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# Hybrid multiplexed pneumoelectrospray system for generation of charged microdroplets

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The article describes the design and presents the characteristics of a hybrid pneumoelectrospray system. It allows to significantly increase the number of charged microdroplets generated per unit time. One of the main purposes of this system is to microdroplet chemical synthesis scale up when operating as part of a microdroplet chemical reactor prototype.

**Keywords:** electrospray, microdroplet chemical synthesis, nebulizer gas, charged microdroplets, transport electrode.

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## Introduction

Electrospray (electrohydrodynamic spraying of liquids) is broadly used in various fields of science and technology. It is used for the production of various micro/nanoparticles [1–3], in space engines [4], microburners [5]. Electrospray of liquids is the basis of a large group of ionization methods used in mass spectrometry for solving various problems in medicine, biology, chemistry and other scientific fields [6–8]. These methods include ESI (electrospray ionization), nanoESI (nanoelectrospray ionization), DESI (desorption electrospray ionization), reactiveDESI (reactive desorption electrospray ionization), ESSI (electrosonic spray ionization), EESI (extractive electrospray ionization). All these methods involve the generation of charged droplets with an uncompensated charge.

It was found in the course of a large number of studies that various chemical reactions accelerate in microdrops compared to their behavior in a liquid homogeneous medium („bulk") [9,10]. It has also been shown that it is possible to obtain substances in microdrops whose synthesis is difficult or impossible in „bulk" [11–13]. All this resulted in the creation of a new method of chemical synthesis — microdroplet chemical synthesis.

The use of microdroplet synthesis for preparative organic synthesis is currently severely limited by its low productivity. The desired productivity begins with values of units of grams per hour [14,15] The productivity of 1 g/h can be achieved with a minimum value of the volume flow rate of the sprayed solution of at least 100 ml/h or 1.7 ml/min (the evaluation was carried out for a conditional second-order reaction occurring with the formation of a product with a molar mass of 200 g/mol; the concentrations of reacting substances are the same and have a value of 0.05 mol/l; 100% yield of the reaction product is assumed). There are currently no specialized systems designed for implementatation of the microdroplet synthesis with practically significant productivity, which is attributable to the novelty of this field and the technical challenges of implementing such systems, as well as, to a large extent, the absence of full-fledged theoretical (or model) concepts that allow for a predictable control of droplet agglomeration. A number of experimental setups are known, in which a number of standard techniques for mechanical droplet generation are used, for increasing the productivity of microdroplet synthesis. In particular, the method of pneumatic spray was used in the setups described in the Ref. [15,16], an ultrasonic method of droplet generation was used in Ref. [17], the number of drops per unit volume was increased compared with electrospray in Ref. [18] using spray nozzles and mesh screens.

It seems that the electrospray of solutions is the most suitable method for the implementation of microdroplet synthesis due to the possibility of generating charged microdroplets. The presence of a charge in all droplets opens up broad possibilities for controlling their characteristics: lifetime, trajectory, charge value. The trajectory of movement can be controlled by the action of electric and/or magnetic fields on charged droplets, which, in turn, can increase or reduce their movement time and change the duration of their evaporation accordingly, which will change their lifetime. Moreover, the presence of a charge in droplets allows for their transportation from the place of origin to the ion source of the mass spectrometer (or before entering into the mass spectrometer) over distances of up to several meters, which makes an online monitoring of the behavior of chemical reactions in them [19,20].

The increase of the productivity of microdroplet synthesis using electrospray is limited by the volume flow rate of the sprayed solution. The problems of its increase are associated with the instability of electrospray, the need to use a drip mode [21], and a significant space charge field. There are two main approaches to increasing the volume flow rate of the sprayed solution during electrospray, which were implemented in practice by various researchers: increasing the number of spray emitters and the use of gas flows. The latter include systems that use a spray gas (Gas-Assisted Elrctrospray) [22,23]. The main idea of their use is to realize the possibility of combining a chromatograph with a mass spectrometer in case of use of which the flow rate of the sprayed liquid ranges from 100 to 1000 $\mu$l/min, and which ensures the operation with highly conductive liquids [24]. The gas supplied along the spray capillary in the direction of movement of charged particles makes it possible to ensure the stability of the electrospray of such flows and reduce the polydispersity of the formed droplets.

Systems with an increased number of emitters are called multicapillary (or multiplexed electrospray). Most of the developed systems can be attributed to three variants: linear construction [1,25], flat arrays of emitters [26] and multi-jet systems [27]. The largest number of schemes proposed in the literature refers to the first two varieties. The first option involves the location of a large number of capillaries working in parallel with each other. Each capillary is connected to two electrodes like in case of a conventional electrospray. The second option is based on creating a large number of holes (emitters) on flat substrates with densities of up to 10,000 sources per square centimeter. It involves the use of three-electrode systems: a spray electrode, an extraction electrode, an counterelectrode or a collector. The sprayed liquid in both approaches is supplied from a common reservoir. Multicapillary systems described in the literature allow obtaining the volume flow rate of the sprayed solution from 10 $\mu$l/min to units ml/min. The flow rate of tens of ml/min is possible only with the implementation of larger multicapillary systems with more than 12,000 sources per square centimeter, which greatly increases the complexity of manufacturing and increases the space charge field of the sprayed aerosol.

Combining electrospray with a spray gas and a large number of emitters can be another approach to increasing the volume flow rate of the sprayed solution. This idea has not yet been fully implemented for practical application. The authors know only one such system, proposed in Ref. [28]. The proposed polycapillary source has several significant drawbacks. Firstly, the extraction electrode is located at a distance of 7 mm from the spray capillaries, which results in a significant loss of ions and charged droplets on it. As the authors showed in their paper, the occurrence of losses is associated with a strong decrease of the velocity of the spraying gas at such a distance. The location of the electrode at the level of the spray capillary allows reducing the loss of charged particles and increasing the efficiency of gas use, as was shown in Ref. [29] for a single capillary. Secondly, it seems incorrect to use the base of the setup from a dielectric. An aerosol of charged droplets and ions can result in strong charging of the dielectric, which, in turn, affects the field at the capillaries, changing the conditions of electrospray. Thirdly, this system has a volume flow rate of the sprayed solution of the order of 1 ml/min, which does not allow for its full usage for many practical tasks of preparative organic synthesis of substances. As shown earlier, the desired values of the volume flow rate of the sprayed solution start from 1.7 ml/min.

The described electrospray systems are not suitable for the implementation of microdroplet synthesis at a practical level. Firstly, this is attributable to the insufficient volume flow rate of the sprayed solution in all the systems described earlier. Secondly, the main task of the proposed systems is the ionization of a sample for mass spectrometry, or the production of droplets with a diameter of less than 10 $\mu$m for various technological applications. The main tasks of ionization sources based on electrospray in the mass spectrometry are obtaining of ions, which are then analyzed in a mass spectrometer, and minimizing the spray solution entering its gas-dynamic interface. This approach is not suitable for microdroplet synthesis, where it is important to minimize the loss of the sprayed sample and collect as many reaction products of interest as possible. Working with small droplets (with a diameter less than 10 $\mu$m) greatly reduces the ability to control their lifetime, as well as other characteristics. Nevertheless, the greatest acceleration of reactions was obtained in paper [9] using a nanoelectrospray.

This paper presents a developed and manufactured hybrid multicapillary pneumoelectric spray system for generating charged microdroplets, devoid of the disadvantages of the described systems and allowing to increase the volume flow rate of the sprayed solution for the implementation of microdroplet chemical synthesis at a practical level. It is one of the main elements of the prototype of a microdroplet chemical reactor designed for the practical implementation of microdroplet chemical synthesis [30]. The multicapillary system was developed on the basis of combining ideas for increasing the volume flow rate of the sprayed solution during electrospray, discussed earlier: gas-assisted electrospray and increase of the number of emitters. The obtained values of the volume flow velocity of the sprayed solution and the loss of charged droplets on the structural elements were experimentally verified.

## 1. The construction of a hybrid multicapillary pneumoelectric spray system

The construction of a hybrid multicapillary pneumoelectric spray system (hereinafter — multicapillary system) is shown in Fig. 1. The system consists of four main

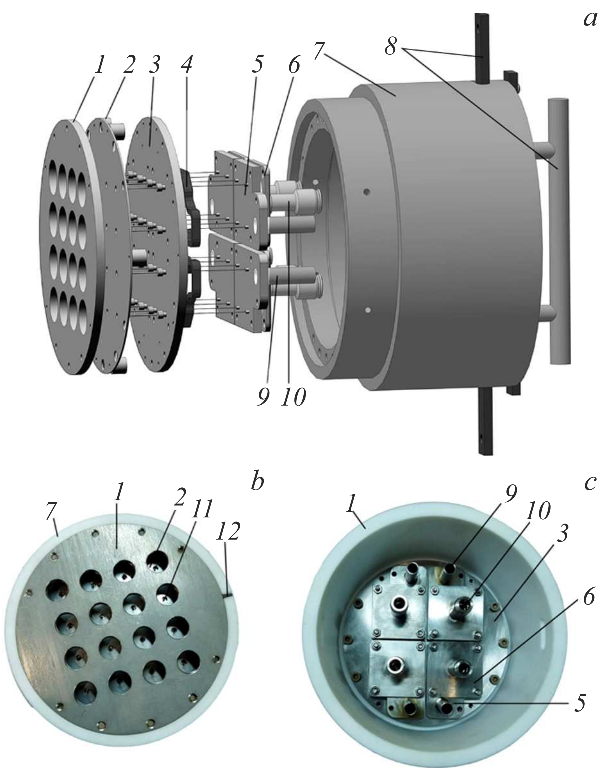

**Figure 1.** The construction of the hybrid multicapillary pneumoelectric spray system, shown on the model (*a*), photos from the front (*b*) and from the back (*c*): *1* — transport electrode, *2* — panel for hiding fasteners, *3* — GIM, *4* — gas seal, *5* — RMIM, *6* — intermediate volume cover, *7* — fluoroplastic casing, *8* — holder assembly, *9* — gas inlet tube, *10* — collet fitting for entering the reaction mixture, *11* —capillary assembly, *12* — the output for connecting the wire.

parts: a transport electrode *1*, gas input module (GIM) *3*, four reaction mixture input modules (RMIM) *5* and a fluoroplastic casing *7*. It has an auxiliary holder *8*, which allows it to be installed vertically. Dimensions of the multicapillary system: diameter — 180 mm, height — 115 mm.

Gas and a sprayed solution were supplied for organizing electrospraying in a multicapillary system, potentials were applied to two electrodes: a spraying and a transport one. The gas was injected through a stainless steel gas inlet tube into four intermediate volumes formed by GIM four gas seals *4* and four RMIM. The liquid was supplied through four collet fittings to enter the reaction mixture *10* into four intermediate volumes, each of which is formed between the RMIM, the lid of the intermediate volume *6* and the liquid seal. Then the gas and liquid separately flowed through *16* spray capillaries, which form *16* capillary assemblies *11*. The structure of the capillary assembly is shown in Fig. 2. It shows its longitudinal section, the name of most positions, corresponding to the parts in Fig.1. The schematic view *A* shows in more detail the liquid capillary *9* and the gas capillary *10*. They contain bases *11* and *12* using which they are installed in the RMIM and GIM, respectively. The outer diameters of the gas capillary and the liquid capillary are 0.8 and 0.4 mm, and the inner diameter are 0.5 and 0.2 mm, respectively. The liquid capillary is extended relative to the gas capillary at a distance of 0.5 mm.

The spray electrode is an assembly of four RMIM, GIM and a panel for hiding fasteners (Fig. 1, 2). There is a special input on the base of the GIM for attaching the wire from the power source. The transport electrode has *16* holes with a diameter of 23 mm, which are located coaxially to the capillary assemblies. The gas capillary is installed at the level of the end face of the transport electrode (Fig. 2). The transport electrode has an input (Fig. 1, *12*) to supply potential to it. The potential difference between these two electrodes allows creating conditions for electrospray.

The concepts used in the developed design of the multicapillary system allow solving the following problems:

1) ensuring the uniformity of liquid and gas supply to all capillary assemblies;

2) ensuring the stability of electrospray;

3) scalability of unit cell-based electrospray.

The first problem was solved by choosing the inlet diameters of the gas and liquid supply elements significantly larger than the diameters of the capillaries. The through diameter of the inlet tubes is 4.8mm for liquid and 7.5 mm for gas. For a liquid, this ensures 1000 times lower hydraulic resistance for the supply tube than for the capillaries included in the RMIM unit, which allows for a uniform outflow of gas and liquid from all capillary assemblies.

The second problem is related to ensuring the stability of the electric field at the end of the capillaries, the influence of the space charge of charged aerosols and the correct choice of geometry for supplying the spraying gas. The transport electrode separates all capillaries from each other, which ensures the independence of the capillary field and allows each capillary to work initially as a single capillary. Calculations of the multicapillary system in Simion-8.1

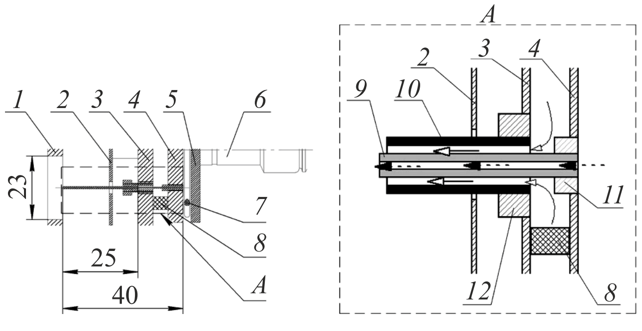

**Figure 2.** Capillary assembly (incision along the axis): view *A* — schematic enlarged image of capillaries (not in scale); *1* — transport electrode, *2* — panel for hiding fasteners, *3* — GIM base, *4* — RMIM base, *5* — intermediate volume cap, *6* — collet fitting for entering the reaction mixture, *7* — liquid seal, *8* — gas seal, *9* —liquid capillary, *10* — gas capillary, *11* — liquid capillary fasteners, *12* — gas capillary fasteners; solid arrow — gas; dotted arrow — liquid, dimensions indicated in millimeters.

showed an inhomogeneous electric field in capillaries of no more than 0.5% for the ideal location of capillary assemblies. The panel for hiding the fastener equalizes the distribution of the electric field at the capillaries and removes the influence of the fastener on it.

It is necessary to observe the geometry according to the expression shown in Ref. [28] for improving the stability of electrospray in the gas stream:

$$D_{orif} \approx (1.2{-}1.4)D_{cap}, \tag{1}$$

where $D_{orif}$ — the inner diameter of the outer capillary, $D_{cap}$ — the outer diameter of the inner capillary. Additionally, the end of the spray capillary should not be extended relative to the end of the gas capillary by more than a distance equal to $D_{orif}$. The liquid injection system is divided into four RMIM units of four capillaries for ensuring accuracy and simplicity of assembly.

## 2. Experimental setup

The volume flow rate of the sprayed solution and the possibility of electrospray were evaluated during the experimental testing of the multicapillary system. The gas was supplied to the system from a QUATTRO ELEMENTI PACIFIC-24 oil-free reciprocating compressor with a power of 0.9 kW, a receiver volume of 24 *l* and a maximum pressure of 0.8 MPa. It flowed through a float flow meter LZM-6T with a valve that allowed to regulate the flow in the system, four ball valves and four polyurethane hoses with a inner diameter of 7.5 mm (185CRT7.5*10B). Each mini-valve allows shutting off four GIM gas capillaries belonging to one RMIM unit.

The solution was supplied into the system using a BT-100M peristaltic pump with an EasyPump VI head with different volume flow rates of the sprayed solution sequentially into each RMIM unit through three collet T-shaped tees. They are connected to four collet fittings for the injection of the reaction mixture using Innovaprene P60 tubes with an inner diameter of 4.8 mm. The multicapillary system is suspended for testing and a collector is installed under it for collecting sprayed charged particles.

An electrical voltage was applied to the electrodes of the multicapillary system *1* and the collector *5* for checking the possibility of electrospray according to the scheme shown in Fig. 3. The voltage was supplied from identical high-voltage blocks Sh0105 provided by LLC „NAUEL" to plus 5 kV *7*. 10 V was supplied on the transport electrode *4* so that it had a known potential. The voltage at the spray electrode consisting of elements *2* and *3*, changed from 1.5 to 4 kV. The total spray current ($I_{\mathrm{sum}}$) and the current supplied to the transport electrode ($I_{\mathrm{te}}$) were measured using multimeters with an internal resistance of 1 MΩ. It was possible to move the collector could be moved to various distances *L* from the multicapillary system, it was grounded, i.e. it was at zero potential.

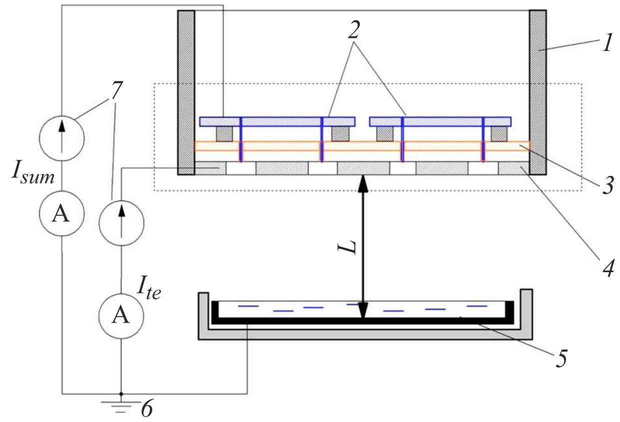

**Figure 3.** Electrical circuit: *1* — multicapillary system, *2* — RMIM, *3* —GIM, *4* —transport electrode, *5* —collector, *6* — ground contact, *7* — high-voltage power supplies +5 kV, *Isum* — multimeter for measuring total current, *Ite* — a multimeter for measuring the current to a transport electrode.

## 3. Results and discussion

At first, the multicapillary system was calibrated according to the volume flow rate of the sprayed solution without electrospray using the main components of sprayed solutions for microdroplet chemical synthesis of — methanol and ethanol. The sample was fed into the spray system according to the following algorithm. First, the spray system was rinsed at the maximum flow rate of the sprayed solution for the peristaltic pump for 20 s, after which the process was interrupted for 20−120 s (the pump is turned off) so that there was no sudden switch to a low liquid flow, i.e. measurements began with a steady flow value. Next, the solution was supplied at different pump speeds: 1, 2, 3, 4 and 5 rpm (revolutions per minute). The solution was collected in a sampling cuvette for a time from 2 to 10 min. The solution was poured from the cuvette into a test tube to measure the volume of the sprayed mixture with an absolute error of ±0.1 ml. The solution was pumped from of the spray system back into the glass by a peristaltic pump after a single measurement

Fig. 4, a shows the obtained calibration curve, and Fig. 4, *b* — the error in determining the volume flow rate of the sprayed solution for measurement times 2, 5, 8 and 10 min. The slope coefficient of the calibration curve for methanol is 1.7 ml/min; *R*-square is 0.99, which shows a very accurate coincidence of the data and the curve. The points were obtained by averaging all data over all times for a given speed of operation of the peristaltic pump, the error was ±0.2 ml/min. It can be seen from fig. 4, *b* that the error in calculating separately for each time is different, it increases with the increase of the time. At the same time, all measurements were carried out at the same distance to the cuvette. This deviation of the error is probably attributable, first of all, to the insufficient number of repeated measurements for times 8 and 10 min (from 5

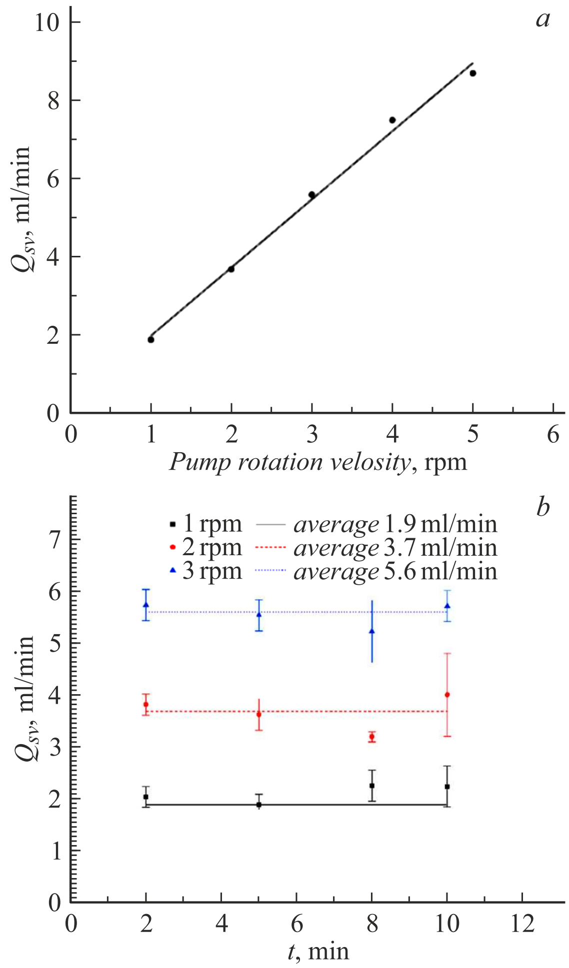

**Figure 4.** Calibration curve of the flow rate of the sprayed solution of the multicapillary methanol system (*a*) and the error of its measurement in time (*b*).

to 10 measurements) and the evaporation of methanol over time. The cooling of the cuvette was observed during the experiments. Other reasons for the error are the ingress of air bubbles into the capillaries of the liquid supply and the impulsive operation of the peristaltic pump, which may not be sufficiently smoothed by the difference in conductivity in the liquid system. A similar situation is observed for ethanol, but the slope coefficient of the curve is 1.9 ml/min. The difference between ethanol and methanol is related to the intensity of their evaporation; the boiling points for ethanol and methanol are 64.7 and 78.4°C, respectively.

Next, the multicapillary system was tested with electrospray in a gas stream. The proportion of charged particles passing through the transport electrode was estimated during the experiments, i.e. generated by a multicapillary system. Spraying was performed using ethanol according to the previously described scheme shown in Fig. 3. Ethanol was used to evaluate the capabilities of the setup in extreme conditions of electrospray. The collector was placed at distances of *L* 55 or 121 mm. The latter size is the maximum in the experimental setup. Then the multicapillary system was washed with a solution, like in case of calibration. After washing, gas was supplied from the compressor at a volume gas flow rate (Qg) 5.5 or 9 Nl/min (normal liters per minute), voltage was turned on at the spray electrode and transport electrode, ethanol was supplied with a sprayed solution volume flow rate (*Qs*) 2 or 9.5 ml/min. The flow rate from Nl/min was converted to the actual flow rate in l/min for the operating pressure of the measured air of 0.4 MPa according to the formula specified in the technical description of the flow meter: the actual volume flow rate is 11 and 18 l/min, respectively. 10−20 values of the total electrospray current and the current to the transport electrode were measured for each voltage at the spray electrode, due to the properties of ethanol, which make spraying unstable even in gas. The values of the currents were averaged, then the ratio of the current at the transport electrode to the total electrospray current was found, which was subtracted from one to obtain the fraction of charged particles that passes through the transport electrode.

The dependences of the proportion of current (charged particles) passing through the transport electrode, the total spraying current and the current passing through the transport electrode on the voltage for different sets of spraying parameters are shown in Fig. 5 and 6, respectively. The modes in which the points in the figures are obtained are marked by numbers *1–6*. The points in Figure 6 were approximated by polynomials of various degrees using the least squares method with the condition of the largest value of R-squared, which was obtained in the range from 0.97 to

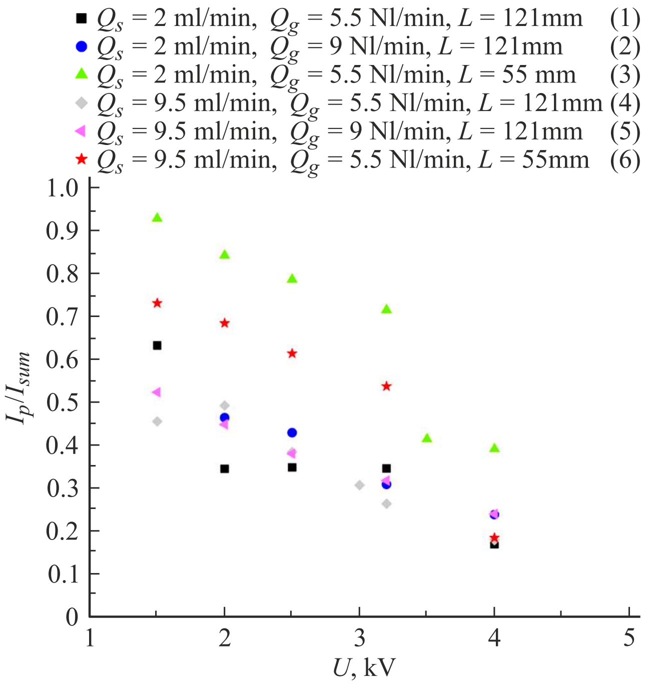

**Figure 5.** Dependence of the proportion of current passing through the transport electrode on the voltage at the spray electrode for various parameters.

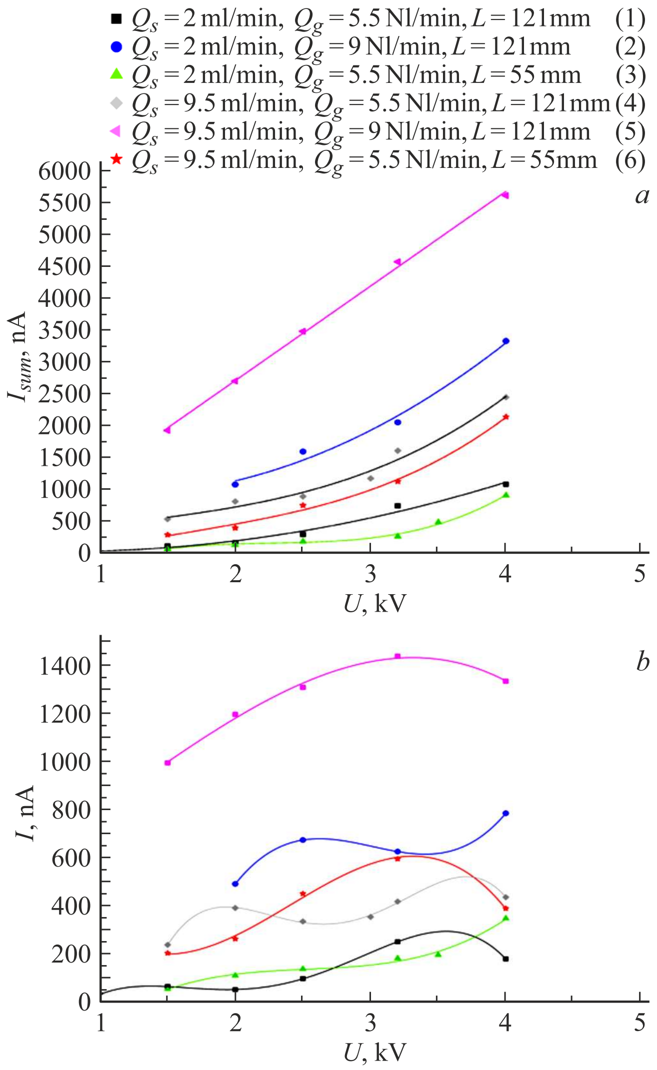

**Figure 6.** Dependences of the current of charged particles generated in a multicapillary system on the voltage at the spray electrode for various parameters: *a* — total current; *b* — current passed through the transport electrode.

0.99. The following polynomials were used for Fig. 6,*a*: modes *1, 3* — polynomial of the second degree; modes *2, 4, 5* — a polynomial of the third degree, *6* — linear dependence; in Fig. 6,*b*: modes *3* and *5* —a polynomial of the fourth degree, the rest of the curves — a polynomial of the third degree

The proportion of charged particles from 10 to 60% is obtained for modes *1, 2* and *4, 5* (Fig. 5). A decrease of the proportion of charged particles with an increase of voltage is associated both with an increase of the total current, as can be seen from Fig. 6,*a*, and with an increase of the pulling field to the transport electrode. An increase of current inevitably increases the space charge, which pushes the particles apart more strongly, resulting in their deposition on the transport electrode or returning to it, i.e. losses. The effect of the space charge on the current passing through the transport electrode is especially clearly visible (Fig. 6,*b*). Thus, the passing current through the transport electrode decreases to values obtained at voltages of about 2.5 kV when the spraying voltage is greater than 3 kV in the modes *1, 4–6*.

As can be seen from the comparison of modes *1, 2* and *4, 5* (Fig. 5) obtained at a distance of 121 mm from the collector, an increase of the volume flow of gas does not result in an increase of the proportion of charged particles passing through the transport electrode at the same sputtering voltages. This can be explained by an increase of the spraying current and, accordingly, the space charge. It turns out that the gas not only carries away the space charge, but also increases the charge density in the aerosol, which produces the opposite effect, which is especially significant with the solution volume flow rates of 9.5 ml/min. Despite this, the multicapillary system shows the efficiency of usage of gas for reducing the space charge. The curve for the mode *2* in Fig. 6,*b* clearly shows that no drop of the transmitted current is observed when the voltage is increased from 3 to 4 kV, unlike the modes *1, 4–6*. With an increase of the volume flow rate of gas to 9.5 Nl/min (18 l/min), the system allows obtaining close fractions of charged particles that have passed through the transport electrode, while increasing the total current by 3.5–4 times. An increase of the proportion of charged particles passing through the transport electrode with a larger gas flow can be expected, which is the subject of further experiments.

The highest proportion of charged particles is typical for the modes *3* and *6* (up to 90%) at a short distance to the collector of 55 mm and low spray currents up to 1000 nA (Fig. 5). A comparison of the modes in pairs *1* and *3, 4* and *6* reveals a significant role of the distance to the collector. For instance, the proportion of charged particles increases at a distance of 55 mm, and the spraying current is greater at a maximum distance of 121 mm. One possible explanation for this phenomenon lies in the field of space charge, which affects the electric field of the capillaries, reducing it. Therefore, the spraying current decreases at 55 mm, since the charge density in the area behind the transport electrode is greater in this case. The effect of the space charge in case of change of the distance to the collector is the most noticeable at a voltage greater than 3 kV when comparing modes *1* and *3* in Fig. 6,*b*.

The system operates simultaneously in the mode of pneumatic spray and electrospray. The main contribution to spraying is made by pneumoelectric spray up to 2.5 kV, since a significant increase of the total current is observed after that (Fig. 6). At the same time, a significant increase of the current at a voltage greater than 3.2 kV results in a decrease of the proportion of charged particles that passes through the transport electrode, which is associated with an increase of the space charge (Fig. 6). The operating voltage for electrospray in a multicapillary pneumoelectropray system ranges from 2.5 to 3.2 kV, so as not to significantly reduce the proportion of charged particles passing through the transport electrode.

The theoretical values of the gas velocity in the region of the transport electrode were obtained for the maximum gas flow used in the experiment. The gas velocity is calculated in the ANSYS Fluent program version 2021R1 by simulating the flow of air through an annular channel formed between liquid and gas capillaries, as shown in Fig. 2. The geometric dimensions correspond to the dimensions of the capillary assembly of the multicapillary system in Fig. 2. We take the overpressure at the inlet to the annular capillary channel 0.4 MPa, gas temperature of 300 K as input data. We use a system of Navier−Stokes equations in an axisymmetric and/or three-dimensional setting for finding the characteristics of the gas flow in a capillary and an annular jet. Numerical integration is performed using the control volume method.

The flow of gas behind the capillary should be attributed to jet streams. A distinctive feature of jet streams is their propagation over a distance much larger than their caliber, i.e. transverse size. The gas outflows from the annular capillary with the flow blockage effect, therefore, it can be characterized as wall-adjacent and underexpanded following the generally accepted classification. The pressure in the jet exceeds the pressure in the surrounding space in this case. The jet expands due to the transverse pressure gradient in the jet, the gas in the jet accelerates to supersonic speeds. As a rule, supersonic zones are closed by a surge of compaction, but at low values of the Reynolds number, a transition to subsonic flow is possible without the explicit formation of a surge of compaction. The gas velocities decrease and range from 100 to 200 m/s at an overpressure of 0.4 MPa in the area of the transport space electrode.

## Conclusion

Thus, the developed hybrid multicapillary pneumoelectrical spraying system allows generating charged droplets during electrospray at a maximum value of the volume flow rate of the sprayed solution of at least 10 ml/min. Such volumes are sufficient for its use in microdroplet synthesis both for real applications in pharmaceuticals, medicine, and for studies of the course of chemical reactions in charged droplets. The proportion of charged particles generated by the multicapillary system ranges from 10 to 60% of all charged particles obtained by electrospray, the rest are deposited on the transport electrode or returned to it. Transmission limitation is associated with various effects of space charge, therefore, for the most efficient operation of the multicapillary system, the correct selection of the sputtering voltage and the volume gas flow rate is required to minimize this effect.

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### Conflict of interest

The authors declare that they have no conflict of interest.

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