Investigation of the effect of condensed phase on the energy conditions for the initiation of the plasma-chemical process of flue gas cleaning by a pulsed electron beam

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Abstract. This work investigates the processes of dissipation of the charge and energy of a pulsed electron beam in gas compositions (nitrogen, carbon dioxide and oxygen) in the presence of ammonium sulphate and nitrate. A pulsed electron beam generated by the TEA-500 accelerator (Tomsk, Russia) with an electron energy of up to 410 keV, a beam current of up to 5 kA (\(I_0\)), and a half-amplitude voltage pulse duration of 60 ns was injected into a 46 cm long drift chamber filled with a gas mixture. The pulsed electron beam current (\(I_{Be}\)) passing through the drift chamber was registered using a sectioned calorimeter with beam charge monitor function, and the efficiency of the current passage of the beam was determined as the ratio \(q_{Be}/q_0\), where \(q_0\) is the beam charge measured at the place of its injection into the chamber drif. The pressure in the drift chamber varied (375, 560 and 760 Torr, humidity value 15% ± 5% and 50% ± 5%). The geometric dimensions of the plasma-chemical reactor for initiating plasma-chemical reactions of flue gas cleaning were determined.

1. Introduction
The use of pulsed electron beams (PEB) interacting with various gas media is determined by their capabilities in the volumetric transfer of high-density energy to a gas and in triggering many plasma-chemical reactions. The interaction of a PEB with gas media can be considered as ensuring effective volumetric absorption of PEB energy by a gas during several chemical reactions, and as ensuring minimum losses during its transportation in the reactor volume at normalized parameters of the gas medium (composition, pressure, temperature, etc.) [1, 2]. The energy carried by the beam is proportional to the beam current. Simultaneously with an increase in the beam current, the difficulties in using the energy stored in the electron beam also increase. This is due to the collective interaction of the beam particles with the generated plasma that ultimately reduces the charge carried by the beam. In addition, in the interaction of PEB with various gas media, it is necessary to take into account the parallel processes: neutralization of the space charge of the beam, current compensation of the beam, the development of instabilities (longitudinal, transverse, resistive, hose) [3, 4]. Most of the research is aimed at studying the physics of the propagation of an electron beam during injection into low-pressure gases. The number of works on studying the interaction of a pulsed electron beam with gas mixtures of increased pressure is limited [5–7]. However, the process of interaction of a PEB with high-pressure gas mixtures is widely used in various physical experiments and applied developments.
(research on controlled thermonuclear fusion [8], energy transport over long distances [9], changes in physicochemical properties of materials [10], excitation of powerful electromagnetic oscillations, pumping of lasers [11], initiation of selective plasma-chemical reactions, generation of pulsed bremsstrahlung X-ray radiation, collective ion acceleration, synthesis of nanocomposites [12], flue gas treatment technologies [13], wastewater treatment and sterilization [14]).

The main problems in the studies of the interaction of a pulsed electron beam with high-pressure gas mixtures are connected with control and prevention of the negative influence of the processes associated with neutralization of the space charge of the beam, current compensation of the beam and the development of instabilities. In addition, this imposes certain requirements not only on the conditions under which the PEB interacts with various gas media: pressure, temperature, and composition of the interaction medium, but also on the design of the drift chamber (wall material, geometric dimensions, and diagnostic elements). A number of plasma-chemical technologies used in metallurgy for the disposal of industrial waste, in the synthesis of hydrocarbons, and in the purification of flue gases requires uniformity of specific absorbed energy (absorbed dose) along the entire length of the drift chamber.

The subject of the presented work was the study of the processes of dissipation of the charge and energy of a pulsed electron beam in gas compositions (nitrogen, carbon dioxide and oxygen) in the presence of ammonium sulfate and nitrate. The obtained experimental data on the effect of the condensed phase (ammonium sulfate and nitrate) on the conditions for initiating the plasma-chemical process of flue gas cleaning by a PEB can be used in the practical determination and selection of modes of flue gas treatment technology using pulsed electron accelerators.

2. Experimental setup

The experiments were similar to those, which we used before [15]; the parameters of the PEB generated by the TEA-500 accelerator were as follows: the electron energy was up to 410 keV, the electron charge transferred over the membrane of the exit window was 430 µC ($q_0$), half-amplitude voltage pulse duration was 60 ns. A drift chamber was used to investigate the effect of the condensed phase (ammonium sulphate and nitrate) on the conditions for initiating the plasma-chemical process of flue gas cleaning by a pulsed electron beam. Figure 1 shows a 3D model of the laboratory bench.

The drift chamber was a metal tube providing the possibility of effective propagation in the injection region of a pulsed electron beam up to 46 cm. The movement of gas mixtures and particles of the condensed phase in a suspended state was performed using two fans, which were mounted in a
closed loop of the drift tube. The air flow rate was controlled using an anemometer. A sectioned calorimeter was used to record the electron energy in the beam cross section [16]. The design feature of the sectioned calorimeter with the beam charge control function was the simultaneous combination of the calorimeter and the Faraday cup in one construction. The collector sections were thermally insulated. The heating of each section was measured using a non-contact temperature sensor. The sensor system was managed by a controller, which, in turn, transmitted data to a computer for automatic processing. To determine the total current and charge of the electron beam, each section of the collector was electrically connected to a low-inductance shunt of the Faraday cup. The electron beam charge from the Faraday cup was measured using an oscilloscope. The electron beam energy was calculated using the developed software [17]. The energy distribution of the electron beam in the cross section was recorded using a sectioned calorimeter [16]. The error in measuring the total energy of the electron beam with a sectioned calorimeter did not exceed ±8%, and the error in measuring the total charge of the beam was not higher than 5% for the Faraday cup.

A mixture of gases was chosen as the test medium: nitrogen, carbon dioxide, and oxygen. The choice of the test medium is determined by the fact that these types of gases are components of flue gases. In experimental studies, the nitrogen, carbon dioxide, and oxygen content was 83%, 14.4% and 2.6%, respectively. The amount of water vapor was estimated by measuring in the drift chamber using a multifunctional meter. Ammonium sulphate and nitrate (particle size 0.05–0.25 µm) were chosen as the condensed phase. Having calculated the material balance of reactions which occurred under the action of a pulsed electron beam onto a mixture of gases (nitrogen, carbon dioxide, and oxygen), the load mass was determined: 3.1 g of (NH$_4$)$_2$SO$_4$, and 0.3 g of NH$_4$NO$_3$ based on the material balance of the cleaning process at initial components: SO$_2$ – 3000 ppm, NO – 500 ppm, NO$_2$ – 500 ppm and purification efficiency: SO$_2$ – 86%, NO – 46%, NO$_2$ – 80%. Also, based on the results of preliminary studies of plasma-chemical processes for cleaning flue gases by a pulsed electron beam, the following values were chosen: 15% ± 5% and 50% ± 5% with the pressure in the drift chamber of 760 Torr, 560 Torr and 375 Torr.

The experimental was as follows: a sectioned calorimeter with a beam charge control function was mounted inside a movable tube at distances of 6, 11, 16, 21, 26, 31, 36, 41, and 46 cm from the exit window of the accelerator. The drift chamber was filled with a gas mixture (nitrogen, carbon dioxide, and oxygen), into which an electron beam was injected. The magnitude of the electron beam was recorded using a Faraday cup, while the energy distribution over the beam cross section was measured using a sectioned calorimeter [16].

3. Results and discussion

Figure 2 shows the characteristic thermal profile of the electron beam imprint obtained using a sectioned calorimeter with the total beam charge monitor function. The initial temperature was 21°C. Section numbering is presented in figure 2 with top numbers in a cell.

The experiments are the same as for earlier experimental studies. When a pulsed electron beam propagates in a gas mixture N$_2$+CO$_2$+O$_2$ at pressure of 760, 560 and 375 Torr in the drift chamber in the presence of ammonium sulfate and nitrate, the value of the pulsed electron beam current that reaches the collector of the Faraday cup ($I_{FC}$) at distances of 6, 11, 16, 21, 26, 31, 36, 41 and 46 cm decreases evenly within the experimental error.

Figure 3 shows the energy diagrams received by each section of the sectioned collector for 10 irradiation pulses. The order of the section numbers on the abscissa axis was selected based on the specified numbering (figure 2).

Analyzing the results of the thermal profiles of a pulsed electron beam recorded at different distances from the anode foil when the electron beam passes through a gas mixture simulating flue gases in the presence of ammonium sulphate and nitrate, we can conclude that the energy distribution over the beam cross section is axis-centered, and the density energy decreases with distance from the geometric centre.
The resulting diagrams of the dependence of the ratio \((k)\) of the charge of the pulsed electron beam that reached the collector of the Faraday cup \((q_{FC})\) to the charge of the pulsed electron beam introduced into the drift chamber \((q_0)\), and the dependence of the average energy density transferred by the pulsed electron beam on the distance \((L)\) at different pressures (figure 4).

**Figure 2.** A thermal profile of the electron beam after 10 successive pulses.

**Figure 3.** Electron beam energy recorded by collector sections at the specified pressures in the drift chamber (ammonium sulfate and nitrate used as a condensed phase).

Figure 4 shows that at a pressure in the drift tube of 375 Torr, the ratio of the charge of the pulsed electron beam \((q_{FC})\) that reached the collector of the Faraday cup to the charge of the pulsed electron beam \((q_0)\) introduced into the drift chamber in the presence of ammonium sulphate and nitrate was higher at a humidity of 50% ± 5% than at a humidity of 15% ± 5% in the drift tube. Meanwhile, the value of the average energy density of the pulsed electron beam over the cross section of the sectioned calorimeter was almost independent of humidity. The maximum value of the average energy density of a pulsed electron beam over the cross section of the sectioned calorimeter was 0.524 J/cm².
Figure 4. Dependence of the ratio of the charge of a pulsed electron beam ($q_{FC}$) reaching the collector of a Faraday cup on the charge of a pulsed electron beam ($q_0$) introduced into the drift chamber (a), and the dependence of the average energy density of a pulsed electron beam in the cross section of the calorimeter on the distance at different pressures (b) (ammonium sulphate and nitrate used as a condensed phase).

At a pressure in the drift tube of 560 Torr, the ratio of the charge of the pulsed electron beam ($q_{FC}$) that reached the collector of the Faraday cup to the charge of the pulsed electron beam ($q_0$) introduced into the drift chamber and the average energy density of the pulsed electron beam over the cross section of the sectioned calorimeter in the presence of ammonium sulphate and nitrate was higher at a humidity of 50% ± 5% in the drift tube. The maximum value of the average energy density of the pulsed electron beam over the cross section of the sectioned calorimeter was 0.429 J/cm$^2$.

At a pressure of 760 Torr, the values of the average energy density of a pulsed electron beam over the cross section of a sectioned calorimeter in the presence of ammonium sulphate and nitrate did not depend on the humidity in the drift tube. The maximum value of the average energy density of a pulsed electron beam over the cross section of the sectioned calorimeter was 0.408 J/cm$^2$.

4. Conclusion

This paper presents the results of comprehensive studies on the dissipation of the charge and energy of a pulsed electron beam in a mixture of gases (nitrogen, carbon dioxide, and oxygen) in the presence of ammonium sulphate and nitrate. The main regularities revealed during this work include the following. In the presence of ammonium sulphate and nitrate, the maximum value of the electron energy recorded by the calorimeter at a long distance (46 cm) from the exit window of the accelerator was recorded at a total pressure of 375 Torr and a humidity of 50% ± 5%. The energy density of the electron beam decreased with the distance from the geometric center. The presence of suspension of ammonium sulphate and nitrate in the amount of 14 mg/L and 1.3 mg/L, respectively, reduces the efficiency of beam propagation. It has been proved that in a plasma-chemical reactor of 26 cm long, plasma-chemical reactions is initiated throughout the volume to clean flue gases using a pulsed electron accelerator with an electron energy of up to 410 keV, a beam current of up to 5 kA and a half-amplitude voltage pulse duration of 60 ns.

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