Spatial distribution and time evolution of metal-containing plasma of a low-current atmospheric pressure discharge with magnesium cathode

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Abstract. The spatial intensity distribution and temporal dynamics of the plasma generated by an atmospheric pressure discharge with magnesium cathode in an argon flow are investigated in a coaxial geometry discharge system. The repetition rate of unipolar pulses was 56 kHz and the pulse duration was 12 μs. The steady-state amplitude of the discharge current was 100 mA at a voltage of about 130 V. Under this operating mode, a local melting of the active cathode surface took place. The evaporated magnesium atoms were captured by the working gas flow and formed a green glow plume around the positive discharge column outside the anode nozzle. The image of the plasma formation was projected onto the entrance slit of the monochromator. The spatial distribution of the radiation intensity and evolution in time of its selected monochromatic components were measured. The radiation spectrum contained groups of ion and magnesium atom lines with wavelengths of 285.21 nm (singlet resonant Mg I); 383.08, 383.36, 383.9 nm (triplet Mg I); 517.3, 517.5, 518.1 nm (triplet Mg I). The results of this work are promising with regard to studying open-type spontaneous radiation sources, as well as the generation of combined gas-metal plasma flows at atmospheric pressure.

1. Introduction
Generation of plasma flows containing metal particles, including atmospheric pressure conditions, is applied for solving practical problems related to the fuel mixture production [1], fabrication of ultra-dispersed powders [2], and creation of spacecraft thrusters [3]. In the fundamental research, the metal-containing plasma is used for generation of optical radiation [4], modeling atmospheric aerosols [5], and synthesis of new materials with unique physical properties [6].

The scope of application of magnesium compounds in the form of powders is wide. Up till 60s of the 20th century, the powder magnesium was applied in photography in the pulse light sources known as “magnesium flash” [7]. Powdered materials based on boron and magnesium compounds are used in the production of jet-propulsion fuels [8], as components of explosives [9], including pyrotechnic devices.

The temperature and electrical properties of magnesium particles determine the efficiency with which its burning reaction is initiated in a gas discharge, when they exist in the form of aerosol [10] or
powder [11], as well as in the presence of the water vapor regarding applications for submarine engines and hydrogen production for the needs of hydrogen power technology [12].

The problem of fabricating magnesium nanoparticles deserves a separate attention. The method of powder generation determines the size of magnesium particles and its burning kinetics in compounded fuels [13]. Ultrafine magnesium powders can be produced by way of dispersion from a solid state as a result of erosion processes in discharge systems [14], by the electron beam action on target materials [15], laser ablation [16], levitation-jet method [17], electrical explosion of small diameter conductive wires [18], evaporation of wires in the plasma of high-current plasma jets [19]. Almost all of the above methods use the action of high energy density on materials to cause a fast phase transition from a solid state to a liquid melt and further to the vapor phase. Of the listed ultra-dispersive powder production methods, the electrical discharge technologies are the simplest. For example, when generating metal vapors in an arc discharge, when melting and evaporating one of the discharge system electrodes [20]. To improve performance, systems with multi-gap arresters consisting of several electrode pairs are used [21].

The methods that employ erosion in cathode spots of spark and arc discharges are disadvantageous in that the size of produced particles is quite large. Their average diameter is tens of micrometers. Therefore, it is important to develop and master new methods of ultrafine powder production based on the thermal erosion of materials which surface is subject to the action of a high-frequency or a glow discharge plasma [22]. This approach allows the powder to be generated with the particle diameter from tens to hundreds nanometers. It is also possible to produce micro-spheres, which are formed during the agglomeration of metal atoms and which can store gases, such as hydrogen [23] - a topical feature in the field of engine fuel materials [24]. The cathode sputtering during a glow discharge burning in an inert gas atmosphere is a source of atomic flows that can be deposited on various surfaces, including plant seeds. This method is beneficial for the seed germination and sprouting [25].

The modern trends in the field dealing with the generation of magnesium atomic flows in atmospheric discharge plasmas was the main stimulus for the present work. Using optical methods, we studied the temporal dynamics and spatial distribution of emitted magnesium particles in an atmospheric pressure discharge with combined flows consisting of gas and metal particles.

2. Experimental setup

Combined gas-metal flows at atmospheric pressure were generated by a plasma source based on a co-axial two-electrode discharge system (figure 1). The cathode consisted of a cylinder with a diameter of 7 mm and a conical tapering with an apex angle of 45-60 degrees. This electrode made of tantalum also served as a crucible. The tip of the cone had a hollow, with a diameter of 2 mm and 6 mm deep, where a magnesium insert was placed. The diameter of the output aperture in the anode nozzle was 2.5 mm.

The distance between the cathode tip and the edge of the anode hole was about 2 mm. Plasma generation processes were studied beyond the anode nozzle output aperture, where the working gas jet mixed with ordinary air at normal atmospheric pressure.

The discharge was electrically fed from a pulse voltage source with an amplitude up to 2000 V. The current was restricted by a 2 kΩ ballast resistor. The discharge voltage was measured using a high-voltage probe Tektronix P6015A with an attenuation ratio 1:1000, the current was measured using a low-inductance shunt of 50 Ω. The signals from the probe and the shunt were input to a Tektronix MDO 3024 oscilloscope. The discharge operated in a pulse mode with a frequency from 50 to 100 kHz and a period duty cycle from 50 to 70%. The discharge current amplitude was about 100 mA at a voltage level of 130 V.

Argon was used as a working gas blown through the discharge system at a consumption rate from 1 to 5 l/min. With the indicated parameter values, within several seconds following the discharge initiation, the discharge started to emanate a visually discernible bright green glow.

The optical radiation spectrum emitted by the plasma was analyzed using an MDR-23 LOMO monochromator. At the monochromator output, radiation in narrow spectral regions fell into a CCD-
camera PI-MAX 2 (Princeton Instruments). The MDR-23 monochromator entrance slit was 30 µm wide. The diffraction grating installed in this monochromator has 1200 lines per 1 mm and the inverse linear dispersion was 1.3 nm/mm. The working wavelength range was 200–1000 nm. The camera spectral sensitivity range was 180–900 nm. The CCD-camera dimensions were 1024×1024 px (linear dimensions were 13×13 mm). The horizontal axis corresponded to the dispersion direction. The instrumental function of the registration system was 0.05 nm.

![Figure 1. Experimental setup.](image)

Time dependence of the radiation in terms of monochromatic components was studied using a Hamamatsu H7732-10 photomultiplier tube (rise time \(\sim 2\) ns). When studying optical characteristics, the plasma source was oriented vertically.

The gas temperature distribution in the discharge burning region outside the anode nozzle was studied using a thermocouple sensor. The thermocouple leads were connected to a digital multimeter to measure the thermo-EMF.

3. Results and discussion

The discharge was initiated after breaking the short circuit between the cathode and anode in the region of the anode output aperture. The discharge burning during several minutes caused local melting of the magnesium insert in the cathode glow region. The measured temperature of the cathode tantalum crucible was 300±50°C, which was significantly lower that the magnesium melting temperature (650°C). However, it was visually noticeable, when looking through the anode hole, that a droplet of liquid metal was forming on the surface of the magnesium insert. The droplet surface had a convex shape. The most power was obviously dissipated near the cathode surface where the liquid-metal area formed. The heat diffusion in the crucible in the direction away from the discharge gap is hampered by at least two factors: the contact area of the magnesium liquid droplet with the cavity walls is too small for an effective heat transfer; and secondly, the convective heat transfer by the argon flow beyond the boundary of the discharge gap. The working gas jet tore away magnesium atomic particles from the surface of the liquid-metal pool. Thus, a mixture of argon and magnesium atoms was transported through the discharge channel, where the atoms were exited and ionized by electron collisions. The plasma glow acquired a green color. (figure 2). Express analysis of optical radiation, which was carried out using an Ocean USB2000 spectrometer, showed that the intensities of lines near
383 and 517 nm corresponding to excited particles of magnesium Mg I were several orders of magnitude higher than the radiation intensity of argon atoms (figure 2). The visually noticeable diffuse green glow was determined by the maximal intensity of the strong line of Mg I corresponding to a wavelength near 517 nm. The plume of decaying plasma also glowed green due to the presence of cooling and agglomerating magnesium atoms in the flow of expanding transport gas [26].

![Emission spectrum](image)

**Figure 2.** Overview of the emission spectrum of the magnesium cathode discharge and a picture of the discharge system during the discharge burning.

![Temporal dependence](image)

**Figure 3.** Temporal dependence of the magnesium cathode discharge: a – current and voltage pulse waveforms; b – discharge optical radiation in corresponding wavelength ranges.

Investigation of the radiation time dependence in narrow spectral regions, corresponding to strong lines of magnesium particles, showed that its dynamics in the wavelength range 272 – 288 nm corresponded to the discharge current pulse duration and also had the length of about 12 µs (figure 3). For the lines in the ranges of 375 – 391 nm and 509 – 524 nm, there was observed a prolonged decay
after the pulse termination. A more detailed study of narrow spectral intervals with the help of the monochromator enabled the spatial intensity distribution to be observed along the discharge system axis outside of the plasmagenerator (figure 4 (b)). Thus, we found that the maximum values of the distributions, corresponding to singly charged magnesium ions Mg II emitting at the wavelengths 280 and 280.66 nm, are reached at a distance of about 4.8 mm from the anode aperture. The distribution maximum of Mg I singlet resonance line at 285.3 nm occurs at a distance of about 4 mm (figure 4).

Figure 4. Characteristics of optical radiation of the magnesium cathode discharge: a – a detailed spectrum in the range 272 – 288 nm; b – intensity distributions of corresponding spectral lines beyond the anode nozzle output.

The Mg I triplet lines, emitted at the wavelengths of 383, 383.36 and 383.9 nm, have the intensity distributions with practically the same positions of maxima at a distance of about 0.9 mm (figure 5). The triplet emitting at longer wavelengths, 517.3, 517.5 and 518.1 nm, has the intensity distributions with the maxima at a distance of about 0.5 mm from the anode aperture (figure 6). Figure 7 shows the dependences obtained by summation over the intensity distributions in the spectral ranges 272 – 288, 375 – 391 and 509 – 524 nm, followed by normalization by the maximum value of each distribution. Thus, one can observe to what extent the intensity distribution shifts outside the anode aperture with decreasing radiation wavelength.
Figure 6. Characteristics of optical radiation of the magnesium cathode discharge: a – a detailed spectrum in the range 509 – 524 nm; b – intensity distributions of corresponding spectral lines beyond the anode nozzle output.

Figure 7. Radiation intensity distribution of strong magnesium lines and temperature of the gas heated in plasma beyond the anode nozzle output along the discharge system axis.

Analysis of the temperature spatial distribution of the gas, heated in the discharge, along the central axis of the plasma generator showed that the temperature dropped monotonously. The maximum temperature was observed directly near the anode exit aperture (about 300°C) at direct contact of the thermocouple with the discharge plasma. With increasing distance from the axis, the temperature decreased exponentially, and at a distance of 13 mm was 120°C.
4. Conclusion

For the discharge-plasma method, when studying the processes of generation of ultra-dispersive magnesium powders, in order to determine the conditions of cooling and agglomeration of magnesium particles, it is important to know the spatial characteristics of the volume filled with plasma. A solution to this problem has been found in the present work. The use of optical method enables one to selectively recover the distribution of radiation generated by a single type of particles. It has been experimentally shown that the maximum of such distribution shifts away from the anode nozzle exit with decreasing wavelength of radiation.

Analysis of the radiation temporal dynamics of strong magnesium lines showed that the radiation emitted by singly ionized magnesium atoms practically repeats the time dependence of the discharge current pulse. The pulse decay of optical radiation of excited magnesium atoms Mg I is less prominent as compared with the decay of Mg II ions. This fact is obviously due to inert dissipation processes of thermal energy in the discharge gap and beyond in the region of the discharge positive column. The active surface of the magnesium insert does not completely cool down within the inter-pulse periods, thus, enabling the generation of magnesium atomic particles to continue.

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