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ENHANCEMENT AND CONTROL OF BURNING PROCESSES AND ATMOSPHERE POLLUTANTS REMOVAL FROM BURNING INSTALLATIONS IN THE ELECTRIC FIELDS

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1. Enhancement and control of burning process in an electric field

1.1. Introduction

Although an interaction of the external electric field with the flame was known as early as XVII century [1], theories accounting for this phenomenon were developed only recently [2-6].

Present investigation of the burning process is aimed at the increase of efficiency, that is decreasing of the specific fuel consumption, increasing of the flame temperature, significant reducing of CO content in the exhaust gases. Owing to these technical and economical, and ecological advantages this problem attracts attention of investigators and developers by its actuality for Moldova and Romania.

Our searches for a principal technical solution resulted in choosing of the transversal scheme of the alternative electric field application. This scheme was taken as the basis at designing and manufacturing of experimental apparatus.

1.2. Experimental

The experimental apparatus consisted of a Seitan 20 gas burner with a fuel flow rate 20 m³N/h, a high voltage source of alternative current, and a combustion chamber with embedded Kanthal electrodes connected to the source of a high voltage.

The combustion chamber was a rectangular container with dimensions $50 \times 50 \times 180 \text{ mm}^3$ made of two fireclay fireproof bricks with dimensions $260 \times 130 \times 60 \text{ mm}^3$ forming sidewalls, and four fireclay insertions with width of 50 mm and thickness of 40 mm forming the other walls of the chamber. There was a window in every sidewall of 50×50 mm size for input of probe, measuring the current density in various areas of flame and for finding the areas with the maximum electric conductivity of the flame.

A system of electrodes consisted of two metal plates made of Kanthal with dimensions $180 \times 130 \times 3 \text{ mm}^3$ was mounted flush with the internal sidewalls of the chamber by means of two Kanthal rods of 5 mm diameter passing through the centers of fireclay bricks sidewalls.

The experimental apparatus was mounted on a specialized testing bench of Bucharest Metallurgic Research Institute, ROMANIA which consisted of a working cylindrical chamber with a concrete fireproof lining with front and side windows to allow for visual observation of the flame; set-up for removing and recuperation of heat of exhaust gases; set-up for gas fuel supply at pressure of 5000 mm H_2O , gas flow rate measuring and fine adjusting.

1.3. Technique of measurements

A controllable difference of potentials in the range of 0 - 12 kV was applied to an interelectrode gap with the width of 50 mm. At voltages 0, 3, 5, 7, 9, and 12 kV and fuel flow rates of 7, 12, 16, 18, 20, and 30 m³N/h were measured the pressure of fuel gases, temperature and composition of exhaust gases, transfer current density in the flame.

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The main indexes of achieving of the maximum efficiency of burning are the content of CO in the exhaust gases and the flame temperature.

The following equipment was used for measurements: Fluke digital multimeter with the range of currents 0 – 10 A, grade of fit 1 μ A/1.2% (for measuring of current in the primary circuit of the high voltage source); voltmeter with the range 0 – 500 V, grade of fit 1.5 (for measuring of voltage in the primary circuit of the high voltage source); microamperemeter with the measuring probe with the range 0 – 200 μ A, grade of fit 1.5 (for measuring of the current transferred by the ions in the flame and finding the regions with the maximum electric conductivity of the flame); cathode oscilloscope with the ranges 0 – 20 kV, 0 – 20 A, 0 – 60 Hz, grade of fit 0.5 (for measuring of voltage in the secondary circuit of the high voltage source and observance of the shape of high voltage tension); thermocouple Pt-Rd 13, 0 – 1800°C, ± 6.5°C (for measuring of the flame temperature); Kent type diaphragm, 0 – 25 m³N/h, ± 2% (for measuring of gas fuel flow rate); *U* – shape manometer, 0 – 1000 mm H₂O, ± 0.5 mm H₂O (for measuring of gas fuel static pressure); TESTO-350 gas analyzer with the electrochemical cells, 0 – 20000 ppm and ± 20 ppm (CO), 0 – 21% and 0.2% (O₂), 0 – 20000 ppm and ± 20 ppm (NO_x) (for measuring of content of CO, O₂, and NO_x in the exhaust gases); INFRALYT 2020 infrared gas analyzer, 0 – 20%, 0.2% (for measuring of CO₂ content in the exhaust gases).

1.4. Experimental results and analysis

The experimental results obtained at variation of the flow rate of fuel gases both in the absence of the electric field and at various voltages are shown in the Figs. 1 - 6.



Fig.1. Pressure-flow rate characteristic of the fuel

Fig. 2. Current-voltage characteristic of the system of electrodes

Analysis of these data shows that electric field doesn't influence significantly on the pressure-flow rate characteristic of the fuel (Fig. 1). The maximum value of the current in the flame at the upper value of high voltage (12 kV) is 32 μ A. Current-voltage characteristic is in the area of Ohm low applicability (Fig. 2). In the absence of the external electric field an existence of intrinsic flame current of the order of 0.4 μ A was revealed. In the absence of the electric field the CO concentration in the exhaust gases exceeds 1.2% (12000 ppm). It means a low burning efficiency and exceeding of the maximum permissible concentration which is regulated by the ecological standard in force (170 mg/m³N). At the voltages on the electrodes exceeding 7 kV and fuel flow rates 15 – 25 m³ N/h, the CO content in the exhaust gases for this burner CO content is equal to 16 ppm or 136.6 mg/m³N. At the flow rates less than 15 m³N/h and voltages in the investigated ranges it was impossible to decrease the CO content till the permissible level that can be accounted for the design of the burner. The air necessary for burning is injected by the fuel gases that makes it impossible to maintain an optimum value of excees air ratio at the low flow rates (Fig. 6).

An increase in voltage on the high voltage electrodes increases the flame temperature that can



Fig. 3. Variation of CO content in the exhaust gases in dependence of the voltage on the electrodes and of fuel flow rate. 1 - 0 kV; 2 - 3 kV; 3 - 5 kV; 4 - 7 kV; 5 - 9 kV; 6 - 12 kV

150

100

50

0 \ 4

NO_x content in the exhaust gases, ppm





Fig. 5. Variation of NO_X content in the exhaust gases in dependence of the voltage on the electrodes and of fuel flow rate. 1 - 0 kV; 2 - 3 kV; 3 - 5 kV; 4 - 7 kV; 5 - 9 kV; 6 - 12 kV

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Fig. 6. Variation of excess air ratio in dependence of the voltage on the electrodes and of fuel flow rate. 1 - 0 kV; 2 - 3 kV; 3 - 5 kV; 4 - 7 kV; 5 - 9 kV; 6 - 12 kV

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1.5. Economical and ecological efficiency of electric field application at the gas fuel burning

A calculation of economical efficiency was conducted by determining of fuel economy at the gas fuel burning due to reducing of CO concentration in the exhaust gases in comparison with the case when the

be accounted for a shift of heat-chemical reaction in the area of primary formation of CO₂ molecules instead of CO ones (Fig. 4).

As soon as this type of burner doesn't allow for obtaining of high temperatures at which NO_x is formed, the influence of the electric field is in the range of accuracy for these experiments and NO_x formation depends mainly on gas fuel flow rate (Fig. 5). In the range of the flame temperatures under consideration NOx content doesn't exceed the maximum permissible value (350 ppm or $450 \text{ mg/m}^3\text{N}$).



Fig. 4. Variation of the flame temperature in dependence of the voltage on the electrodes and of fuel flow rate. $1 - 0 \, kV; \, 2 - 3 \, kV; \, 3-5 \, kV;$ 4 - 7 kV; 5 - 9 kV; 6 - 12 kV

electric field is absent. For the burner under consideration of Seitan 20 type this economy is 5% or 1 m³N/h. Taking into consideration the electric energy expenses due to electric field presence (0.2 kWh/h USD) and cost of gas fuel (0.080 USD/m³N), and assuming that annual duration of work is 4000 h/year we find that annual saving is 292 USD/year per a burner. Capital investments for equipping of a burner with means of electric field creation are 30 USD. In this way, the amortization term is 0.1 year.

As soon as this type of gas burners has a disadvantage of formation of large quantities of CO in the exhaust gases their equipment with the means of electric field creation turns the gas burners into more ecological friendly ones.

2. Atmosphere pollutants removal from burning installations in an electric fields

2.1. Introduction

The technologies of NO_x and SO_2 simultaneous removal from exhaust gases are based in the following conceptual scheme: the exhaust gases get energy necessary for ionization and formation of radicals of O, OH, HO₂, which oxidize molecules of NO_x and SO_2 , being further transformed in corresponding acids – sulphuric through the reaction of hydrolyze and neutralization by means of introduction of ammonia in gas, and getting of sulphate and nitrate of ammonia in the fine particulate form, which are seized by electro-filter. These technologies are distinguished by the mode, in which the exhaust gases receive the energy necessary for ionization.

The old technologies, in which the exhaust gases were ionized by beams of accelerated electrons [7-9], were replaced by the new based on the treatment of the exhaust gases with "cold plasma" induced in the corona discharge system with electrodes [6, 10 - 16], in the microwaves system without electrodes [17, 18] or in combined fields of corona discharge and microwaves [19].

In the case of electrical discharges, energy for ionization of the exhaust gases is getting due to application of high voltage (negative direct, pulsing or alternating positive) to the system of electrodes, which consist of group of filament electrodes, placed in the center of a gas channel formed from two parallel metallic plates connected to the ground [6, 10 - 16]. If the electric field intensity is high enough that accelerated electrons could ionize the exhaust gases molecules, the presence of the free electrons in this region is sufficient for developing of corona discharge and formation of the other electrons, positive and negative ions, active radicals (excited atoms and molecules) required for the process.

In these conditions, the corona discharge takes place in the exhaust gases running from the filament electrodes to the ground electrodes in time by 20 - 40 ns. The corona discharge will be completed if the total duration of the current impulse is about 1 μ s. The positive corona discharge obtained through the application of short impulses of high voltage – large peak voltage at the high impulse rise rate (by order of nanoseconds), conducts to the biggest production of ions, free electrons and active species. The energy transfer from the active electrode to gas must be on the level of 1 J/impulse per m of electrode. In comparison with the technology based on the treatment of the exhaust gases with beams of accelerated electrons, the corona discharge, especially at the application of high voltages in very short impulses, is less effective in the nitrogen and sulphur oxides trapping, however, the cost of technology realization is considerably lower. And another advantage of this technology is not required the special protective measures.

Plasma induced by microwaves [17, 18] produces secondary electrons of very high concentrations, and that is extremely important at once initiated in microwave field, plasma kept up by gas pressure more high (to double atmospheric pressure) than kindling pressure. Acting through the all cross-section of the gas flux, the microwaves are introduced when the electric field intensity is higher than 300 V/cm, the duration of the impulses is $10^{-8} - 10^{-7}$ s and the frequency of the impulses is 200 - 1000 MHz. Cost of 1 kW of microwave power is 10 - 15 times less than of 1 kW of accelerated electrons.

The technology of the simultaneous trapping of the sulphur and nitrogen oxides from the exhaust gases through combined using of induced plasma by corona discharge and induced plasma by microwaves [19] is developed currently by S.C. ICPET S.A. Bucharest (Bucharest Institute for Research and Design of Thermoenergetical Equipments) in collaboration with INFLPR Bucharest (Bucharest National Institute for Research and Development in Laser, Plasma and Radiation Physics), unifies the advantages of both methods of gas treatment. The degree of trapping of the sulphur oxide is estimated on maximum level 95%, but for the nitrogen oxide this index is about 80%. The specific energy required for process is 8 - 20 Wh/Nm³.

The factors influencing the efficiency of trapping of the sulphur and nitrogen oxides are the next: the concentration of the water in the exhaust gases, the required amount is 8 - 30%; the amount of ammonia determined from stoichiometric calculation in function of the initial concentration of SO₂ and NO_x; the time when the ammonia is introduced (during exhaust gases irradiation or after, although with less efficiency but with the inhibition of the reaction of N₂O forming as advantage); the optimal temperature interval of the ex-

haust gases is $60 - 90^{\circ}$ C; the existing technologic additives or special additives; the initial concentrations of SO₂ and NO_x etc.

2.2. Mecanism of chemical transformations of nitrogen and sulphur oxides in the field of corona discharge

The principal chemical reactions leading to oxide of nitrogen and sulphur transformations in corresponding acids can be grouped in the next scheme [13]: the primary formation of the radicals $(O\bullet, OH\bullet, HO_2 \bullet)$ and other substances / species activated through the collision of the molecules with electrons of 5 to 20 eV energy:

$$e^{*}(5.1 \text{ eV}) + \text{H}_{2}\text{O} \rightarrow \text{H} + \text{OH}^{-};$$

$$e^{*}(7 \text{ eV}) + \text{O}_{2} \rightarrow \text{O} + \text{O}^{-}e^{*}(>15.7 \text{ eV}) + \text{N}_{2} \rightarrow \text{N}_{2}^{+}; e; \text{N}^{+}; \text{N}; \text{N}_{2}^{*};$$

$$e^{*}(>12.1 \text{ eV}) + \text{O}_{2} \rightarrow \text{O}_{2}^{+}; e; \text{O}^{+}; \text{O}; \text{O}_{2}^{*};$$

$$e^{*}(<5 \text{ eV}) + \text{O}_{2} \rightarrow \text{O}_{2}^{-};$$
(1)

the secondary forming of new radicals $(O\bullet, OH\bullet, HO_2 \bullet)$ through the effect of the neutralization and transfer of charge in the result of the collision of the molecules, ions and electrons:

$$O_{2}^{+} + O_{2}^{-} \rightarrow O_{3} + O \bullet;$$

$$O_{2}^{+} + H_{2}O \rightarrow O_{2}^{+}H_{2}O;$$

$$O_{2}^{+}H_{2}O + HO_{2} \rightarrow H_{3}O^{+}OH + O_{2};$$

$$H_{3}O^{+}OH + n \cdot H_{2}O \rightarrow H_{3}O^{+}(H_{2}O)_{n} + OH \bullet;$$

$$H_{3}O^{+}(H_{2}O)_{n} + O_{2}^{-} \rightarrow (n+1)H_{2}O + HO_{2}\bullet;$$
(2)

the transformation of NO_x and SO_x in corresponding acids:

$$NO + HO_{2} \bullet \rightarrow NO_{2} + OH \bullet;$$

$$NO + O \bullet \rightarrow NO_{2};$$

$$NO_{2} + OH \bullet \rightarrow HNO_{3};$$

$$NO_{2} + O \bullet \rightarrow NO_{3};$$

$$NO_{2} + NO_{3} \rightarrow N_{2}O_{5};$$

$$N_{2}O_{5} + H_{2}O \rightarrow 2 \cdot HNO_{3};$$

$$SO_{2} + OH \bullet \rightarrow HSO_{3};$$

$$HSO_{3} + OH \bullet \rightarrow H_{2}SO_{4};$$

$$SO_{2} + O \bullet \rightarrow SO_{3};$$

$$SO_{3} + H_{2}O \rightarrow H_{2}SO_{4};$$

$$SO_{3} + H_{2}O \rightarrow H_{2}SO_{4};$$

$$SO_{3} + H_{2}O \rightarrow H_{2}SO_{4};$$

the neutralization of the formed acids by means of base compounds (through the injection of ammonia NH₃):

$$HNO_{3} + NH_{3} \rightarrow NH_{4}NO_{3}$$

$$H_{2}SO_{4} + 2 \cdot NH_{3} \rightarrow (NH_{4})_{2}SO_{4}$$
(4)

2. 3. The experimental research on the realization of technology of SO_2 and NO_X removal by pulsed corona discharge

The italian firm ENEL produced the installation for experimental research of NO_x -SO_x removal with using of three reactors (Fig.7) [14]. Two of the last ones (TR 1000/1 and TR 1000/2) were of plates type and another was of tubular type. The constructive characteristics of all three reactors are presented in Table 1. The investigation was carried out at the next experimental conditions: the temperature of the exhaust gases at the entrance was in the range $80 - 100^{\circ}$ C; the gas yield $- 500 - 600 \text{ m}^3/\text{h}$; the mean gases composition:

 $N_2 - 73\%$, $O_2 - 8\%$, $H_2O - 6\%$, $NO_x = 500 - 550$ ppm, $SO_2 = 350 - 400$ ppm; the amount of injected ammonia corresponding to molar ratio $NH_3/(NO_x+SO_2) = 0.7 - 0.8$.



Fig.7. Scheme of the installation for $NO_x - SO_x$ removal in electric field

For the supply of the electrodes it was using the high voltage generator with value of voltage in pulsed regime about 80 kV that corresponds to current density through the ground electrode on the level of 30 A/m^2 . The duration of the impulse was from 1 to 2 µs with rise of impulse 20 ns. The frequency of the impulses repetition was up to 300 impulses/s.

It was established that geometry of reactor plays important role in the process of energy transfer to the exhaust gases. For the reactor TR 1000/2 the saturation threshold of the NO_x removal process is higher than for TR 1000/1 due to larger volume. The similar efficiency in NO_x removal can be achieved in the reactor with smaller volume (TR 1000/3) but complete active for processing.

The sulphur oxides can be reduced in even more great scale than the nitrogen oxides due to thermochemical reaction with ammonia even in the absence of the energization.

The energization influences the process leading to the reaction rate growth and increasing of amount of the particles of ammonium sulphate. The efficiency of the sulphur oxides removal is about 75% at the temperature of the exhaust gases 100° C and about 90% at the temperature 70° C. At the absence of the ammonia the efficiency is varied from 12 to 20% on the dependence on the energy transferred to the exhaust gases. The efficiency of the nitrogen oxide removal is about 50% at the energy consuming on the level 12 - 15 Wh/Nm³. It will depend on the initial concentration of NO_x in the exhaust gases being higher at the low initial concentrations.

Reactor type	TR1000/1	TR1000/2	TR1000/3
Reactor geometry	with plates	with plates	tubular
Number of channels	3	2	16
Gas velocity, m/s	0.36	0.27	0.43
Time of passage, s	3.9	5.6	3.5
Channel longitude, m	1.4	1.5	1.5
Channel wide, mm	200	200	200
Total volume, m ³	0.84	1.2	0.75
Total surface of plates, m ²	8.4	12	15.1
Number of emitting electrodes	14	20	16
Distance between filament electrodes, mm	200	150	200
Longitude of filament electrodes, m	1.5	2	1.5
Diameter filament electrodes, mm	3	3	3

Table 1. Constructive characteristics of reactor

$2.\,4.\,SO_2$ and NO_x removal by combined using of pulsed corona discharge and alternating electrical field

The efficiency of NO_x and SO_2 removal can be increased by means of the application of an alternating electric field to ionized by pulsed corona discharge exhaust gases [16]. The effective intensity of alternating field is in the range 5 – 30 kV/cm and the frequency (Hz) is determined by the condition:

$$f = (1.10 - 1.25) \frac{k_{\max} \cdot \overline{E}}{l},\tag{5}$$

were k_{max} is the maximum magnitude of the mobility of the ions, participating in the active radicals formation in accordance with the reactions (2), $[\text{m}^2/(\text{V}s)]$; \overline{E} – the effective intensity of the alternating electric field, [V/m]; l – the distance between electrodes, [m].

The ionized particles are involved in the oscillating movement in the alternating field that increases the probability of the collisions between them and the neutral molecules, intensifying the reactions of NO_x and SO_2 conversion.

The tests carried out with using of new process [6, 16] have confirmed the possibility of the efficiency improvement at the nitrogen oxides removal up to 90% and the sulphur oxides removal up to 95 – 98% (Tables 2, 3).

Table 2. Influence of alternating electric field effective intensity \overline{E} on the efficiency of NO_x and SO₂ removal from exhaust gases

\overline{E} , kV/cm	0	4	5	10	20	30	35
<i>f</i> , kHz	0	0.79	1.0	2.0	4.0	6.0	7.0
NO _x removal, %	50	52	59	81	86	90	Electrical break-down
SO _x removal, %	75	76	80	90	93	95	in exhaust gases

As follows from the Tables 2 and 3, the efficiency of NO_x and SO_x removal from the exhaust gases is higher in the case of the presence of alternating electric field with the intensity in the range 5 - 30 kV/cm than in the case of DeNO_x – DeSO_x technology (Table 2, column $\overline{E} = 0$, f = 0), but the variation of the field frequency, regarding reference value of f = 4 kHz, determined from Eq.(5), leads to the diminution of the efficiency of the removal process. For the frequency f < 4 kHz this decrease is explained by the fact that a part of the ions are neutralized on the wall of the tube connected to ground and does not participate in the reactions (2) and (3). In the case, when f > 4 kHz, the oscillating movement of the ions in the alternating electric field takes place in restricted space that decreases the probability of the collisions between ions and between ions and the neutral molecules and as a consequence, the efficiency of the nitrogen and sulphur oxides removal.

Table 3. Influence of the frequency of alternating electric field on the efficiency of NO_x and SO_2 removal from exhaust gases

\overline{E} , kV/cm	2.0	3.0	3.6	4.0	6.0	8.0
<i>f</i> , kHz	20	20	20	20	20	20
NO _x removal, %	23	35	48	86	64	57
SO _x removal, %	31	56	70	93	86	81

The technologies of the nitrogen and sulphur oxides removal by electric field have suchadvantages as reduced consumption of ammonia, small losses of pressure through installation and obtaining of some products which can be possibly used as fertilizers [4].

3. Conclusions

1. An usual gas burner of Seitan 20 type was equipped with the means of transversal electric field creation which was applied to the flame that resulted in the flame temperature increase from 1100 up to 1190°C due to decrease of CO percentage from 1.2 up to 0.012%. This resulted in reduction of the fuel consumption about 5% in comparison with the nominal case and creation of ecological friendly gas burner. Economical efficiency of a proposed gas burner application is 292 USD/year at the amortization of the capital investments within 1.2 month.

2. The old technologies in which the exhaust gases were treated by the beam of accelerated electrons, were displaced by the technologies based on the treatment of the exhaust gases with plasma induced in the corona discharge (for example, $DeNO_x - DeSO_x$ technologies elaborated by ENEX, Italy), by means of microwaves field or through the combination of both technologies.

3. In alternating field the ionized by pulsed corona discharge particles are involved in oscillating movement that increases the probability of the collisions between the negative and the positive ions and between them and the neutral molecules, intensifying the reactions of NO_x and SO_2 removal.

4. The obtained experimental results have shown the possibility of the efficiency improvement of the nitrogen oxides removal from 50 to 90% and of the sulphur oxides removal from 75 to 95 - 98%.

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Summary

An apparatus, consisting of a Seitan 20 gas burner with a fuel flow rate 20 m³N/h, supplied with a system of Kanthal electrodes and an adjustable source of a high voltage, was investigated on the experimental bench of Bucharest Metallurgic Research Institute, ROMANIA. It was found that an increase of voltage up to 12 kV, and current up to 32 μ A, respectively, results in the evident improvement of the burning process: increasing of flame temperature from 1100 up to 1190°C, decreasing of CO percentage from 1.2 up to 0.012%, reducing of the fuel consumption about 5%. Annual savings resulted from the using of such apparatus is around 292 USD. A new technology of nitrogen and sulphur oxides removal in the installations for burning of organic fuels, which is based on the oxidation of the last ones up to the corresponding acids used pulsed corona discharge and alternating electric field, neutralization of the formed acids by introduction of ammonia in gas and receiving of nitrate and sulphate of ammonia used as fertilizers, is proposed.