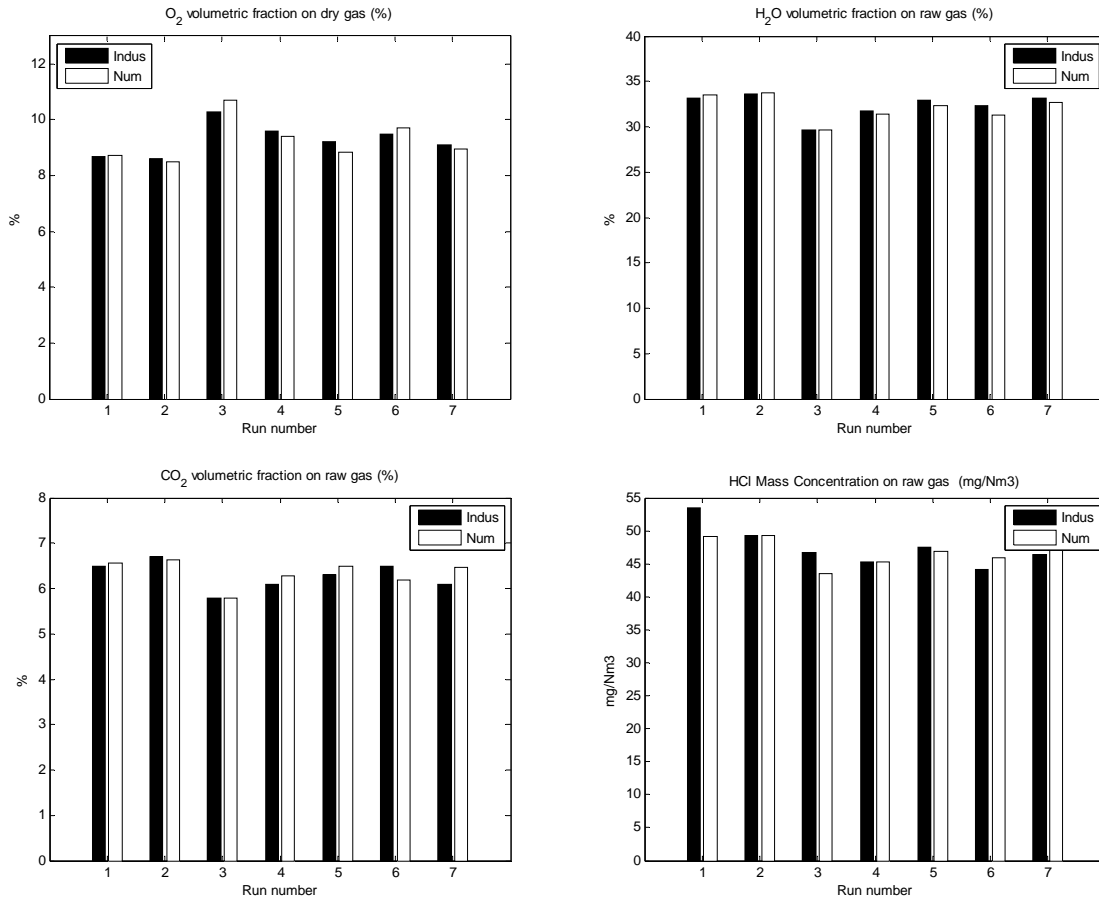


Figure 6: Value of the composition of the gas (main compounds) during the test runs. (Industrial at air Flume before the flue gas treatment unit, Numerical at the outlet of the equivalent reactor)

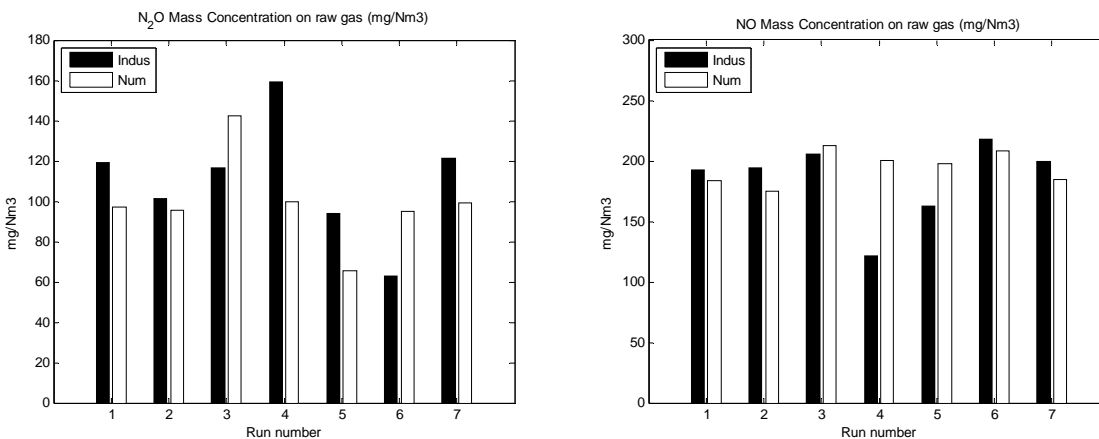


Figure 7: Value of the N₂O and NO concentration of the gas during the test runs. (Industrial at air Flume before the flue gas treatment unit and with the SNCR unit switched off, Numerical at the outlet of the equivalent reactor)

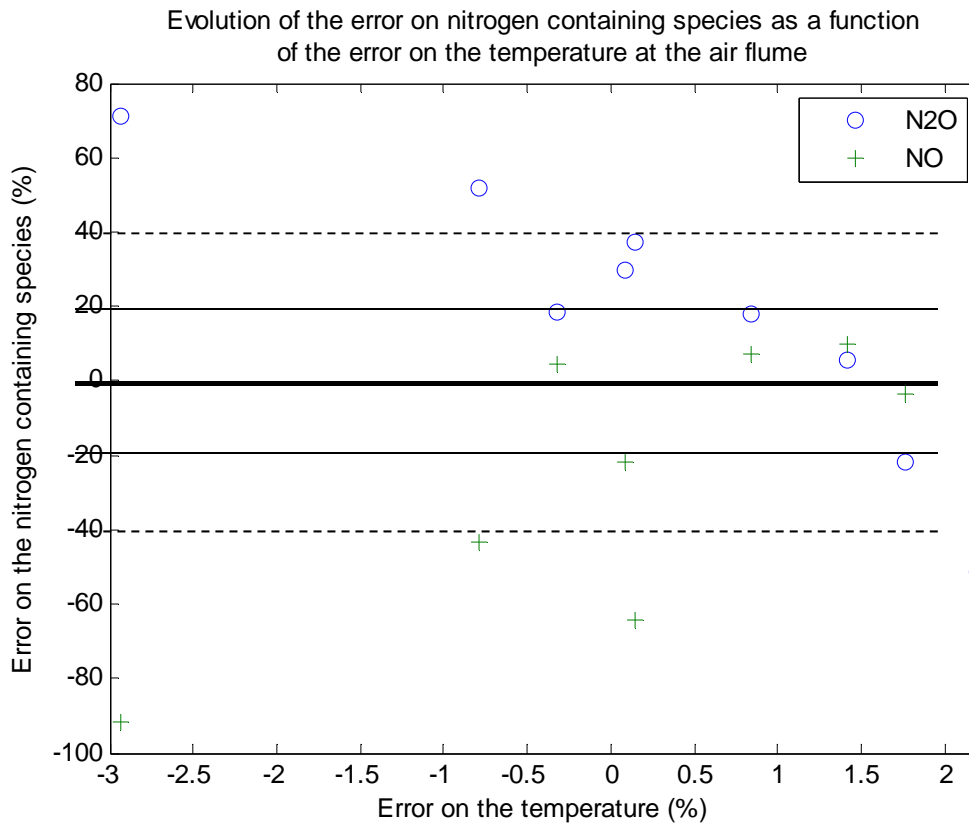


Figure 8: Correlation between the errors obtained on the estimation of the nitrogen containing species and on the error on the estimation of the temperature of the air flume (Error are defined as the absolute value of the difference between numerical and industrial data divided by industrial data).

4 Conclusions

This paper has presented some very recent work that has been done on the modelling of nitrogen conversion within a fluidized bed devoted to sludge combustion. It provides results that were obtained through collaboration between an industrial partner (Veolia Environnement Company) and an academic laboratory (LaTEP, Université de Pau et des Pays de l'Adour). The main goal of this collaboration was to build a model allowing description of the relevant phenomena occurring within such a combustor. This model, considers the reactor as an assemblage of several zones, including a 2D one relating the transfer and reaction phenomena occurring within the surrounding of the bubbles crossing the dense bed. It also takes into account the transport of the reactive particles of sludge ongoing drying, pyrolysis and heterogeneous reactions. All of these reactions are included in the model with their respective kinetic data, which also ensure conservation of the elements involved in the transformation (carbon, hydrogen, oxygen, nitrogen, sulfur and chlorine). The existing model has been updated to account for the specificity of nitrogen transformation during the overall process, and more particularly during pyrolysis and further conversion into NO and N₂O. The ability of the model to properly describe this transformation has been proven thanks to comparison with data obtained from an industrial operating unit own by Veolia Environnement Company

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