Investigation of the Energy Balance in the Spark Discharge Generator for Nanoparticles Synthesis

D A Mylnikov, A A Efimov and V V Ivanov
Moscow Institute of Physics and Technology, 9, Institutsky per., 141700
Dolgoprudny, Russia

mylnikov.da@yandex.ru

Abstract. In this paper we investigate the balance of energy in the discharge circuit of a spark discharge generator (SDG) for nanoparticles synthesis. The released energy consists of several parts: the energy in a discharge gap and the energy dissipated in the other elements of the circuit. In turn, in the gap a one part of the energy releases in preanode and precathode regions and the other part in an arc between electrodes. We measured these parts and proposed ways to optimize energy efficiency of the nanoparticles production.

1. Introduction
Nanoparticles of ultra-small sizes (1 – 10 nm) are in demand for a number of promising applications: gas and optical sensors (Ag, SnO\textsubscript{2}, etc.) [1], luminophores (Si, Ge, etc.), different methods of aerosol printing [2], manufacturing of inks for jet printing (Ag, Al, Si, Ge, SiO\textsubscript{2}, C, etc.). With these inks by methods of printed electronics it is possible to create transistors [3], the elements of solar panels [4] (Si, Ag, Al, etc.), antennas, and other devices.

There are various methods of nanoparticles gas-phase synthesis: the electric explosion of wires, the laser vaporization of the target, the electrical erosion of electrodes in the pulsed gas discharge. The electric explosion, in this series, was historically the first method, which gives nanoparticles with a large mass yield of the order of hundreds of grams per hour, but the resulting particles are of sufficiently large diameters, on the order of 20 – 300 nm. The electric explosion is by far the most energy-efficient gas-phase method (~ 50 kWh/kg), as all the energy input goes directly to the evaporation of a wire and particle formation. The laser vaporization method produces much smaller particles (10 – 30 nm), but requires specially trained targets of compressed powder. Energy costs account for 80 – 300 kWh/kg. The gas-discharge synthesis is a unique method that allows one to obtain particles of very small size, up to clusters of a few atoms [5]. Other advantages include possibility to obtain nanoparticles of any conductive material, including metals and doped semiconductors. There are only two requirements to a workpiece material, not too high electrical resistance and an ability to manufacture a rod from the material to be used as an electrode. At the same time, this method requires a lot of specific energy input about of 1600 kWh/kg. For the industrial use of the method it is of interest to find ways to minimize particle synthesis energy.

In the aerosol SDG a capacitor charged to a high voltage discharges through a system of the two electrodes with the gas gap between them. The energy releases in the discharge gap (useful energy) and in the capacitor and busbars. It is clear that heat released out of the gap should be kept to a minimum, this can be done by reducing the resistance of an external part of the circuit. The ratio of the energy released in the gap to full one is usually about of 50 – 100%.
In the gas discharge gap a part of the energy goes into a gas heating, plasma generation and light emission, and another part is spent to the erosion of the electrodes surfaces and the formation of nanoparticles. Mechanisms of the erosion of the electrodes are not fully elucidated, there are several hypotheses. The formation of nanoparticles is possible due to a strong local heating of the electrodes, which leads to evaporation of the substance, with following condensation into nanoparticles. Heating could be a result of a Joule heat by flowing current or radiative and convective energy flows from plasma channel and ions bombardment, because the resulting plasma channel has a high temperature of the order of several thousand Kelvin. Detachment of droplets of molten substances or solid particles is possible under the influence of locally concentrated electric field having an explosive nature. It is necessary to experimentally investigate the energy mechanism of electrodes erosion, although in the literature there are several theoretical hypothesis [6, 7]. Evaluation of energy consumed for evaporated mass gives a value of only about 0.2% of the energy stored in a capacitor [6]. Therefore it is of interest to study the energy processes in the gap and methods to control the energy balance with the aim of increasing the portion of a useful part of energy.

2. Experimental setup

We use the spark discharge generator in a rod-to-rod configuration with 3 electrode pairs connected in series [8]. A scheme is based on the multigap switches previously proposed by G. A. Mesyats [9]. The discharge circuit comprises of the 1.0 μF capacitor, connecting busbars and three pairs of electrodes connected in series. The breakdown is produced by a high-voltage pulse of 1 – 10 ms in duration and 20 kV in amplitude. The pulse is applied to one of the medium electrodes. The electrodes are 10 mm in diameter titanium and copper rods, and the gap width between them ranges from 0.5 to 1.5 mm in the current work. The electrodes are arranged in an insulating ~ 30 mm diameter plastic pipe, through which a constant air flow of 100 l/min is fed.

Experiments are carried out with one and three pairs of electrodes. In the experiments with a single pair the two others have been replaced by copper rods, overlapping the discharge gap. Thus the inductive component of the circuit resistance remains the same, only the component changed that directly related to a voltage drop across the electrode gap.

There are several approaches for measuring a voltage and current in the circuit including resistive current detectors. Mostly commercial high-voltage probe and current coils are used, while we use custom-made Rogovskii coil with coefficient of $2.1 \times 10^8$ A/(V·s) and 1:500 capacitive voltage divider. Advantage of our coil is that it is made to measure not current but derivative of the current and allows us to see cathode-anode voltage jumps.

Measurements of both current and voltage in the discharge gaps allows us to calculate the energy released in them by the equation

$$E = \int U_{gap}(t)I(t)dt$$

Measurements of power is quite challenging, for several reasons. It requires precise phase synchronization between the measured current and voltage. The voltage on a stray inductance of the circuit exceeds several times the active voltage across the discharge gap. Also, the signal from the Rogovskii coil and a voltage divider is of the order of 1 – 10 V and is under the influence of electromagnetic noise from the discharge circuit with a current of 3 kA. Therefore, we used our measurements scheme protected against a noise [10].

3. Results and discussion

The equation of oscillations of our circuit is:

$$LI + IR + \frac{d}{C} = 0,$$

where $C$ is the capacitance of the storage capacitor, $R$ is the total resistance of the circuit including the internal resistance of the capacitor and the impedance of the electrode spaces, $L$ — capacity of the circuit.

In this circuit current oscillations can be evaluated described by the formula at assumption of constant resistance:
\[ I = U_0 \sqrt{\frac{C}{L}} \omega_0 e^{-\delta t} \sin \omega t \]  

where \( U_0 \) is the initial voltage on the capacitor, \( \delta = R / 2L, \omega_0 = \sqrt{1/LC}, \omega = \sqrt{\omega_0^2 - \delta^2} \). For our circuit, we can assume that the current in a circuit is directly proportional to the initial voltage.

Typical waveforms of voltage and current derivative are shown in figure 1. We integrated the measured curves of \( dI/dt \) and fitted them by formula (3).

\[ W = W_C + W_{\text{arc}} + W_{\text{drop}} \]  

\[ W = \int I(t)^2 R_{\text{C}} dt + \int I(t) U_{\text{arc}}(t) dt + \int I(t) U_{\text{drop}}(t) dt \]

Here \( U_{\text{drop}}(t) \) is a square wave, i.e., equal to \(+U_{\text{drop}}\) during the first half of the period of oscillation, then the current direction is reversed, and the second half period of the current oscillations, the voltage drop becomes equal to \(-U_{\text{drop}}\), etc.

We introduce the resistance \( R_{\text{arc}} \), the equivalent active resistance of the arc discharge. It is such a resistance that dissipates the same power as the discharge gap given the waveform of the current. \( R_{\text{drop}} \) is the equivalent resistance due to the voltage drop at the cathode and the anode in total. The total resistance of the discharge gaps is \( R_{\text{gap}} = R_{\text{arc}} + R_{\text{drop}} \):

\[ W = \int I(t)^2 R_{\text{C}} dt + \int I(t)^2 R_{\text{arc}} dt + \int I(t)^2 R_{\text{drop}} dt \]

We measured \( R_{\text{C}} \) at the operating frequency by RLC-meter and got a value of 60 m\( \Omega \).

Rompe and Weizel proposed the following formula for the arc resistance without taking into account the voltage drop in the near-electrode regions [11]:

\[ R_{\text{arc}}(I,t) = \frac{d}{I(t)^2 dt}, \]
where \( d \) is the length of the arc.

Equating the members relating to \( W_{\text{drop}} \) in (5) and (6) we can calculate \( R_{\text{drop}} \), at the assumption that the dependence of the current on time \( I(t) \) is given:

\[
R_{\text{drop}} = \frac{U_{\text{drop}}}{I_0} \left( \frac{4 \delta}{\omega} \left( 1 + \frac{2}{\exp(\pi \delta / \omega)} - 1 \right) \right),
\]

where \( I_0 \) is the amplitude of the current in the circuit, \( U_{\text{drop}} \) is the sum of cathode and anode voltage drop.

\( R_{\text{arc}} \) and \( R_{\text{drop}} \) depend on the current, both values decrease while current increase, and \( R_c \) is a constant. Therefore, if you change the current in the circuit the ratio of \( W_c \) and \( W_{\text{arc}} + W_{\text{drop}} \) changes.

Useful energy, energy released directly in the gap can be controlled by changing the current through the voltage variation, the inductance of the circuit, width and number of gaps.

As a first approximation we can assume that the resistance \( R_{\text{arc}} \) and \( R_{\text{drop}} \) depends only on the current amplitude \( I_0 \): \( R_{\text{arc}} \sim 1/I_0^2 \) and \( R_{\text{drop}} \sim 1/I_0 \). With the increase of the current \( R_{\text{drop}} \) decreases at a slower rate and begins to prevail in the resistance of the gap.

We measured the voltage and current waveforms for different voltages across the capacitor and different gaps, and calculated on the overall circuit resistance, the gaps resistance and the fraction of energy delivered directly in the discharge gap according to the formula (1). Figure 2 shows that with increasing voltage, and thus with the increasing current, the resistance of the circuit decreases. This is due to the decrease of the resistance of the gaps as the parasitic resistance of the circuit remains constant. Therefore, the efficiency of the energy delivering to the discharge gap (gap efficiency factor) also decreases accordingly. Theoretically, it is equal to \( \eta = R_{\text{gap}} / R \). The measured value is qualitatively consistent with this one.

\[\text{Figure 2.}\] The dependence on the voltage a) for the gaps resistance of Cu and Ti electrodes for one and three gaps; b) for the corresponding value of the gap efficiency factor.

If you increase the length of the discharge gap \( R \) arc the gap efficiency factor should increase. It can be seen if we compare measurements for the Ti 0.7 mm gaps and 1.5 mm gaps.

One can see a strong difference in the resistance of the circuit and, as a consequence, gap efficiency factor for single and three gaps for copper. For the three gaps the resistance is \( \approx 100 \text{ m\Omega} \), for the one is \( \approx 33 \text{ m\Omega} \), which gives the resistance of the single gap of \( \approx 33 \text{ m\Omega} \). In the case of three gaps, they consume a large share of the initial energy of the capacitor, increasing the efficiency gap factor. Energy redistribution towards the greater release in the gap is possible by reducing the circuit and capacitor resistance. This should lead to an increase in the mass yield of particles as well.
Figure 3 shows the relationship between \( R_{\text{drop}} \) and total resistance of the gaps. Which direction we must shift the balance between energy input in the arc and near-electrode region, the question is not as straightforward, but it’s safe to say that is possible to displace this balance both in the direction of increasing energy in the arc (by decreasing the current and increasing the distance between the electrodes) and in the direction of increasing energy in the areas of near-electrode voltage drop by increasing the current.

![Figure 3](image.png)

**Figure 3.** The dependence of the ratio of the equivalent resistance of near-electrode regions and the gap resistance on the voltage.

Figure 4 shows a weak dependence of the nanoparticles specific energy consumption on the voltage, which allows one to draw conclusions about the management capabilities of the efficiency of nanoparticles generation in the spark discharge by changing the discharge parameters.

![Figure 4](image.png)

**Figure 4.** The dependence of nanoparticles specific energy consumption on the initial discharge voltage. Three pairs of titanium electrodes in series are used.

4. Conclusion

We have measured current, voltage and energy release in various processes in the discharge circuit. There are three major consumers of energy — the resistance of the capacitor and lead bars \( R_C \), the resistance of the plasma in the discharge \( R_{\text{arc}} \) and the resistance in areas of near-electrode voltage drop \( R_{\text{gap}} \). We have shown theoretically and experimentally that the energy release in these three parts of the circuit can be controlled. The energy balance between the release in the gaps and dissipation in the circuit can be controlled by changing the parameters of the electrical circuit, current and parasitic resistance. This balance needs to be shifted in the direction of increase of energy release in the gaps, for example, by selecting a capacitor with low resistance, reducing the length of the bars and increasing their diameter, e.g. choosing copper bars and copper wires of large diameter and making good contacts. We can control gap efficiency factor in the range of 50 – 100%. Resistance of the
plasma column $R_{pw}$ and near-electrode regions $R_{sep}$ is also could be to changed. The regularities of the dependence of energy release on the supply parameters of the circuit allow us to study their influence on the synthesis of nanoparticles and to optimize the design of high-efficiency nanoparticles generators and increase a mass production rate.

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