

Propagation of a pulsed electron beam in gas compositions of carbon-containing composite nanomaterial synthesis reagents

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Abstract. The paper presents the results of comprehensive studies of the efficiency of the propagation of a pulsed electron beam through a mixture of gases: titanium tetrachloride and hydrogen, titanium tetrachloride and methane, titanium tetrachloride and oxygen. These substances are the initial reagents or products of plasma-chemical reactions of the synthesis process using pulsed electron beams. The energy distribution over the beam cross section is uniform, and the temperature decreases with distance from the geometric center. The efficiency of the process of dissipation of the energy of a pulsed electron beam during its passage through gas compositions has been studied depending on the energy of the beam electrons (350–450 keV). The studies were carried out on a test bench including a TEA-500 pulsed electron accelerator, a drift chamber, and a sectioned cut-off calorimeter with a beam charge control function. The research results are of practical importance and are the main ones in the technological processes of pulsed plasma-chemical synthesis of carbon-containing nanocomposites.

Keywords: pulsed electron beam, carbon-containing composite, energy dissipation, charge dissipation, gas compositions.

1. Introduction

Since 2000, the scientific community has become more and more widely discussed topics on the subject of nanoparticles, methods for their production and applications. Since 2010, there has been a growing interest in the development and study of nanocomposite materials, in which the particle size (at least in one direction) is no more than 100 nm. At present, carbon-containing nanocomposites of various types (carbon nanotubes, carbon fibers) are of great interest both for scientific research and for various practical applications. Using carbon nanoparticles of a certain type in given quantities, due to their specific geometry and characteristics, it is possible to obtain a nanocomposite material with a desired set of performance characteristics. The physicochemical properties of carbon-containing nanomaterials depend on the synthesis method [1, 2].

The main method for the synthesis of carbon-containing nanomaterials is the sol-gel method [3–5]. The synthesis process in the sol-gel method is carried out at high temperatures, and the use of catalysts is also required, which must be removed from the final product at the end of the process.

It seems promising to use the pulsed plasma-chemical method to obtain carbon-containing nanocomposites [6]. Efficient input of energy into the gas by a pulsed electron beam due to the elementary processes in plasma significantly reduces the energy costs of the implementation of the production process. However, the studying the processes occurring during the interaction of pulsed electron beams with objects with a complex chemical composition, which are the main ones in the technological processes of pulsed plasma-chemical synthesis of nanocomposites, remains topical. The purpose of this work was to conduct comprehensive studies of the efficiency of the passage of a pulsed electron beam through a mixture of gases: titanium tetrachloride and hydrogen, titanium tetrachloride and methane, titanium tetrachloride and oxygen.

2. Measurement of absorbed dose

2.1. Experimental setup

During the research, a TEA-500 pulsed electron accelerator was used [7]. This accelerator has the parameters shown in Table 1. A distinctive feature of the design of the TEA-500 pulsed electron accelerator is a matching autotransformer. It provides matching of a low-resistance water double forming line with a high-resistance impedance of an explosive emission planar diode.

Table 1. Operating beam parameters of the TEA-500 electron accelerator

Parameter	Value
Electron energy	350–450 keV
Extracted electron current	up to 10 kA
Voltage pulse duration (FWHM)	60 ns
Pulse repetition rate	up to 10 pps
Pulse energy	up to 200 J

The stability of the accelerator was controlled using a Rogowski coil and a capacitive voltage divider (the spread of current and voltage values recorded by the sensors did not exceed 5%).

The scheme of experiments was similar to that which we used in [8], but within the framework of this work, a cut-off sectioned calorimeter with a beam charge control function was used (Fig.1).

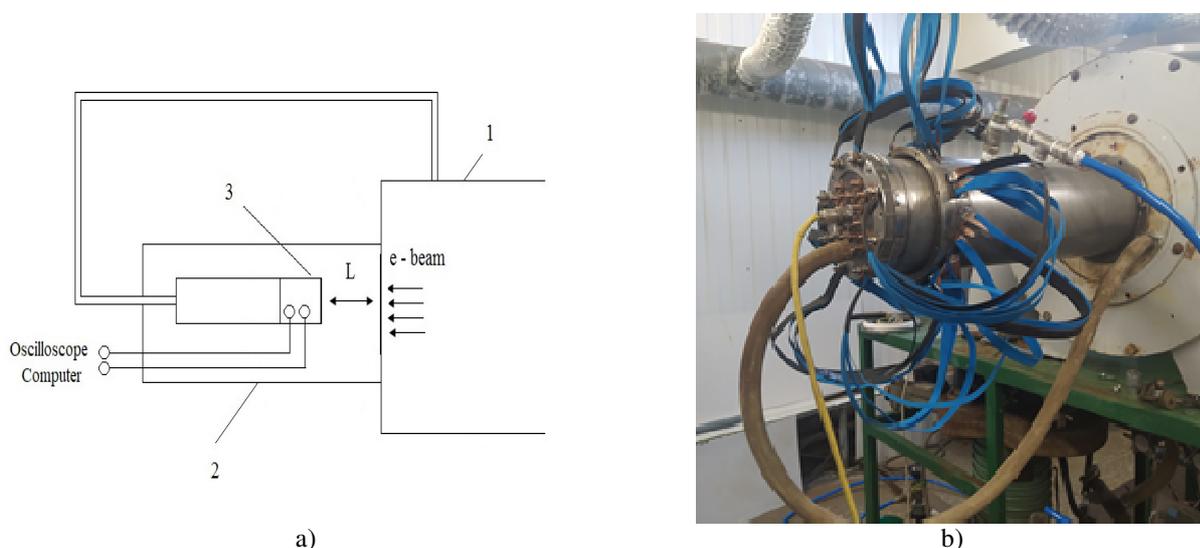


Fig.1. Diagram (a) and photograph (b) of the experimental stand: 1 – diode chamber of the TEA-500 pulsed electron accelerator, 2 – drift chamber, 3 – sectioned calorimeter with the function of controlling the total beam charge.

A sectioned cut-off calorimeter with a beam charge control function was installed inside a mobile tube at distances (L) of 8, 15, 25, 35, and 45 cm from the accelerator output window. In the experiments, the drift chamber was filled with three gas mixtures: 1) titanium tetrachloride (6 mmol) and hydrogen (18 mmol), 2) titanium tetrachloride (6 mmol) and methane (18 mmol), 3) titanium tetrachloride (6 mmol) and oxygen (36 mmol)) into which an electron beam was injected. The charge of the electron beam was recorded using a Faraday cup and the energy distribution over the beam cross section was recorded using a sectioned calorimeter. The current measurements recorded by the reverse current shunts were made by removing the voltage drop across the resistors (0.06Ω). After that, the voltage signal was attenuated by a factor of 25 to protect the oscilloscope from high voltage. In this regard, the sensitivity of reverse current shunts is ~ 150 A in amplitude. The technique makes it possible to record the current generated by the beam electrons and the electrons generated as a result of the ionization of the medium; however, this factor depends on the plasma electron concentration and its lifetime.

To study the efficiency of the energy dissipation process of a pulsed electron beam when passing through gas compositions, experiments were carried out at different parameters of a pulsed electron accelerator. The accelerating voltage varied from 350 to 450 kV, while the total accelerator current varied from 7.5 to 4.2 kA.

3. Results and discussion

3.1. Research on the efficiency of propagation of a pulsed electron beam through gas compositions of reagents for the synthesis of carbon-containing composite nanomaterials

Fig.2 shows the typical thermal profiles of a pulsed electron beam recorded at various distances from the anode foil during the propagation of an electron beam through a mixture of gases.

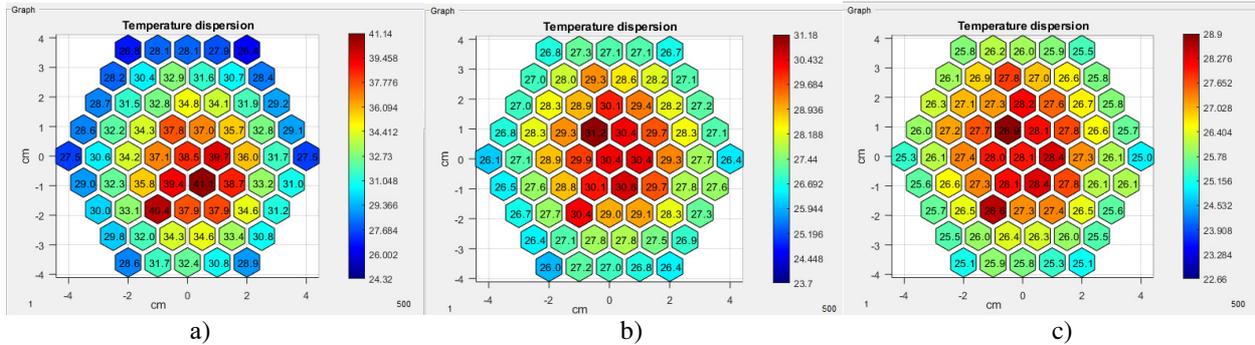


Fig.2. Typical thermal profiles of a pulsed electron beam recorded at distances of 8 cm (a), 25 cm (b) and 45 cm (c) from the anode foil during the passage of an electron beam through a mixture of gases TiCl_4 (6 mmol) + CH_4 (18 mmol).

For these imprints, the initial temperature was (before irradiation) 21°C . The sections were numbered in a spiral from the reference points indicated in the figure clockwise. The effective diameter of the sectioned calorimeter was 114 mm. With increasing distance from the anode foil, the thermal profile of the imprint of the pulsed electron beam had a more uniform distribution in the cross section. Fig.3 shows the dependences of the average energy transferred by the pulsed electron beam and the charge of the pulsed electron beam that reached the collector of the Faraday cup (q) on the distance for gas compositions of titanium tetrachloride, oxygen, hydrogen, and methane.

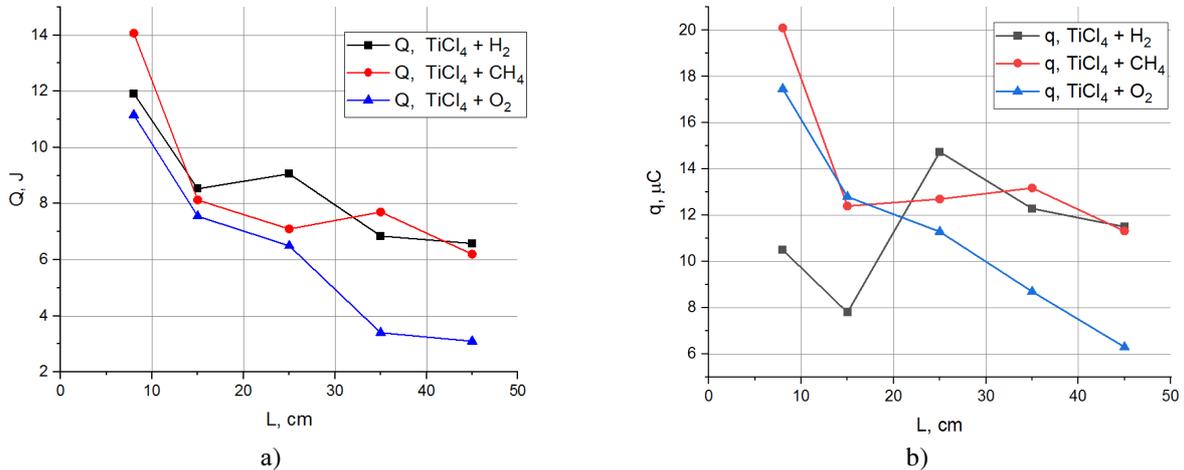


Fig.3. Dependence of the average energy transferred by the pulsed electron beam (a) and the charge of the pulsed electron beam that reached the collector of the Faraday cup on the distance for gas compositions of titanium tetrachloride, oxygen, hydrogen, methane (b).

The energy distribution over the beam cross section was uniform, and the energy value decreased with the distance from the geometric centre. The electron beam propagates more efficiently in the $\text{TiCl}_4 + \text{H}_2$ gas mixture than in other studied mixtures. For the $\text{TiCl}_4 + \text{H}_2$ gas mixture, there is an increase in the energy value at a distance of 25 cm (9.06 J) compared to a distance of 15 cm (8.53 J).

The dependence of the charge recorded by the Faraday cup on the distance for the $\text{TiCl}_4 + \text{H}_2$ gas mixture had a non-linear form. It can be seen that there are points on the diagram where the value of the total charge of the beam sharply decreases, and then increases with increasing distance. The experiments were performed in duplicate and repeated two times with similar results.

The peculiarity of the decrease in the charge of a pulsed electron beam at a distance of 15 cm of the drift space and the increase in the charge value by 25 cm in the mixture can be explained as follows. Firstly, the saturation vapor pressure of titanium tetrachloride is low. Secondly, hydrogen is differed from oxygen and methane by the fact that losses per unit mass thickness are two times higher due to the ratio of the nuclear charge to the mass number. This ratio is 1 for hydrogen and 0.5 for oxygen. Hydrogen is characterized by a low ionization cross section, which affects the value of the ion background integral. In addition, at a low pressure of titanium tetrachloride and a distance of 15 cm from the collector of the Faraday cylinder, the beam scatters due to the fact that the necessary ion background is not formed, due to which the beam would be neutralized. A pattern similar to the behavior of a beam in vacuum is observed (an increase in the distance (up to 35 cm) to which the beam is propagated in the medium causes an increase in the integral of the ion background, due to which the beam charge is neutralized).

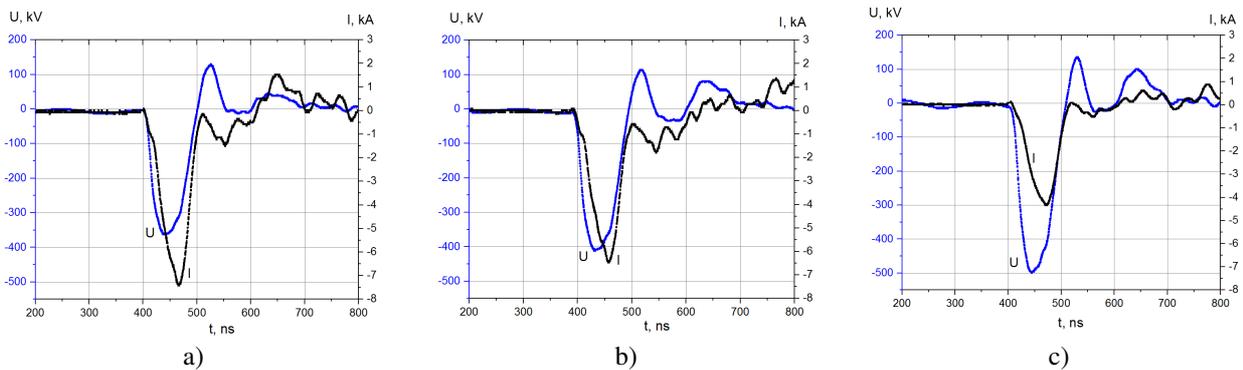


Fig.4. Typical oscillograms of the accelerating voltage and total current for three operating modes of the accelerator: 350 kV (a), 400 kV (b), and 450 kV (c).

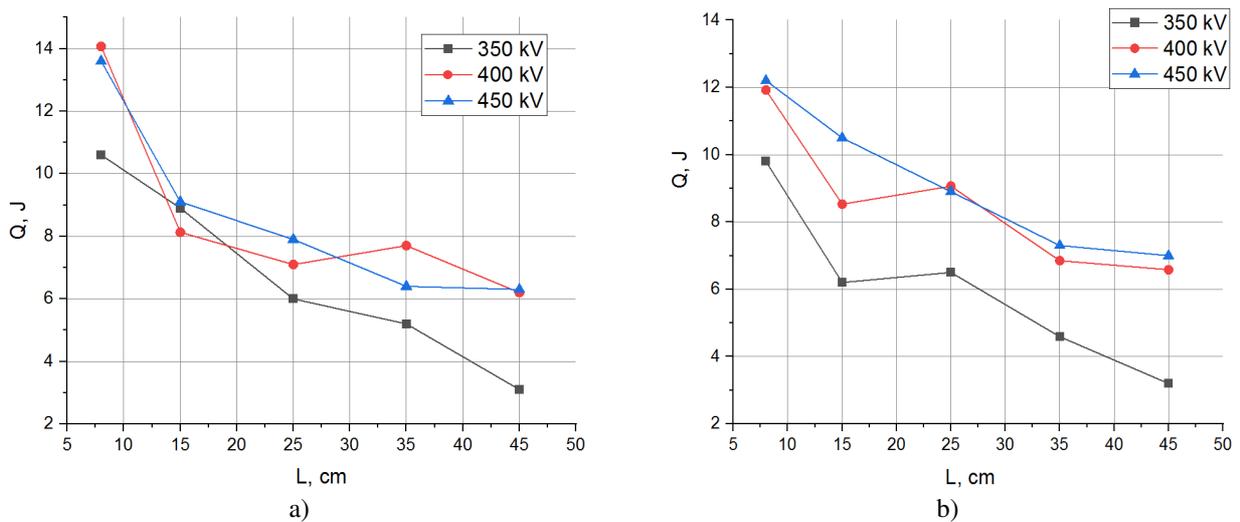


Fig.5. Dependence of the energy of a pulsed electron beam passing through gas compositions on distance at different beam electron energies: $\text{TiCl}_4 + \text{CH}_4$ gas mixture (a) and $\text{TiCl}_4 + \text{H}_2$ gas mixture (b).

3.2. *The results of studies of the efficiency of the energy dissipation process of a pulsed electron beam when passing through gas compositions of titanium tetrachloride, hydrogen, methane, depending on the beam electron energy (350–450 keV)*

Fig.4 shows typical oscillograms for three operating modes of the accelerator. There is the dependence of the energy of a pulsed electron beam passing through gaseous compositions on distances at different energies of beam electrons in Fig.5. Analyzing the obtained data, we can conclude that the change in the energy of the electron beam from 350 to 450 kV had little effect on the process of dissipation of the energy of the pulsed electron beam when passing through the gas compositions of titanium tetrachloride and hydrogen, titanium tetrachloride and methane.

4. Conclusion

This paper presents the results of comprehensive studies of the charge and energy dissipation of a pulsed electron beam in a mixture of gases: $\text{TiCl}_4 + \text{H}_2$, $\text{TiCl}_4 + \text{CH}_4$, $\text{TiCl}_4 + \text{O}_2$. The electron beam is propagated more efficiently in the $\text{TiCl}_4 + \text{H}_2$ gas mixture, however, the dependence of the charge recorded by the Faraday cup on the distance for this gas mixture has a non-linear form. A change in the energy of the electron beam from 350 to 450 kV slightly affects the dissipation of the energy of a pulsed electron beam when passing through gas compositions of titanium tetrachloride and hydrogen, titanium tetrachloride and methane. Revealing regularities in the propagation of pulsed electron beams is important, since the performance of the synthesis technology using pulsed electron beams is determined by the pressure in the reactor and its volume. The main problems in the implementation of studies of the interaction of a pulsed electron beam with high-pressure gas mixtures lie in the area of control and prevention of the negative impact of processes associated with the neutralization of the beam space charge, beam current compensation, and the development of instabilities. And this imposes certain requirements not only on the conditions under which the interaction of pulsed electron beams with various gaseous media occurs: pressure, temperature and composition of the interaction medium, but also on the design of the drift chamber (wall material, geometric dimensions, diagnostic elements). A number of plasma-chemical technologies used in metallurgy for industrial waste disposal, in the synthesis of hydrocarbons, the synthesis of nanocomposites, and flue gas cleaning require uniformity of the specific absorbed energy (absorbed dose) along the entire length of the drift chamber.

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5. References

- [1] Costa E., Zamora P., Zarbin A., *J. Colloid. Interface. Sci.*, **368**, 121, 2012; doi: 10.1016/j.jcis.2011.10.028
- [2] He X., Wu M., Ao Z., Lai B., Zhou Y., An T., Wang S., *J. Hazard. Mater.*, **403**, 124048, 2021; doi: 10.1016/j.jhazmat.2020.124048
- [3] Wang F., Zheng J., Qiu J., Liu S., Chen G., Tong Y., Zhu F., Ouyang G., *ACS Appl. Mater. Interfaces.*, **9**, 1840, 2017; doi: 10.1021/acsami.6b14748
- [4] Hussain M.Z., Yang Z., Linden B.V.D., Huang Z., Jia Q., Cerrato E., Fischer R.A., Kapteijn F., Zhu Y., Xia Y., *J. Energy Chem.*, **57**, 485, 2021; doi: 10.1016/j.jechem.2020.08.048
- [5] Udayakumar S., Ibrahim N., Chien C.Y., Rahman S.A., Mohd Noor A.F., Ramakrishnan S., *Bull. Chem. React. Eng. Catal.*, **15**, 687, 2020; doi: 10.9767/BCREC.15.3.8195.687-697.
- [6] Kholodnaya G., Sazonov R., Ponomarev D., *Fuller. Nanotub. Carbon Nanostructures*, **29**, 7, 487, 2021; doi: 10.1080/1536383X.2020.1820994

- [7] Remnev G.E., Furman E.G., Pushkarev A.I., Karpuzov S. B., Kondrat'ev N. A., Goncharov D.V., *Instrum. Exp. Tech.*, **47**, 394, 2004; doi: 10.1023/B:INET.0000032909.92515.b7
- [8] Kholodnaya G., Egorov I., Sazonov R., Serebrennikov M., Poloskov A., Ponomarev D., Zhirkov I., *Laser Part. Beams*, **38**, 3, 197, 2020; doi: 10.1017/S0263034620000257