Gaseous Pollutants Removal by Electron Beam Based Hybrid Systems

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This paper presents the results obtained in the gaseous pollutants removal from industrial flue gases by combined irradiation with electron beam (EB) and microwave (MW). The important role of the catalyst under EB and MW irradiation in the complete oxidation of volatile organic compounds (VOCs) was shown. The goal of the research is to improve pollutants removal efficiency and to reduce the EB irradiation dose. Two innovative hybrid installations for SO_2, NO_x and VOCs removal are presented. One of them permits experiments with successive EB and MW processes in two distinct reactors. The other one allows experiments with simultaneous EB and MW processes in the same reactor. All reactors contain catalyst bed in the case of the VOCs removal. The experiments show that combined EB and MW treatment applied to SO_2 and NO_x removal as well as simultaneous EB, MW and catalyst treatment applied to VOCs removal, provides satisfactory efficiency values compared with separate EB or MW method.

Keywords: electron beam, microwave, catalyst, NOx, SO_2, volatile organic compounds

Gross emission of pollutants emitted during industrial processes from steel plants, power stations, chemical plants and other activities is tremendous worldwide. Different pollutants as fly ash, sulphur oxides, nitrogen oxides and volatile organic compounds (VOC) that are emitted during the combustion process are present in the atmosphere in such conditions that they can affect population and environment. At the present time, many kinds of processes for gaseous pollutants removal have already been adopted for industrial goal. Classical technologies like wet, dry and semi-dry flue gas desulphurization (FGD) and selective catalytic reduction (SCR) can be applied for flue gas treatment and SO_2 and NO_x emission control, but all these technologies are complex chemical processes and wastes like wastewater, gypsum and used catalyst, are generated [1,2]. The principal competing technologies for the removal of volatile organic compounds from gaseous and aqueous phase waste streams include incineration, concentration by condensation/absorption/adsorption systems and biological treatment for the toxic residue that must be disposed. These technologies are relatively expensive and considered environmentally undesirable [2].

Electron beam (EB) technology is among the most promising advanced technologies available for the treatment of flue gases, particularly in the light of the drawbacks of conventional technologies [3-6]. It is a dry scrubbing process for simultaneous SO_2 and NO_x removal with no generation of waste. Performed studies have shown that irradiation of flue gases with an electron beam can bring about chemical changes that make easier the removal of sulphur and nitrogen oxides [7]. Electron beam technology is now under investigation for its application in volatile organic compounds decomposition [8, 9]. The studies that have been carried out so far prove that the EB irradiation determines the transformation of VOCs from gases. Usually this transformation does not only lead to CO_2 and H_2O (if VOCs are hydrocarbons) but also to the formation of partial oxidation products. For the complete conversion of VOCs, the catalytic treatment is useful. The combination of a non-thermal plasma (NTP) with in situ heterogeneous catalysis, denoted as NTP-catalysis (NTPC), is considered as a promising method to improve the oxidation of VOCs and the energy efficiency of the plasma process [10,11].

The electrical energy consumption is evaluated to be about 2 to 4 per cent of total electrical energy produced in the power station for effective purification of flue gases by EB irradiation. Thus, for industrial scale installation, the problem of reducing the electrical energy consumption for flue gases cleaning as well as the electron beam cost is important.

In view of these arguments, we have been attracted, during the last few years, to the concept of microwave (MW) energy additional use to electron beam energy [12-17]. The main idea of this method is to combine the advantages of both energy sources, electron beam and microwave in gas energizing process, i.e. electron beam high efficiency in producing high densities of free electrons, radicals and other species with microwave remarkable easiness in volumetric producing and sustaining active species to optimum level, in order to enhance gaseous pollutants removal efficiency and to reduce the electrical energy consumption and removal costs. Also, in the case of VOCs removal, the use of the catalyst under the EB or MW irradiation causes its activation, and as a consequence the temperature of the oxidation processes decreases.

The additional use of MW to the EB energy could increase the number of active species due to the presence of a higher number of free electrons induced by a possible free electron multiplication mechanism. This effect depends on several parameters, such as MW electric field amplitude, field distribution, energy distribution, and MW applicator geometry. Although extensive prior art exists in the MW field, our attention was focused on the improved design of some microwave applicators which are especially adapted to plasma processes at high power level for both MW and EB.

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Table 1
CHARACTERISTICS OF THE INSTALLATIONS

Installation	Radiation source	Reactor type	
SPI-1		EB-NTP	MW-NTP
	EB	ALIN-10 electron linear accelerator of 6.23 MeV and power up to 164 W	
	MW		MW source of 2.45 GHz and power up to 850 W
SPI-2		EB+MW-NTP	
	EB	ILU-6M electron linear accelerator of 1.8 MeV and power up to 10.8 kW	
	MW	MW source of 2.45 GHz and power up to 4.2 kW	

In view of these arguments, two innovative semi-pilot installations (SPIs) for SO_2, NO_x and VOCs removal were carried out.

Experimental part

Two innovative semi-pilot installations (SPIs) for SO_2, NOx and VOCs removal were developed:

1)SPI-1 for experiments with successive EB non-thermal plasma (EB-NTP) and MW non-thermal plasma (MW-NTP) processes into two distinct reactors, both containing inside a catalyst bed in the case of VOCs removal;

2)SPI-2 for experiments using simultaneous EB and MW irradiation (EB+MW-NTP processes) in the same reactor, having inside a catalyst bed in the case of VOCs removal.

The main components of the two installations are presented in figures 1 and 2, and the characteristics of the irradiation sources are given in table 1.

For SP-1, two distinct reactors were built: one for EB-NTP process and other for MW-NTP process, namely EB reactor (EBR) and MW reactor (MWR). At the bottom area of the EBR was installed a V_2O_5 catalyst layer. The electron beam is introduced at the front area of the EBR through

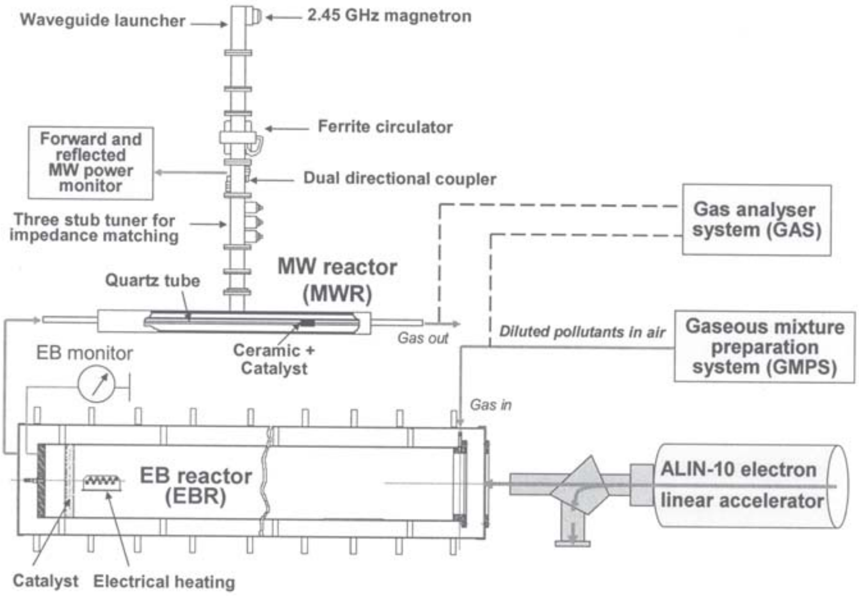

Fig. 1. Schematic drawing of the SPI-1 installation

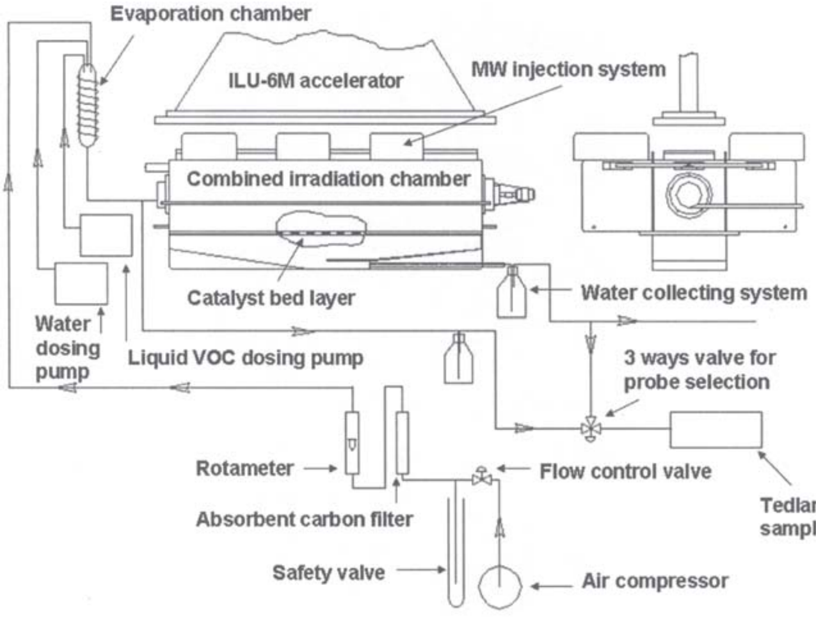

Fig. 2. Schematic drawing of the SPI-2 installation adapted for VOCs removal

two entrance windows made of 100 μm-thick aluminum foils. Because EBR is designed like a Faraday cage consisting of two concentric metallic cylindrical vessels, the EB current was collected, integrated and displayed on the control desk. The MWR is an elongated cylindrical cavity adapted to operate in the TM_{010} mode and is excited with a rectangular waveguide propagating the microwave electric field from a 2.45 GHz magnetron, parallel to its axis. Inside MWR is placed a concentric quartz tube containing a V_2O_5 catalyst bed.

For SP-2 was built a multimode rectangular reactor, MRC, used as EB+MW-reactor. MRC contain an EB window of 100 μm thick Al rectangular foil and six magnetrons of 2.45 GHz and 700 W maximum output power each, supplied by separate power controlled electrical sources.

For both SPI-1 and SPI-2, the gaseous mixture preparation system (GMPS) mainly consists of: a gaseous mixing chamber, a water dosing pump, a NH_3 dosing pump (in the case of SO_2 and NOx diluted in air), VOCs dosing pumps (in the case of VOCs diluted in air), an air compressor, reducing valves, flow meters, evaporation chambers, safety valves, electrical heating, buffer vessels, temperature controller devices and others.

The gas analyzing system, GAS, consists of:

1)A gas analysis system, EDNA-600 series (HORIBA, Japan), for experiments with SO_2 in the range 0 – 5000 ppm and NOx in the range 0 – 1000 ppm;

2)A system of three gas analyzers, for experiments with VOCs: TLV Panther Industrial Precision PID Monitor, Fisons 8330 Gas chromatograph and Buck Scientific-Multiple Gas Analyser #1.

Results and discussions

In order to achieve a better assessment on both removal and oxidation processes, two efficiency types have been determined: removal efficiency, Er (the efficiency of converting VOCs into any products) and oxidation efficiency, Eo (the efficiency of the VOCs conversion into carbon oxides) (eq. 1 and 2):

$$\mathrm{Er} = \frac{[VOC]_0 - [VOC]_t}{[VOC]_0} \times 100 \qquad (1)$$

$$\mathrm{Eo} = \frac{[VOC]_{ox}}{[VOC]_0} \times 100 = \frac{\dfrac{[CO_2]_t - [CO_2]_0 + [CO]_t}{N.C.}}{[VOC]_0} \times 100 \qquad (2)$$

where:

$[VOC]_0$ is the initial concentration of VOC, (ppmv);

$[VOC]_t$ - the concentration of VOC after treatment, (ppmv);

$[VOC]_{ox}$ - the amount of VOC completely oxidized (determined from the concentrations of CO_2 and CO after treatment);

$[CO_2]_0$ - the initial concentration of CO_2, (ppmv);

$[CO_2]_t$ and $[CO]_t$ - the CO_2 and CO concentrations, respectively (ppmv), after treatment; N.C. is the average number of carbon atoms of VOCs.

Figures 3-10 of this paper present only several representative results performed with SPI-1 and SPI-2, in order to compare the effects of different treatments: EB-NTP, MW-NTP and EB+MW-NTP on SO_2, NOx and VOCs removal in the following conditions:

1) SO_2 and NOx of different concentrations in gaseous mixture containing air, NH_3 (given as stoichiometric ratio $SR(NH_3) = NH_3/(2\times SO_2+NOx)$), CO_2 (10%) and H_2O (7%).

2) Hexane (H) and Toluene (T) separate or together (H+T) of different concentrations in gaseous mixture containing air and H_2O (7%).

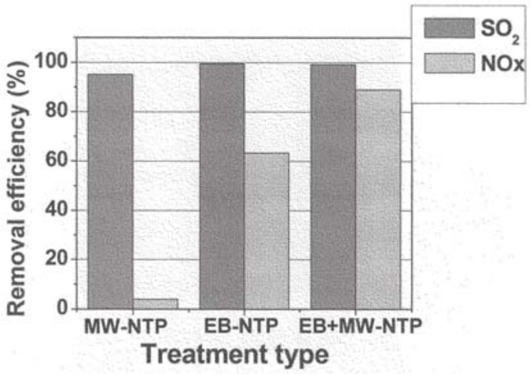

Fig. 3. Effect of treatment type (MW, EB and MW+EB(succesive)) on SO_2 and NO_x removal from flue gases. Initial concentrations: $[SO_2]_i$=2000 ppmv; $[NO_x]_i$=400 ppmv; [water vapors]$_i$=7%, $[CO_2]_i$=10%. Stoichiometric ratio of NH_3 = 0.8. Flow rate of gases = 1000 L/h; temperature = 65÷70^{0}C. Power used for irradiation: P_{MW} = 0.7 kW, P_{EB} = 5 W. Installation used SPI-1.

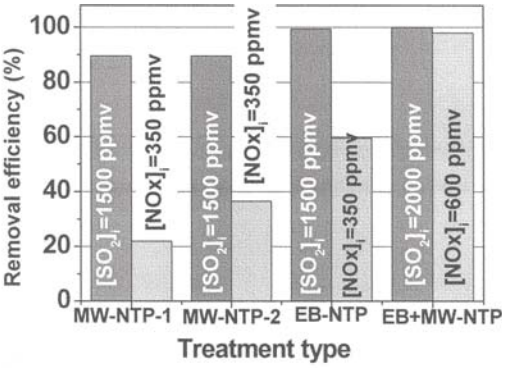

Fig. 4. Effect of treatment type (MW, EB and MW+EB (simultaneous)) on SO_2 and NO_x removal from flue gases. Initial concentrations: $[SO_2]_i$ = 1500÷2000 ppmv; $[NO_x]_i$=350÷600 ppmv; [water vapors]$_i$=7%, $[CO_2]_0$=10%. stoichiometric ratio of NH_3 = 1.16. Flow rate of gases = 1500 L/h; temperature = 65÷70^{0}C. Power used for irradiation: P_{MW} = 1.5 kW for MW-NTP-1 and 1.85 kW for MW-NTP-2, P_{EB} = 88 W for EB-NTP and P_{MW}=1.18 kW + P_{EB}=88 W for EB+MW-NTP. Installation used: SPI-2.

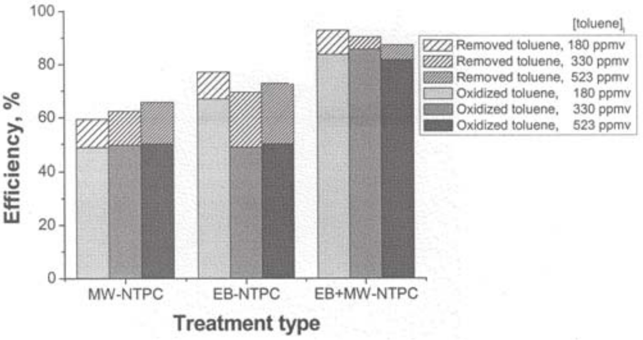

Fig. 5. Effect of treatment type (MW, EB and EB+MW (successive)) on catalytic toluene removal from air. Initial concentration of toluene: 180; 330 and 523 ppmv. Initial concentration of water vapors 7%. Flow rate of gases = 400 L/h; catalyst temperature = 200^{0}C. Power used for irradiation: P_{MW}=400W and P_{EB}=105W. Installation used: SPI-1.

Two kinds of EB+MW-NTP processes were used: EB-NTP and MW-NTP successively applied in SPI-1 and EB-NTP and MW-NTP simultaneously applied in MRC of SPI-2.

The analysis of these results led to the following main observations and conclusions:

- all treatment types, EB-NTP, MW-NTP and EB+MW-NTP are very effective for SO_2 removal at low and high

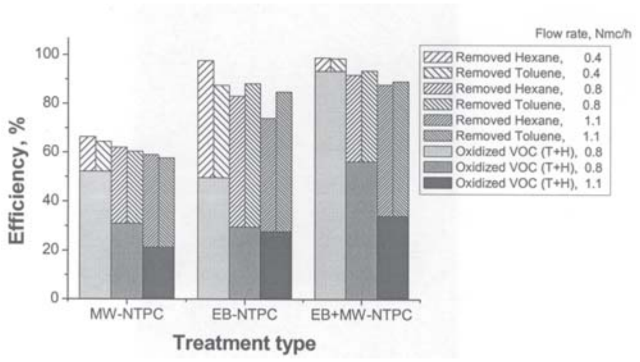

Fig. 6. Effect of treatment type (MW, EB and EB+MW (successive)) and air flow rate on catalytic toluene and hexane removal from air. Initial concentrations: $[toluene]_i$=285 ppmv, $[hexane]_i$=285 ppmv, $[water\ vapors]_i$= 7%. Flow rate of gases = 400, 800, 1100 L/h. Power used for irradiation: P_{MW}=400W and P_{EB}=105W. Installation used: SPI-1.

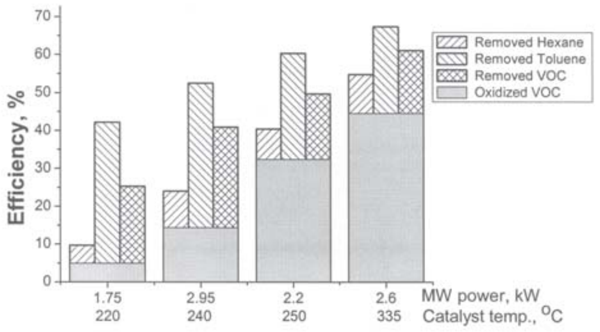

Fig. 7. Effect of MW power used on catalytic toluene and hexane removal from air. Initial concentrations: $[toluene]_i$=1000 ppmv, $[hexane]_i$=1000 ppmv, $[water\ vapors]_i$= 7%. Flow rate of gases = 1000 L/h. Power used for irradiation: P_{MW}=1.75÷2.6 kW. Installation used: SPI-2.

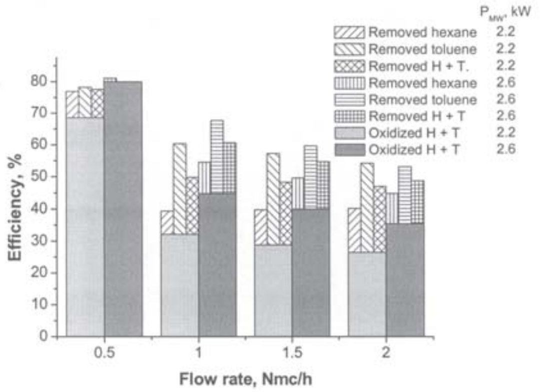

Fig. 8. Effect of MW power used and air flow rate on catalytic toluene and hexane removal from air. Initial concentrations: $[toluene]_i$=1000 ppmv, $[hexane]_i$=1000 ppmv, $[water\ vapors]_i$= 7%. Flow rate of gases = 500÷2000 L/h. Power used for irradiation: P_{MW}=2.2÷2.6 kW. Installation used: SPI-2.

initial SO_2 concentrations ($[SO_2]_i$), as well as at low and high flow rates (figs. 3 and 4). There can be several pathways for SO_2 oxidation, depending on the conditions [18]. The most important are the radio-thermal and thermal reactions [19]. Radio-thermal reactions proceed through radical oxidation of SO_2 in the following reaction:

$$SO_2 + {}^{\cdot}OH + M \rightarrow HSO_3 + M \qquad (3)$$

Then HSO_3 creates ammonium sulfate in the following steps:

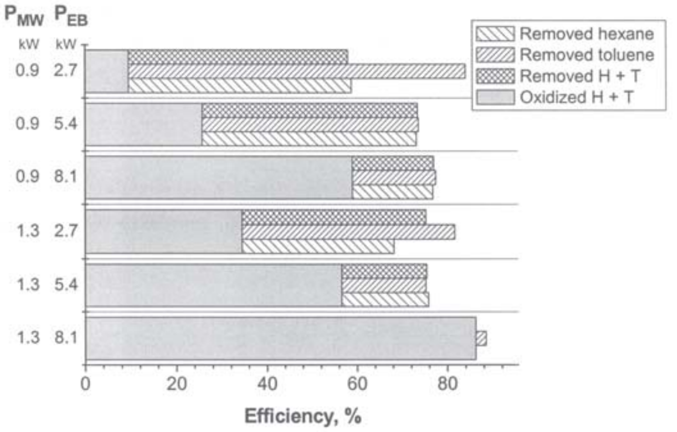

Fig. 9. Effect of power used for simultaneous irradiation (EB+MW) on catalytic toluene and hexane removal from air. Initial concentrations: $[toluene]_i$=1000 ppmv, $[hexane]_i$=1000 ppmv, $[water\ vapors]_i$= 7%. Flow rate of gases = 1000 L/h. Power used for irradiation: P_{MW}=0.9÷1.3 kW, P_{EB}=2.7÷8.1 kW. Installation used: SPI-2.

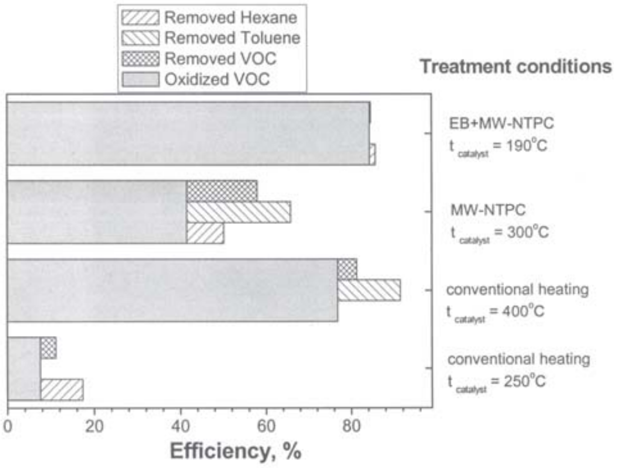

Fig. 10. Effect of treatment type (conventional heating of catalyst, microwave non thermal plasma catalysis (MW-NPTC) and electron beam + microwave non thermal plasma catalysis (EB+MW-NTPC) on catalytic toluene and hexane removal from air. Initial concentrations: $[toluene]_i$=1000 ppmv, $[hexane]_i$=1000 ppmv, $[water\ vapors]_i$= 7%. Flow rate of gases = 1000 L/h. Installation used: SPI-2.

$$HSO_3 + O_2 \rightarrow SO_3 + HO_2 \qquad (4)$$
$$SO_3 + H_2O \rightarrow H_2SO_4 \qquad (5)$$
$$H_2SO_4 + 2NH_3 \rightarrow (NH_4)_2SO_4 \qquad (6)$$

The thermal reactions are [19]:

$$2SO_2 + 3NH_3 + 2\,H_2O \rightarrow (NH_4)_2SO_3 + NH_4HSO_3 \ \ (main\ reaction) \qquad (7)$$

$$2SO_2 + 3NH_3 + 2\,H_2O + O_2 \rightarrow (NH_4)_2SO_4 + NH_4HSO_4\ (side\ reaction) \qquad (8)$$

- In our conditions (temperature, humidity and oxygen content), the thermal reactions are the most important. The values near 100% for SO_2 removal efficiency are easily obtained even by MW-NTP, which is generally less effective than EB-NTP for gaseous pollutants removal. The experimental results suggest that the MW-NTP process could become a viable and commercial attractive method for SO_2 removal in comparison with other technologies.

- Several pathways are known for NO oxidation. In the case of electron beam treatment, the most important are as follows [20]:

$$NO + O(^3P) + M \rightarrow NO_2 + M \tag{9}$$
$$O(^3P) + O_2 + M \rightarrow O_3 + M \tag{10}$$
$$NO + O_3 + M \rightarrow NO_2 + O_2 + M \tag{11}$$
$$NO + HO_2^{\cdot} + M \rightarrow NO_2 + {}^{\cdot}OH + M \tag{12}$$

After the oxidation, NO_2 is converted into nitric acid in the reaction with $^{\cdot}OH$:

$$NO_2 + {}^{\cdot}OH + M \rightarrow HNO_3 + M \tag{13}$$

HNO_3 aerosol reacts with NH_3, giving ammonium nitrate:

$$HNO_3 + NH_3 \rightarrow NH_4NO_3 \tag{14}$$

NO is partly reduced to atmospheric nitrogen during the reaction with radicals obtained from NH_3.

- The NOx removal efficiency is much smaller than SO_2 removal efficiency for both EB-NTP and MW-NTP processes, but it strongly increases up to 89% by EB+MW-NTP applied in SPI-1 and up to 98% by EB+MW-NTP applied in SPI-2 (figs. 3 and 4). In addition, NOx removal efficiency is less effective by MW-NTP than by EB-NTP technology. Depending on many parameters, such as gas composition (pollutants concentration, SR(NH_3) and H_2O concentration), gas mixture temperature (t_{GM}), gas flow rate (FR), MW power (P_{MW}) level and EB power (P_{EB}) level, the maximum value obtained for NOx removal efficiency was in the range of 75-85% for EB-NTP technology and up to 6% for MW-NTP applied in SPI-1 and up to 40% for MW-NTP applied in SPI-2. It is important to note that in the case of MW-NTP process the most important parameter for NOx removal increasing is P_{MW}. It was observed that NOx removal rises slowly but significantly versus P_{MW} increasing.

- In the case of VOCs decomposition, the process itself is based on the similar principles as primary reactions concerning SO_2 and NOx removal, i.e. free radicals attack on organic compounds chains or rings causing VOCs decomposition [5]. For aromatic hydrocarbons, VOCs decomposition will mainly go through [18]:

1. Positive ions charge transfer reaction:

$$M^+ + RH\ (RH = VOC,\ e.g.\ Benzene\ or\ PAHs) = M + RH^+ \tag{15}$$

Because RH has low ionization energy (for example benzene: IE = 9.24 eV), part of VOC will be decomposed by rapid charge transfer reactions.

2.Radical—neutral particles reactions, $^{\cdot}OH$ radical plays a very important role for VOC decomposition, especially when water concentration is high. $^{\cdot}OH$ radicals react with VOC in two ways:

$^{\cdot}OH$ radical addition to the aromatic ring (e.g. toluene):

$$^{\cdot}OH + C_6H_5CH_3 \rightarrow R_1^{\cdot} \tag{16}$$

and H atom abstraction (for the alkyl-substituted aromatic compounds and for n-alkanes) or H atom elimination (for benzene, naphthalene and the higher polycyclic aromatic hydrocarbons):

$$C_6H_5CH_3 + {}^{\cdot}OH \rightarrow R_2^{\cdot} + H_2O \quad \text{(H atom abstraction)} \tag{17}$$
$$C_6H_6 + {}^{\cdot}OH \rightarrow C_6H_5OH + H \quad \text{(H atom elimination)} \tag{18}$$

Radicals ($R_1^{\cdot}$, $R_2^{\cdot}$) formed above will go though very complex reactions: O_2 addition, O atom release, aromatic –CHO (-dehydes), –OH (-ol) formed compounds or ring cleavage products:

$$R^{\cdot}\ (R_1^{\cdot}, R_2^{\cdot}) + O_2 \rightarrow RO_2^{\cdot} \tag{19}$$
$$2\ RO_2^{\cdot} \rightarrow 2RO^{\cdot} + O_2 \tag{20}$$
$$RO_2^{\cdot} + NO \rightarrow RO^{\cdot} + NO_2 \tag{21}$$
$$RO^{\cdot} + O_2 \rightarrow HO_2^{\cdot} + products\ (aromatic\ –CHO,\ \text{-}O) \tag{22}$$
$$RO^{\cdot} \rightarrow aliphatic\ products \tag{23}$$

For n-alkanes, only radical-neutral particles reactions are possible.

If only the EB irradiation is used, the main reaction products are the aerosols, with a complex chemical composition. Our objective was to combine EB irradiation with the catalytic oxidation, in order to transform the main part of the active species into total oxidation products (carbon oxides and water, in the hydrocarbons case). For the process efficiency computation, it is enough to determine initial and final VOC concentrations and the formed carbon oxides quantities. The process is considered optimum when VOCs are removed and transformed in the same time into carbon oxides.

- The results presented in figure 5 and figure 6 show that the experiments performed with EB+MW-NTP-catalysis (EB+MW-NTPC) applied in SPI-1 give higher values than separate EB-NTPC or separate MW-NTPC, for both efficiencies, Er and Eo. As shown in figure 5 the toluene removal and oxidation efficiencies increase significantly for the EB+MW-NTPC treatment as compared with MW-NTPC and EB-NTPC treatments, at low and at high toluene initial concentrations. The results presented in figure 6 illustrate the influence of airflow rate and treatment type on removal and oxidation efficiencies for a mixture of hexane and toluene. In these cases, the best values are obtained for a combined EB + MW + catalyst, but the influence of flow rate is very strong, only at low flow rates the reactor residence time is enough effective to obtain meaningful efficiencies.

- The results presented in figures 7 and 8 are obtained by experiments performed with SPI-2. All efficiencies, Er(H), Er(T), Er(H+T) and Eo(H+T) increase continuously versus P_{MW} level but Eo(H+T) increases strongly versus P_{MW}, starting with 2.2 kW (fig. 7). Er(T) is always higher than Er(H) in the P_{MW} tested range from 1.75 kW to 2.6 kW (fig. 7). At low FR (0.5 m^3/h) and high P_{MW} levels (2.2 kW and 2.6 kW) there is no notable difference between Er(H), Er(T) and Er(H+T) but Eo(H+T) strongly increases compared to the values obtained at FR of 1 m^3/h and 2 m^3/h (fig. 8). The FR increasing from 1 m^3/h to 2 m^3/h does not substantially modify the level of Er(H), Er(T), Er(H+T) and especially the level of Eo(H+T) (fig. 8).

- The results presented in figure 9 show that Eo(H+T) increases strongly, up to 85%, versus $P_{EB}+P_{MW}$ for EB+MW-NTPC applied in SPI-2. For a given level of $P_{EB}+P_{MW}$ there are little differences between Er(H), Er(T) and Er(H+T). This fact means that EB+MW-NTPC perform the same Er for the two different pollutants, H and T (fig. 9).

- The comparative results obtained by catalytic oxidation with classical heating (CH), MW-NTPC and EB+MW-NTPC (fig. 10) demonstrate that satisfactory removal efficiencies are obtained with CH only when the temperature of the catalyst exceeds 350°C. With MW-NTPC, the oxidation process begins at a lower temperature and good efficiencies can be obtained in the range of 250 - 270 °C. With EB+MW-NTPC, high removal efficiencies begin in the range of 175 - 200 °C.

- The EB+MW-NTP (or EB+MW-NTPC) processing obtained by combined EB-NTP (or EB-NTPC) with MW-

NTP (or MW-NTPC) into two different reactors (SPI-1), EBR and MWR, connected in series, is less effective than EB+MW-NTP (or EB+MW-NTPC) process obtained by simultaneous application of EB-NTP (or EB-NTPC) and MW-NTP (or MW-NTPC), in the same reactor (SPI-2). However, successive application of EB-NTP (or EB-NTPC) and MW-NTP(or MW-NTPC) into two distinct reactors connected in series is more easily to be performed from the technological point of view.

Conclusions

The electron beam treatment of flue gases is well known and applied on an industrial scale for the conversion of SO_2 and NO_x from flue gases into fertilizers (a mixture of ammonium sulphate and ammonium nitrate). The main idea of our research was to combine the complementary characteristics of several gas treatment techniques:

- the very high efficiency of EB in generating very reactive species (OH radical especially) which are efficient in the oxidation of pollutants;
- the ability of MW to produce and sustain non-thermal plasma in large electrode-less reaction vessels.

When using combined irradiation (EB+MW), the SO_2 removal is done more easily. The removal process of NOx is more difficult and, as a result, it is necessary to use higher EB irradiation doses. The fact that the NOx removal efficiency rises with the increase of the microwave power used (at the same EB irradiation dose) makes us believe that good results could be obtained when a part of the needed EB power is replaced by the MW (the MW irradiation is much cheaper than the EB irradiation).

If a proper catalyst is added this process can also be used for the removal of VOC. The MW has the capacity of selectively heating the oxidation catalyst, which makes the catalytic oxidation possible at gas temperatures which are lower than in the conventional heating. The EB can also activate the catalyst and by generation of active species the VOC are easily converted into oxidation products.

The experimental results demonstrate that the EB+MW-NTPC treatment improves both removal and oxidation efficiencies with respect to those obtained by EB-NTPC or MW-NTPC.

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