Contributions to the Mathematical Modeling of Electron Beam Flue Gas Treatment

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This paper aims to present the contributions to the mathematical modeling of atmospheric pollutants' removal using the electron beam flue gas treatment (EBFGT), after an in-depth look at its current status and perspectives for future development. Despite having been demonstrated at industrial level, EBFGT's market penetration is still low. The bulk of experimental and modeling work dedicated to the EBFGT has been carried out in order to understand how the operating parameters influence the removal efficiency of the process and how they impact the overall cost of the technology. The importance of developing new accurate mathematical models in order to allow a better prediction of EBFGT performances and, consequently, a high acceptance on industrial scale is presented as well as the authors' personal contribution in this field. The paper discusses the influence of the most important operating parameters affecting to a higher or lower degree the removal process efficiency (the irradiation dose, the water and ammonia content of the treated gases, the temperature of the gases entering the irradiation chamber, the irradiation time and the initial concentration of pollutants).

Keywords: mathematical modeling, water droplets, absorption, electron beam, flue gas

With the continuous rise in population and industrial activities during the last decades, the control and removal of harmful emissions represent one of the key challenges in assuring a better quality of the environment. Two of the most important atmospheric pollutants, sulfur and nitrogen oxides (SO_2 + NO_x), cause acidiphication, eutrophication and ozone depletion, contributing to climate change [1]. The most widely employed methods for their destruction are wet flue gas desulfurization (WFGD) and selective catalytic reduction (SCR), methods that have long proved their advantages: high efficiencies for pollutant removal, relatively simple installation and operation, etc. [2].

However, due to important limitations of both WFGD and SCR, several alternative methods have been devised for the removal of sulfur and nitrogen oxides from exhausts: adsorption (using activated coke) or absorption technologies (employing either oxidizing or complexation reagents), membrane filtration, microbiological treatment, photo-catalytic oxidation, microwave and plasma based technologies [3]. A special class encompasses the plasma based depollution methods, in particular on the non – thermal plasmas, as they have been commercialized for industrial coal fired energy plants for almost two decades [4].

The EBFGT is an established technology for the removal of sulfur and nitrogen oxides, reaching high efficiencies for the pollutants' removal on industrial scale demonstration plants: more than 95% for SO_2 and over 70% for NO_x [5]. Moreover, the use of electron accelerators was successfully extended to the removal of several volatile aromatic organic compounds: benzene, toluene, and a wide variety of poly-aromatic hydrocarbons (PAHs), etc. but still on a laboratory scale [6-8]. The EBFGT first appeared in laboratory in 1970, as a result of the cooperation between the Ebara Corporation and the Japan Atomic Energy Research Institute [9]. The experiments showcased the suitability of using linear accelerators for the removal of SO_2 without the use of additives, at relatively low temperatures (~100°C) and atmospheric pressure [5]. The treatment was consequently extended to the simultaneous removal of SO_2 and NO_x, emphasizing high efficiencies for both pollutants. Following these developments, several pilot scale facilities have been developed during the following decades in USA, Germany, Japan, Poland and China [10]. The following step was the development of the first industrial scale treatment facility in Chengdu, China, focused on treating high sulfur concentration flue gases emitted from coal burning [5]. Since then, several other industrial scale applications have been developed, mostly in Poland and China (fig. 1).

The operation schematics of an electron beam facility for the treatment of the gases emitted from coal or oil

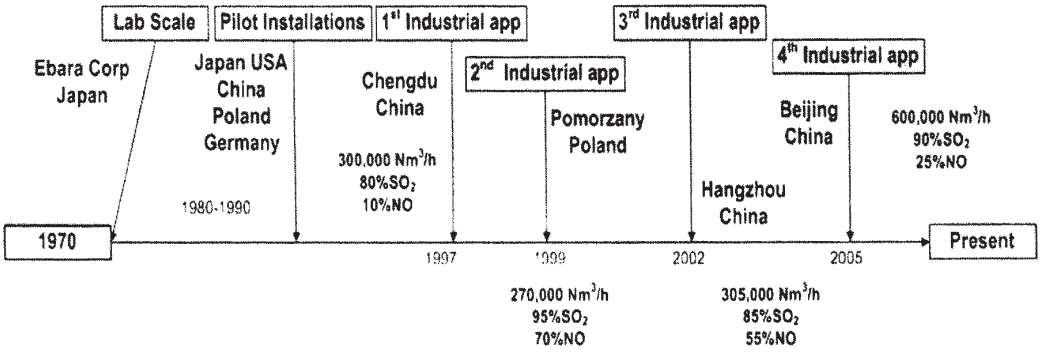

Fig. 1. Historical development of EBFGT

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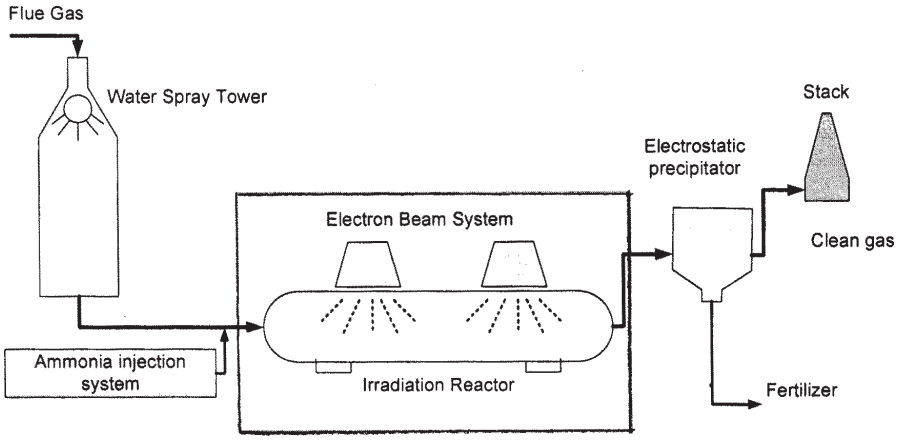

Fig. 2. Schematics of an EBFGT facility, adapted from [4]

burners are presented in figure 2: before entering the installation, the gases from boilers are de-dusted in electrostatic precipitators to remove particulate matter and other solid pollutants. The partially purified gas enters a spray cooler, where the water evaporation process takes place. As a result of this process the temperature of the gases is lowered to 65– 80°C and the humidity level can be increased up to 10–16% (volume). The importance of the plasma reactor's configuration has been highlighted [11, 12], therefore the development of good mathematical models able to accurately predict the performance of the former became of utmost importance.

The investigations into the mechanisms of non-thermal plasma gas depollution have revealed that the collisions between the generated accelerated electrons and the main species of the flue gas (N_2, O_2, CO_2 and H_2O vapor) are the first steps in the pollutants removal chemistry. These interactions are termed primary radiolysis phenomena and occur in the span of 10^{-14} – 10^{-7} seconds, encompassing ionization, excitation, dissociation and charge transfer reactions. Due to both the complexity of the phenomena involved during the primary radiolysis stage and the extremely fast reaction rates associated to them, it has become common practice to use the concept of radiochemical yield to describe the behavior of the main components of the gas during this stage [13-15], in order to simplify the mathematical approach. Currently the values proposed by [16, 17] for these radiochemical yields or G-values (defined as the number of molecules destroyed or formed per 100 eV of absorbed irradiation) are used as stoichiometrical coefficients, as presented in eqs (1 – 4):

$$4.14\,N_2 \rightarrow 0.885\,N^2D + 0.295\,N^2P + 1.87\,N^4P + 2.27\,N_2^+ + 0.69\,N^+ + 2.96\,e^- \quad (1)$$

$$5.3\,O_2 \rightarrow 2.98\,O + 2.25\,O^1D + 2.07\,O_2^+ + 1.23\,O^+ + 3.3e^- \quad (2)$$

$$6.7\,H_2O \rightarrow 0.51\,H_2 + 4.25\,OH + 4.15\,H + 0.46\,O^3P + 1.99\,H_2O^+ + 1.99e^- \quad (3)$$

$$7.54\,CO_2 \rightarrow 4.72\,CO + 5.16\,O + 2.24\,CO_2^+ + 0.51\,CO^+ + 0.07\,O^+ + 2.82e^- \quad (4)$$

In order to quantify the effect the primary irradiation phenomena has on the main gas components, the following mass balance equation (eq. 5) was proposed by [18].

$$\frac{dN_i}{dt} = D^* G_i \quad (5)$$

where D* is the irradiation rate and G_i is the radiochemical yield of the irradiated species.

Further studies have shown that the irradiation intensity depends on the density of the medium and that the accelerated electrons interact with gas components proportional to their composition, and thus have adjusted the mass balance equation to reflect that (eq. 6).

$$\frac{dN_i}{dt} = \rho D^* G_i X_i \quad (6)$$

where ρ is the density of the irradiation medium and X_i – the molar fraction of the irradiated species.

The reactive species listed in the eqs (1-4) interact with each other and with the gas components through ion – ion recombination, radical – radical and radical – neutral reactions, in what is denoted as secondary radiolysis phenomena, with a time frame of about 10^{-7} – 10^{-1} seconds.

The first kinetic models developed for these interactions started from the simplest reaction systems, formed only of N_2, O_2 and NO, and considered only 32 chemical reactions [18]. The work of [18], even though it proposed a small reaction system, had succeeded to offer an insight of the most important pollutant removal mechanisms: oxidation vs. reduction. The kinetics of the gas phase irradiation process have been further studied in various works [16,19], improving the size of the reaction system gradually until reaching over 1100 chemical reactions and identifying more than 90 participating chemical species.

The extensive kinetic study of [16] provided an in depth analysis of the most important reaction pathways leading to the removal of pollutants, concluding that the oxidation mechanism is best represented by the reactive oxygen species generated as a result of the primary radiolysis phenomena (O^1D, O, OH) further react with each other, yielding other species with a strong oxidizing capacity such as O_3 and HO_2, eqs (7-10).

$$O + O_2 \rightarrow O_3 \quad (7)$$

$$OH + O \rightarrow HO_2 \quad (8)$$

$$OH + O_3 \rightarrow HO_2 + O_2 \quad (9)$$

$$HO_2 + O_3 \rightarrow OH + 2O_2 \quad (10)$$

All of them will further interact with the pollutant molecules, promoting the oxidizing pathways for the transformation of sulfur and nitrogen oxides into their corresponding acids, as presented in figures 3 and 4.

In addition to the oxidizing pathway, NO is removed as a result of reduction reactions prompted by the presence of nitrogen and amidogen radicals, leading to the formation of molecular nitrogen. Early studies have shown that the reduction reactions are responsible for around 23% of the total removal efficiency of NO_x [20] in the absence of ammonia addition, findings substantiated by the works of [16,19], who have also evaluated the interactions between

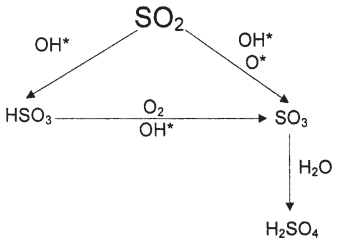

Fig. 3. SO_2 removal mechanism

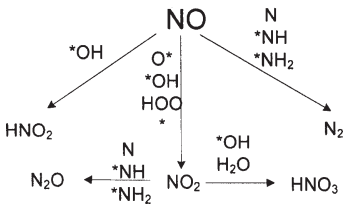

Fig. 4. NO_x removal mechanism

ammonia and the gas components, pollutants and secondary electrons.

However, the computational effort associated to these complex kinetic systems has prompted researchers to find methods of reducing the size of the reaction system and find innovative computational pathways, such is the case with [21], who proposed a reduced kinetic model starting from the simplest reaction system and steadily evolving to consider 103 chemical reactions. The adopted solving strategy follows the procedures detailed in [22], taking into account that the model of EBFGT is of DAE type with a strong stiff differential system. The paper stresses the beneficial effect of the CO_2 and SO_2 presence in the system and the improvement to the mathematical model predictions when taking them into account.

More recent modeling studies are those performed by [23, 24] who aim to provide a complex kinetic model for the gas phase interactions in order to better understand the mechanism of the EBFGT. The works focus on identifying the OH radical sinks and analyzing the pathways through which the OH radicals are formed and destroyed. The first paper presents a kinetic system obtained from the work of [16], considering additional chemical species (e.g. ion hydrates) and proposing different reaction rate constants – published by NIST and IUPAC more recently. The second paper investigates the contribution of CO and CO_2 to the generation and consumption of OH radicals, by using the model proposed earlier. The main novelty of the work is the fact that the model considers the irradiated flow cell as a continuous stirred tank reactor, as opposed to the norm in modeling, which considers discontinuous reactors. The authors stressed the importance of the oxygen presence in the irradiated mixture and its influence on the overall process.

In addition to this radiolytical reaction pathway, early experimental studies showed that the sulfur dioxide is also removed by direct reaction with ammonia via the thermo – chemical pathway. The reaction mechanism (described by eqs 11 –15) was initially proposed by [25], who used an overall rate of the process with kinetic constants obtained through a regression analysis performed on a limited set of experimental data, later refined by [26]. In the absence of irradiation, the thermo – chemical pathway is solely responsible for the removal of sulphur dioxide, which can be as high as 80%, depending on the temperature inside the irradiation chamber.

$$SO_2 + NH_3 \rightarrow NH_3SO_2 \quad (11)$$

$$NH_3SO_2 + NH_3 \rightarrow (NH_3)_2SO_2 \quad (12)$$

$$(NH_3)_2SO_2 + 0.5\ O_2 \rightarrow NH_4SO_3NH_2 \quad (13)$$

$$(NH_3)_2SO_2 + H_2O \rightarrow (NH_4)_2SO_3 \quad (14)$$

$$NH_4SO_3NH_2 + H_2O \rightarrow (NH_4)_2SO_4 \quad (15)$$

In addition to these homogeneous routes, early studies have also recognized the huge impact of the heterogeneous chemistry, especially the interactions between SO_2 and NH_3 on the filter cake, after irradiation and the formation of a liquid phase due to sulphuric acid nucleation. However, the extent to which this heterogeneous chemistry influences the overall removal of pollutants has been a topic under debate. Matzing et al. [27] evaluated different possibilities for gas-aerosols interactions that could account for the second heterogeneous pathway – as small increases in humidity were reportedly accompanied by large increases in SO_2 removal efficiency. However, the reported results showed that even when passing a stream of concentrated sulphuric acid through the irradiation chamber would not significantly impact the removal efficiency of sulfur dioxide. These finding are contradicted by those of [28], who have observed that the liquid droplets can improve the removal of both NO_x and SO_2. Moreover, [29] have demonstrated the importance of the heterogeneous reaction mechanism for the removal of SO_2 (using pulsed corona streamers instead of EB irradiation in the experimental work). The authors proposed a kinetic system for the liquid phase reactions centered on the SO_3^- and OH radical, investigating the heterogeneous mechanism in the presence and absence of NH_3. For the kinetics of the liquid phase reactions the reaction system both [28] and [29] employed a condensed version of the one presented by [30] for aqueous phase atmospheric chemistry, where the transformations of S(IV) to S(VI) ionic compounds are believed to occur via a chain mechanism, with the main termination reactions presented in figure 5.

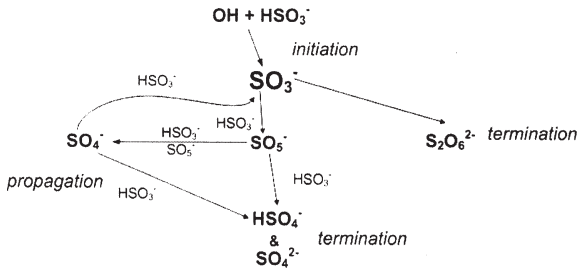

Fig. 5. Liquid phase chemical reactions

Mathematical modeling principles

As stated above, due to the fact that the size of the kinetic system greatly impacts the computational capacity, more recent modeling studies have only taken into consideration a fraction of these chemical reactions [31] or have resorted to empirical or semi-empirical approaches [32]. Due to the low liquid to gas ratio observed experimentally [28], researchers have mostly neglected the formation and behavior of the liquid phase resulted from the sulphuric acid nucleation in their treatment of EBFGT phenomena.

The aim of the current section is to present the mathematical models developed by the authors, which can accurately describe the behaviour of the sulfur and nitrogen oxides subjected to electron beam irradiation.

The mathematical models were developed so far following the main phenomena involved in the treatment of flue gases with electron beams (fig. 6).

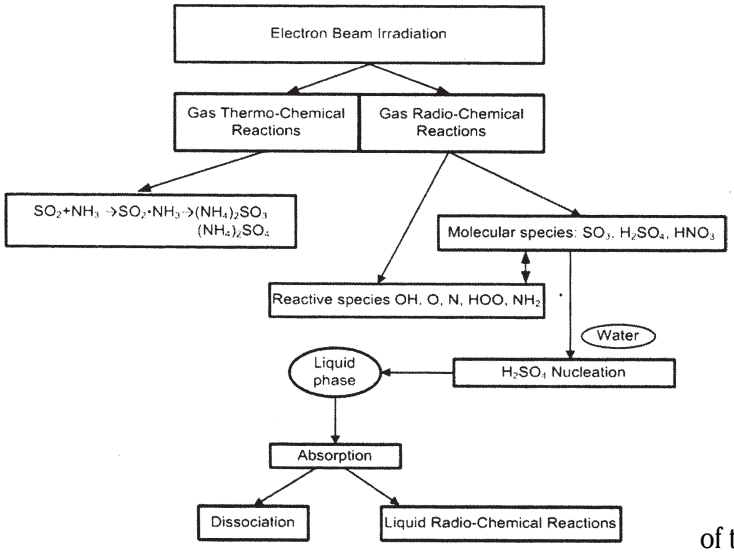

Fig. 6. Modeling algorithm

Gas phase transformations

The generation of reactive species as a result of irradiation was described using equations (1-4), while the secondary radiolysis phenomena and the molecular reactions are presented in [33]. The kinetic system assembled is derived from the work of [21, 23], analyzing the reaction system in order to achieve a computationally affordable model with high accuracy. The developed kinetic system considers 40 chemical species taking part in 90 radio-chemical reactions and is able to predict with sufficient accuracy the process efficiency while being computationally economic.

For the proposed steady–state plug flow (PF) modeling approach, eq. (16) represents the mass balances, where the volume fraction of liquid (f_L) changes at every integration step. In addition to this the dose is believed to be absorbed by the gas and liquid phases proportional to their weight.

$$\frac{dN_{Gi}}{(1-f_L)\cdot dV} = D_G^{*}\rho_G G_i X_i + \quad (16)$$

Condensation process

The nucleation phenomena (the formation of critical clusters of water and sulphuric acid) was modeled using the parameterizations proposed in [34] for the critical composition of the clusters and the one proposed by [35] for the nucleation rate. The further growth of the nuclei as a result of sulphuric acid absorption and the coagulation phenomena were neglected in the modeling. The values obtained for the nucleation rates computed using the parameterizations ($\sim 10^{14} - 10^{15}$ cm^{-3}) and the composition of the critical clusters (~0.15 molar % H_2SO_4) are in good agreement with the results reported for atmospheric experiments conducted in similar conditions [36].

Absorption phenomena

The absorption of ammonia, nitrous acid, nitric acid, oxygen and sulfur dioxide in the liquid phase, followed by their dissociation into ionic species was modeled employing solubility coefficients. Their use is superior to the Henry's Law approach as the latter holds only for the infinite dilution case, while the droplets contain a significant amount of sulphuric acid. The solubility approach has led to a better overall fit between the results of the model (using the discontinuous – DC – model for the reactor) and those obtained experimentally, as discussed in our previous work [33]. On the other hand, the presence of sulfate and sulfite anions in the droplets can modify the solubility coefficients for the other species and this aspect must be further studied.

Liquid phase dissociation and chemical reactions

The water radiolysis and the chemical reactions of the ionic and free radical species are described in the Supplementary Material in [33]; eq. (17) represents the mass balance for the liquid phase components, similar to that written for the gas phase species.

$$\frac{dN_{Li}}{f_L \cdot dV} = D_L^{*}\rho_L G_i X_i + \quad (17)$$

The resulting model takes the shape of a system of differential algebraic equations embedding mass and charge balances for the chemical species listed in table 1. The resulting system is solved with an in-house written

Gas Phase		Liquid Phase	
Radical Species	$H\cdot SO_4$, $O\cdot$, $N\cdot$, $\cdot OH$, $HOO\cdot$, $H\cdot SO_3$, $H\cdot$	Radical Species	$\cdot OH$, $HOO\cdot$
Ionic Species	O_2^-, O^+, N_2^+, N^+, O_2^+, NO^+, NO_2^-, CO_2^+	Ionic Species	HSO_3^-, SO_3^{2-}, SO_3^-, SO_5^-, HSO_5^-, HSO_4^-, SO_4^-, SO_4^{2-}, $S_2O_8^{2-}$, $S_2O_6^{2-}$, H^+, O_2^-, NO_2^-, NO_3^-, HO^-, NH_4^+
Meta-stable Species	$N(^4S)$, $N(^2D)$, $O(^1D)$, $O(^3P)$	Molecular Species (absorbed from gas)	H_2O, CO_2, CO, NO, NO_2, NO_3, O_3, H_2SO_4, NH_3, HNO_2, SO_2, HNO_3, O_2, H_2O_2
Stable Molecular Species	N_2, O_2, CO_2, H_2O, NO, SO_2, NH_3, NO_2, H_2SO_4, CO, O_3, N_2O, N_2O_5, SO_3, NO_3, HNO_2, HNO_3		
End Products	$(NH_4)_2SO_4$, $(NH_4)_2SO_3$ NH_4NO_3, NH_4HSO_4, NH_4NO_2		
Complex (short-lived) species	$HOSO_2O_2$, $HOSO_2ONO$, $HOSO_2ONO_2$, H_2SO_5, $H_2S_2O_6$, $NH_4SO_3NH_2$, $(NH_3)_2SO_2$, NH_3SO_2		

Table 1
CHEMICAL SPECIES CONSIDERED IN THE MATHEMATICAL MODEL

Experimental conditions							
Experiment #	Temperature (°C)	Humidity (%)	Dose (kGy)	Residence time (s)	$[NO]_{initial}$ (ppm)	$[SO_2]_{initial}$ (ppm)	NH_3 ratio
1	58.6	12.0	10.0	14.43	127	383	0.92
2	59.2	10.7	10.0	14.36	171	364	0.89
3	60.4	8.6	10.2	4.11	161	673	0.89
4	54.9	8.2	10.0	13.4	129	359	0.88
5	60.3	7.7	10.1	4.05	196	467	0.88
6	78.8	6.9	10.1	6.02	216	430	0.9
7	55.1	7.9	12.5	3.56	157	465	0.91
8	55.8	8.0	12.7	3.63	159	484	0.88
9	78.8	6.7	10.1	5.99	216	421	0.91
10	61.2	8.1	7.1	4.37	181	427	0.87
11	62.3	7.8	5.1	4.41	186	515	0.91
12	59.8	7.8	2.8	4.22	182	510	0.87
13	59.1	9.0	8.0	4.03	146	462	0.93
14	59.3	8.0	10.4	4.13	158	624	0.91
15	60.9	8.2	10.2	4.11	194	443	0.89
16	60.8	9.8	10.1	11.94	175	314	0.91
17	59.0	12.4	11.4	13.78	181	358	0.9
18	60.6	10.7	12.1	14.36	168	377	0.87
19	59.8	7.7	12.1	4.08	190	386	0.9
20	61.8	7.7	10.2	4.13	185	398	0.9

Table 2
EXPERIMENTAL CONDITIONS USED TO COMPARE MODELING RESULTS [31]

routine based on Gear's method for stiff differential equations.

Results and discussions

In order to validate the plug flow model and evaluate its results, we compared the predictions of the plug flow approach with the set of experimental data presented in [32], listed in table 1. As seen in figure 5 and 6, the PF approach shows a satisfactory agreement with the experimental data, better than the DC modeling approach. The mean relative deviation in the PF approach is 13.4% for NO and 9% for SO_2, while the values for the DC model, presented in [33], are 13.9 and 9.3 % respectively.

The irradiation dose has been identified as the most important parameter in the removal of nitrogen oxides, as its magnitude is directly linked to the amount of primary reactive species generated during radiolysis. Although irradiation doses as high as 50 kGy have been demonstrated for almost complete removal of NO_x and SO_2, much lower levels of irradiation are usually employed. These levels vary in the range of 2 – 10 kGy, depending on the efficiency targets set for pollutant removal. In the case of sulfur dioxide, doses as low as 3 - 5 kGy ensure suitable removal efficiencies at relatively low temperatures and humidity contents, as can be observed from experiments 11 and 12. Increasing the magnitude of the irradiation dose above this threshold does not significantly affect the SO_2's removal efficiency. However, at this level of absorbed dose, the removal efficiency of NO_x is in the range of 47 – 65 %, even for small pollutant concentrations, as can be seen from table 1. Nevertheless, increasing the irradiation dose will lead to higher operating costs, which represent one of the more serious drawbacks of the EBFGT.

The temperature of the process is also an important operating parameter that must be controlled as it has opposite effects on the behaviour of SO_2 and NO_x: as the temperature increases, the rates of the radiochemical reactions also increase, as collisions between gas molecules become more frequent, thus making the removal of NO_x easier. On the other hand, the high temperatures diminish the rates of the thermo-chemical reaction pathways for the sulfur dioxide and thus the pollutant's removal efficiency, which drops below 70% in the case of temperatures higher than 75°C (experiment 6).

The temperature and the humidity of the gases entering the irradiation chamber are strongly dependent on each other, as the flue gases are cooled in a water tower before the electron beam treatment and thus the humidity content is dictated by the temperature difference between the gas exiting the 'stack' and the target for the temperature in the irradiation chamber [37]. This, however, is not the case in pilot and laboratory scale applications, where the temperature and humidity values can be set independently. High humidity contents are beneficial for both the SO_2 and the NO_x, as the higher water content is linked to a greater rate of hydroxyl radical formation, promoting the oxidation pathways for the pollutants' transformation, as illustrated by experiments 1 and 2.

The ammonia content of the flue gases also impacts positively the removal of both pollutants, promoting the transformations of acidic compounds (H_2SO_4, HNO_2 and HNO_3) into their corresponding salts. The ammonia is usually added to the gases in less than stoichiometric proportion to the concentration of SO_2 and NO_x, as it has to be closely monitored to avoid the release into the atmosphere of high quantities of un-reacted ammonia. The removal efficiency of SO_2 is believed to show a more pronounced dependence on the ammonia stoichiometry, as the thermo-chemical reaction rates are more dependent on the ammonia concentration. However, due to the fact that the ammonia content was not significantly varied during the experiments, this hypothesis could not be definitely confirmed.

The initial concentration of the pollutants is also important for the overall efficiency of the process: higher

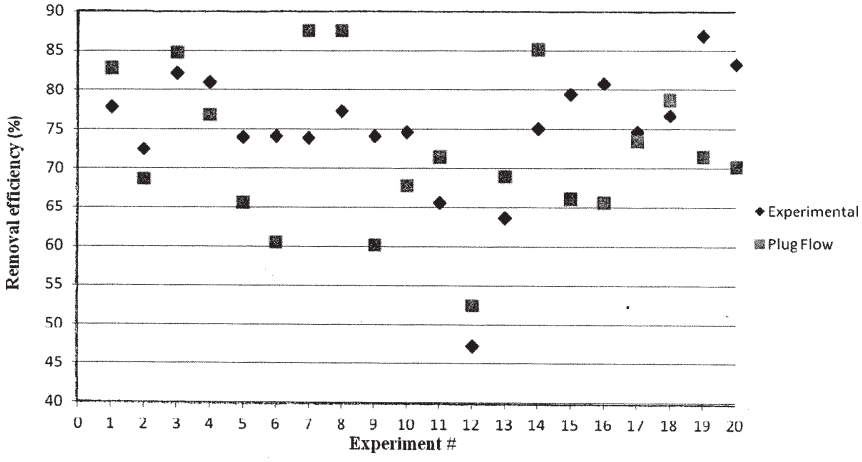

Fig. 7. Plug flow modeling results vs. Experimental data for NO removal

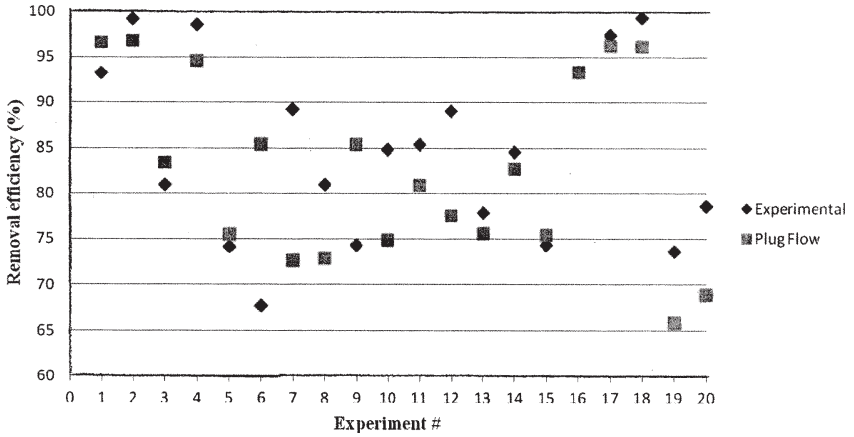

Fig. 8. Plug flow modeling results vs. Experimental data for SO_2 removal

initial concentrations of NO lead to poorer removal efficiencies for NO_x, as the ratio of reactive species to pollutant molecules is lower. In contrast, a higher SO_2 concentration leads to a higher removal efficiency, as the rate of the thermal reactions will thus increase. The higher SO_2 initial concentrations also have a slightly beneficial effect on the NO removal process, as the pollutants interact during irradiation.

The residence time and, implicitly, the flue gas flow rate have been shown to have a marginal effect on the overall performance of the process: higher residence times imply a longer time for the molecular reactions to take place, which leads to a slightly better removal but it also diminishes the treatment capacity of the plant.

Conclusions

The EBFGT is a competitive technology compared to the more traditional ones and even with other non-thermal plasma treatment methods for the treatment of SO_2 and NO_x in terms of operation and investment costs, scalability, flexibility and operability. Its features make it suitable for the treatment of a wide variety of other pollutants such as VOCs, dioxins and other gaseous chlorinated compounds. Further developments in the field of electron accelerators, irradiation methods and reactor design are required to ensure larger market penetration of this technology. The mathematical modeling presented in this work takes into account both gas and liquid phenomena while the modeling results show a good agreement with the experimental data collected from literature. In addition, the paper proposes the novel plug flow approach to the modeling of the EBFGT, as it offers more optimization opportunities and shows a better agreement with the experimental data than the discontinuous modeling approach.

Acknowledgments: This work is supported by the Sectoral Operational Programme Human Resources Development (SOP HRD), financed from the European Social Fund and the Romanian Government under the contract number POSDRU/159/1.5/S/137390.

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Manuscript received: 29.01.2015