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A NEW APPROACH IN  $e$ -BEAM TREATMENT  
OF FLUE GASES  
AND OTHER GAS-PHASE PROCESSES

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Новый подход к электрон-пучковой обработке отходящих газов и других газофазных процессов

Предлагается принципиально новый подход к электрон-пучковой технологии очистки отходящих газов ТЭЦ, металлургических предприятий, котельных агрегатов и т.д. без использования аммиачного метода. Предлагаемый подход включает две стадии. На первой отходящий газ подвергается воздействию жесткого ультрафиолетового излучения (с подходящим спектром) в постоянном электрическом поле, что приводит к частичному разделению окислов NO и SO<sub>2</sub> от остальной массы газа. На второй стадии приготовленная таким образом газовая смесь с частично отделенными окислами азота и серы подвергается комбинированному воздействию электронного пучка с энергией 300 – 400 КэВ и быстрой газовой струи, состоящей из смеси газов N<sub>2</sub> + H<sub>2</sub>. В условиях данной геометрии эта активированная газовая струя приводит к восстановлению одновременно NO до N<sub>2</sub> + H<sub>2</sub>O и SO<sub>2</sub> до свободной серы и воды вместо нитратов и сульфатов аммония в традиционном подходе «Эбара». Помимо этого, предложенный двухступенчатый процесс может привести к значительному уменьшению энергопотребления (энергии быстрых электронов) и резкому снижению фона гамма-квантов. В итоге процесс имеет намного более высокий потенциал для коммерциализации и широкого распространения.

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A New Approach in *e*-Beam Treatment of Flue Gases and Other Gas-Phase Processes

A principally new approach avoiding the conventional ammonia (or limestone) technology for the *e*-beam treatment of flue gases in coal-fired electric power stations, boilers and ore smelting industries is suggested. The approach includes two stages. At the first stage the flue gases undergo hard ultra-violet irradiation (with an appropriate spectral distribution) in permanent electric field which leads to a partial separation of NO and SO<sub>2</sub> from the rest of flue gases. At the second stage this partially separated mixture of NO and SO<sub>2</sub> undergoes a combined impact of a fast gaseous jet consisting of a molecular mixture N<sub>2</sub> + H<sub>2</sub> together with longitudinal collinear electron beam with maximal electron energy around 300 – 400 KeV only. This low energy electron irradiation leads in case of such a geometry to a simultaneous reduction of NO and SO<sub>2</sub> oxides to the clear air components (i.e. N<sub>2</sub> + H<sub>2</sub>O) and elementary sulphur respectively instead of a large mass of ammonium nitrates and sulphates in the traditional Ebara approach. Besides, the suggested two-stage process may lead to a very significant reduction in the energy consumption (of the accelerated electrons) and a sharp decrease in the gamma-radiational background. Due to its characteristic features, the process has a much higher potential for commercialisation and wide spreading.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR and at the D.V.Skobel'syn Institute of Nuclear Physics, Lomonosov Moscow State University.

Communication of the Joint Institute for Nuclear Research. Dubna, 1999

# 1 Introduction

In the last decades the ecological problems are rousing all growing interest around the world of both the experts and also general public (e.g. in the form of rapidly rising activity of the "Greens" and the Greenpeace movement). It is undoubtedly related to serious threats to the mankind surviving and to modern civilization itself from the technogenic factors and antropogenic activities which destroy the environmental natural biota. It is quite enough to note here, as an example, the demolition of the Earth's ozone layer or greenhouse effect which results in continuous rise of the average temperature in the northern hemisphere for last 50 years.

Among all these global ecological problems the large scale emission of hazardous nitrogen and sulphur oxides into the atmosphere from many large industrial units (e.g. in oil- and glass industries, metallurgy, power- and electric stations, coal-fired boilers etc.) evokes all growing anxiousness of the public in many countries. It is because these oxides, subjecting to many transformations in atmosphere, lead to hazardous pollutions, acid rains, smog and general poisoning of the environment. They also effect somehow on the demolition of the ozone layer etc [1,2]. As a result, emission control legislation has been enacted in many countries, especially in USA and Germany. E.g. the new source performance standards enacted in the US call for a reduction in the sulphur dioxide emission from coal-fired utility boilers of 70 to 90%, depending on the level of the uncontrolled emission. The Federal Republics of Germany has set a target stack-gas sulphur dioxide concentration of 140 mL/L, with a ten-year compliance period for both new and old plants. More stringent sulphur dioxide/nitrogen oxide emission control legislation is expected in the near future.

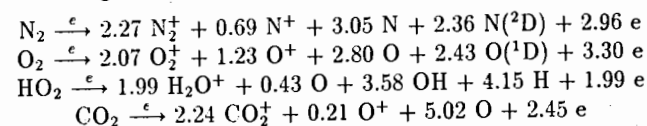
Several different technologies for the control of nitrogen oxide and sulphur emission from coal-fired electrical generating stations are currently available [1-3]. Of these only the wet lime/limestone flue gas desulphurization (FGD) technique for sulphur dioxide emission control has seen extensive implementation at the utility scale in the US. In the FGD technology, a slurry of lime, or limestone, in water is injected into the flue gas stream in a spray tower. The sulphur dioxide in the flue gas reacts with the lime slurry and produces a mixed calcium sulphite/calcium sulphate sludge. The major drawbacks associated with the FGD technology, such as low process unit availability, the need for reheating the stack gas and sludge disposal problems, have led the utilities to consider alternative emission control strategies. The characteristics of an ideal emission control technology would include minimal capital investment, low input of energy and chemical reagents, low maintenance, and easily saleable by-product with a large potential market.

In recent years for the solution of the above very complicated problems some principally new approaches such as a e-beam treatment have got an essential development. These approaches employ high power electron beams generated by industrial electron accelerators [3-10]. Two radiation-based processes for de-NO<sub>x</sub> and de-SO<sub>x</sub> from boiler flue gasses have been developed:

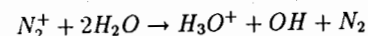
- so called EBARA process [2-4];
- Research-Cottrel process (see e.g. D.J. Helfritch and P.L. Feldman, Radiat.Phys.Chem. 24, 129 (1984)).

The EBARA process (or Ebara-like processes) is currently considered as the most well developed radiation-based technology for the removal of NO<sub>x</sub> and SO<sub>2</sub>. The process consists from a few stages. In the first step, the flue gases from burner with temperature 800-1000°C are cleaned from fly ash and are fed to a spray tower which is supplied with

a very fine water sprayer. After passing the tower the temperature of the flue gases decreases down to 70-80°C. At the second step, an ammonia water is admixed to the cooled flue gas and the gas mixture is fed to reaction vessel for radiational treatment. In the vessel the following basic reactions proceed [11]:



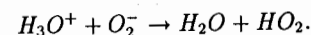
with a subsequent transformation of the radicals and ions as follows:



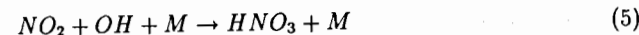
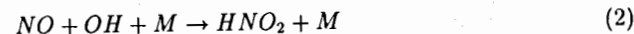
(the water decomposition with a charge transfer)



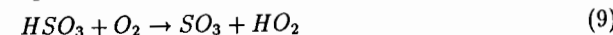
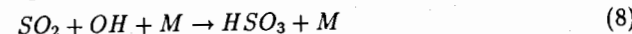
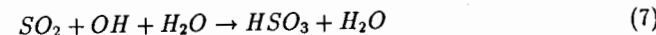
and a subsequent neutralization of the ions:



Thus one pair of ions N<sub>2</sub><sup>+</sup> and O<sub>2</sub><sup>-</sup> offers one radical OH and one radical HO<sub>2</sub>. Now one can outline the basic ways for de-NO<sub>x</sub> and de-SO<sub>2</sub> in the Ebara process [6-8, 12]:



and also for SO<sub>2</sub>:



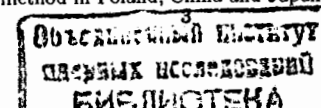
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As a result of such reactions the NO<sub>x</sub> and SO<sub>2</sub> oxides, at presence of the water, first transform to nitric and sulphuric acids and these eventually react with ammonia giving ammonium salts (nitrates and sulphates).

And in the last stage, these fine powder-like salts are gathered in electrostatic precipitator and clear flue gases are exhausted into the air. These salts are rather good fertilizers and as a by-product may improve noticeably the commercial potential of the Ebara process.

The removal degree for SO<sub>2</sub> and NO<sub>x</sub> in this process may reach 90% and thus the process can meet the recent emission control requirements [6-8]. Today almost all the details of the Ebara process seem to be well studied and the process can be considered as one of the most promising for wide dissemination [13,14]<sup>1</sup>.

<sup>1</sup>The solid proof for this is a real construction of full scale industrial cleaning units for flue gas e-beam treatment based on the Ebara method in Poland, China and Japan.



## 2 Disadvantages of the Ebara process

Unfortunately this method also suffers from many serious disadvantages which noticeably prevent its widespread implementation. Let's discuss these disadvantages in a descending order of their importance.

(i) One of the main drawback of Ebara technology is very high e-beam energy consumption, i.e. high radiational doses around 1 Mrad or so [4-8,13]. In a previous version of the process the energy consumption was even higher 1 Mrad at the degree of SO<sub>2</sub> and NO<sub>x</sub> removal being less than 85%. As a result of these high doses required the total e-beam energy consumption for the typical energy station with power 500 MWt might reach 5 MWt and higher which is hardly reachable with the modern technology for the electron accelerator production. In any way the capital costs for such accelerator set should be enormously high.

(ii) It is evident that at such a very high power of electron accelerators as given above, the gamma-ray background also should be very intensive. And thus the cost of radiational shielding will be also rather high. This important drawback constitutes another negative factor of the Ebara method.

(iii) Third negative factor of the method is related to the hazardous ammonia utility. This also gives rise to serious ecological problems. It is necessary to bring regularly to the plants a large amount of ammonia and to remove even much larger amounts of ammonium salts. This regular delivery of ammonia can lead to some ecological risks related to a uncontrolled leakage of ammonia to the atmosphere.

(iv) And finally the last negative moment is related to large-scale utilization of by-product in the Ebara process. In a number of recent reviews (see e.g. [13,14]) the production of the ammonium salts is considered as an additional benefit of the technology because the commercial sale of the by-product will decrease the cost of e-beam treatment of flue gases.

Unfortunately it is only true in a near short time perspective. And in a more long term perspective, this conclusion is likely not true. It is due to the fact that at rather wide-scale dissemination of the Ebara process the amount of ammonium salts, especially for ammonium sulphate, will much exceed all future needs in such salts and inevitably the serious problems of their disposal will appear. For example, according to the estimations made by German authors in the middle of the 80-ies for the US [15] the utilization of the whole ammonium sulphate produced (with the Ebara technology) by only one middle power station in USA will cover a half of a year's need in the sulphate in the country! Thus, if even to use the highly increased estimates for the needs in the (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> over the world [13], even in such a case the utilization of the whole ammonium sulphate produced from coal fired electrical generating stations in one country only (e.g. in Germany) will cover with an excess all-world needs in this substance [15]. Thus, in any case the serious disposal problem is remained.

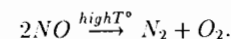
Hence one can summarize the whole situation with the Ebara method as follows: this method, by solving one ecological problem, will give rise simultaneously to the new ones, generally quite serious, such as:

- high radiational gamma background;
- some risk of the ammonia leakage during the ammonia transportation;
- utilization of huge amounts of ammonium salts.

All the above factors enforce to look for other alternative approaches to the treatment of flue gases which are free from these drawbacks.

## 3 New approach to removal of NO<sub>x</sub> and SO<sub>2</sub> from flue gases

First of all it is worth to try to circumvent the ammonia technology at all. For this goal one can use an electron-beam version of the so called "Exxon" process [16-18]:



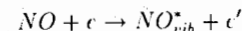
The Exxon process is based on the observation that at very high temperatures T° ~ 2000°K and higher the nitrogen oxide (which is thermodynamically unstable) will dissociate back into free nitrogen and oxygen but with a small rate constant. However at higher temperatures the dissociation rate increases significantly. see e.g. the Table 1.

Table 1. Dependence of the characteristic time for the NO dissociation at normal pressure on temperature.

T° K 10 <sup>3</sup>	1.0	1.7	2.0	2.3	3.0	4.0
Dissoc.time,s	2.2 10 <sup>12</sup>	140	1	5.3 10 <sup>-3</sup>	7.8 10 <sup>-5</sup>	7.2 10 <sup>-7</sup>

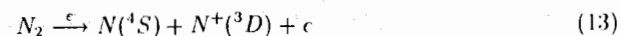
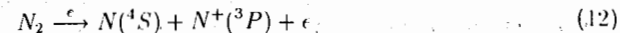
From the table it is seen the dissociation time of NO at normal pressure is exponentially shortened (and the rate constant grows respectively) and thus the dissociation rate at 2300° is rather high.

Electron beam, at appropriate conditions (e.g. in decreasing the average energy of the electrons down to a few KeV [19]) should initiate strongly the excitation of the vibrational modes in NO:

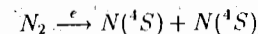


so that in case of efficient pumping only the 2-3 vibrational quanta in the NO molecule the Exxon process will proceed with a noticeable rate at the temperatures around 800-1000° C really existing in flue gases.

Another efficient way for inverse conversion of NO into N<sub>2</sub> and O<sub>2</sub> is a selective excitation and subsequent dissociation of the gaseous nitrogen by electron beam while feeding the nitrogen into the reaction zone in the form of a fast gaseous jet by the following chain of reactions:



and also



and further



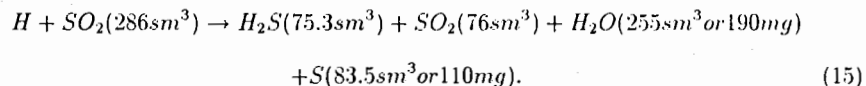
etc.

In such a process the nitrogen oxide will be reduced to N<sub>2</sub>+O<sub>2</sub> rather than be oxidized to NO<sub>2</sub>. However due to very short life time of N-radicals the decisive condition for carrying out such a reduction of the NO to the free nitrogen will be a process geometry and the velocity of the reagent mixing. We propose to realize the feeding nitrogen atoms into the reaction zone by combining the electron beam collinearly with a fast gaseous jet consisted of the mixture N<sub>2</sub>+H<sub>2</sub>, as displayed on Fig.1.

At such a geometry for fast electrons and reagent feeding into the stream of flue gases along the axis of a side pipe socket A-A, some excessive nonequilibrium concentration of

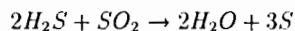
free nitrogen and hydrogen atoms and also of free electrons appears inside the main duct. All these three active components (i.e.  $\epsilon + N^* + H^*$ ) will be mixed up to the main stream of flue gases incorporating NO and SO<sub>2</sub> and will result in a reduction of the NO to the N<sub>2</sub> due to the reactions (12)–(14) and the SO<sub>2</sub> to the free sulphur (see below).

The latter process has been studied as early as in 30-ies [20] by Hartek et al. The authors have found that by adding free H-atoms to SO<sub>2</sub> the following products have appeared:



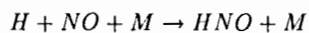
In other words, in the process (15), three quarters of the initial quantity from some initial amount of SO<sub>2</sub> oxide are transformed into H<sub>2</sub>S, H<sub>2</sub>O and free sulphur.

It is also well known from the oil chemistry that at higher temperatures and/or presence of a catalyst the H<sub>2</sub>S reacts easily with SO<sub>2</sub> giving a free sulphur and water (the so called Klaus process). Thus, being radiationally initiated the reaction:

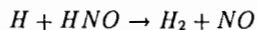


will easily proceed.

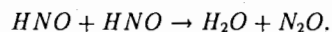
As a result of such a treatment of the flue gases with fast jet of free atoms (N+H) and at appropriate conditions (e.g. at high temperatures) there appears mainly (N<sub>2</sub>+H<sub>2</sub>O+free S) together with a small amount of N<sub>2</sub>O from the reaction:



with subsequent reactions:



and



Certainly some residual small amounts of SO<sub>2</sub> and NO will appear as well.

This, instead of huge volumes of ammonium salts which are emerged in the Ebara process, there appear in our approach only the components of the clear air and relatively small amount of free sulphur. The latter represents likely the more costly by-product as compared to the ammonium sulphate emerged in the Ebara method.

To make the reduction process even more selective and efficient we propose to insert to the process some initial stage where the NO and SO<sub>2</sub> oxides will be partially separated from the rest of flue gasses. This stage includes a pretreatment of flue gases with combined action of hard ultraviolet irradiation with an appropriate spectrum and permanent electric field. The ultra-violet irradiation results in selective ionization of NO and SO<sub>2</sub> components in flue gases while the permanent electric field will lead to a partial segregation of the oxides from the rest of flue gases [23]. Thus the e-beam + gas jet treatment of a such partially segregated gas mixture should lead to noticeable enhancement of the process yield and optimization of energy losses (see Fig.1 where the scheme of the process is shown).

Now let's consider the question of energy consumption and gamma-ray background in our approach. Surely it would be rather difficult to give realistic estimates for the total power of electron beam needed in our approach. However it is highly probable that the

energy consumption and energy losses in our case will be much reduced as compared to the Ebara method. It is explained by the evident fact that in the latter method, one has to treat (by e-beam) the whole large mass of the exhaust gas in which content of NO<sub>x</sub> is only 0.5–1% or so. While the e-beam energy in our approach is spent mainly to the excitation and dissociation of gas jet fed in the main duct, the mass of the jet is being still a small fraction of the whole mass of the flue gasses. So that, if even to assume the 300% losses of active radicals generated in the jet the necessary mass of the feeding N<sub>2</sub>+H<sub>2</sub> should be much less than the total amount of the flue gases. Roughly the energy consumption in our case will be less in the same ratio.

Other advantage of the present approach is the fact the length of pipe socket with a nozzle can be made as short as only 15–20 sm whereas the density of the feeding gas can be taken as a small fraction of that for flue gases but due to a high velocity of the jet the active radicals generated in the pipe socket will penetrate deeply inside the main stream of flue gasses. Significant additional saving the energy will be also obtained due to the partial segregation of the oxides in UV pretreatment of flue gases. This pretreatment makes it possible to reduce additionally the maximal energy of the electron beam in our approach down to 300–400 KeV. So that both above facts, i.e. decrease of the total mass of the gas jet treated by e-beam in our case together with significant reduction of maximal electron energy to 300–400 KeV result in a drastic decreasing the cost of both the radiational shielding and the electronic accelerators needed in our approach.

The high efficiency of the above method may be additionally enhanced by employment of an electron accelerator with a multibeam outlet [21] recently developed in JINR. In such a construction each e-beam feeds fast electrons separately in each pipe socket with own jet. And by varying the composition of gas jet and its velocity in each channel one can achieve a maximal efficiency of the whole construction.

## 4 Conclusion

A new approach towards the radiationally stimulated removal of NO<sub>x</sub> and SO<sub>2</sub> from flue gases suggested in the present work is distinguished in a few important aspects from traditionally employed now the ammonia method "Ebara".

(i) Instead of oxidizing NO<sub>x</sub> and SO<sub>2</sub> to nitric and sulphuric acids with their subsequent transformation into ammonium salts, we use active N and H radicals generated in a high velocity gas jet by collinear e-beam to reduce the oxides to free nitrogen, sulphur and water. In this approach the employment of hazardous ammonia and all the drawbacks related to this are completely avoided.

(ii) Due to a rejection from ammonia method the total mass of by-product in our case is decreased drastically (about in 7 times).

(iii) The cost of by-product (i.e. free sulphur) in our case is also higher as compared to that of ammonium sulphate in the conventional method.

(iv) Due to much less amount of the e-beam treated mass of gas jet in our approach and reduced maximal energy of fast electrons (down to 300 KeV) the e-beam energy consumption in our case is expected to be less essentially. However the exact estimates are difficult to do on this preliminary stage. One can derive likely reliable estimates for the energy consumption and the process yield from theoretical modelling the radical kinetics in our scheme.

(v) It should be emphasized that the general approach suggested here for de-NO<sub>x</sub> and

de-SO<sub>2</sub> of flue gases can undoubtedly be employed for many other cases where one needs to treat selectively only one (or a few) component(s) in a gas mixture. In traditional e-beam technologies the electron beam spends its energy in general non-selectively. It leads often to very high doses required for conducting many radiational processes. On the contrary to this, in case of selective excitation of some sort of molecules in the gas mixture, the yield should be much increased due to decreasing the energy losses and thus the energy consumption will be reduced.

It is quite similar to catalytical processes in chemistry. It is well known that passing to a catalytic conducting some process makes it possible to enhance the process yield and to reduce the energy consumption. Using the method for de-NO<sub>x</sub> and de-SO<sub>2</sub> from flue gases suggested here, some experimental tests are started now in JINR and first results should appear in near future.

### Acknowledgments

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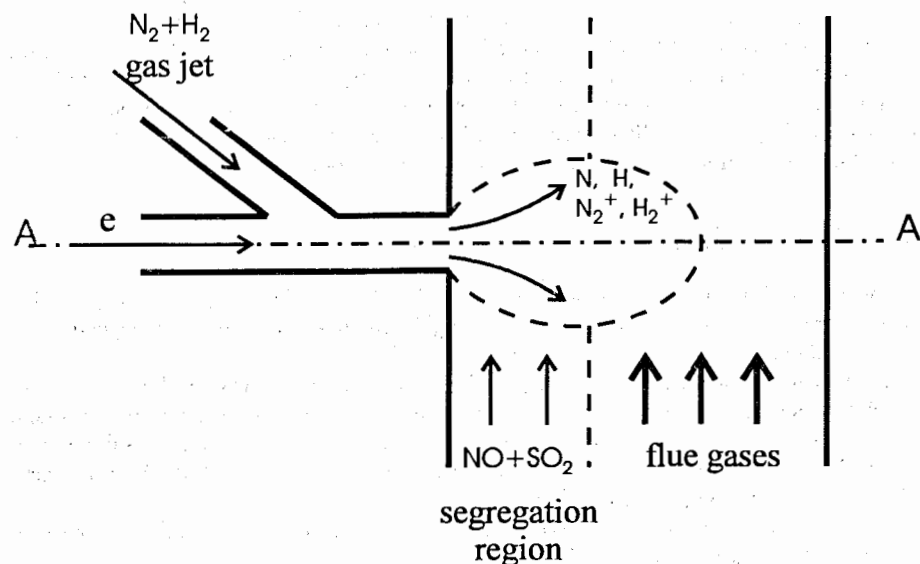


Fig.1 A scheme illustrating the reagent and e-beam feeding in our approach. A partial segregation of NO+SO<sub>2</sub> admixtures from the rest of flue gases is reached in the UV-pretreatment – see the text (not shown on the figure).

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