Investigation of Composition and Charge State of Vacuum Arc Single- and Multi-Component Plasma

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Abstract – To form coatings for various functional purposes and to modify properties of materials, vacuum arc discharge (VAD) and non-self-maintained arc discharge with an incandescent cathode are widely used. To develop the technologies of ion-beam and plasma processing of materials and articles, knowledge of the plasma and ion stream parameters is required.

In the present work, laws of change in the mass composition and charge state of gas and metal plasma ions are investigated versus the working gas pressure in ion-plasmous installation. Single- (Ti, Zr, W, Ar, and N_2) and multi-component (Ti-N and Ti-Zr) plasmas were studied in our experiments. Investigations were carried out with the use of a plasma-immersion time-of-flight spectrometer.

1. Introduction

Modern investigations of technological applications of ion beam and plasma streams have demonstrated the necessity of knowledge of not only basic parameters including plasma concentration, ion current density, and average ion energy but also specific parameters including the charge state, mass ion composition, and presence of external impurity in plasma. At present, various methods of investigating the charge and mass states of ions in ion beams and plasma are widely used [1–3].

A developed plasma-immersion time-of-flight spectrometer (PITFS) differs by a simple design and good resolution and provides the opportunity of analyzing the mass and charge ion states in different plasmas [1].

In our investigations, the vacuum-arc evaporator equipped with a plasma filter [4] and the plasma generator based on non-self-maintained arc discharge with the "PINK" heated cathode [5] were used for plasma generation.

Since the "PINK" is used to ionize the working gas in the processes of a finish ion-plasmous clearing, surface activation and coating deposition assisted by plasma [5], investigations of the plasma parameters for this source are important.

2. Experimental setup

The plasma-immersion time-of-flight spectrometer was placed in a vacuum chamber of the NNV6.6-I1 installation coaxially with the plasma source (the vacuum-arc evaporator or a plasma generator "PINK").

Fig. 1 shows the block diagram of the installation.

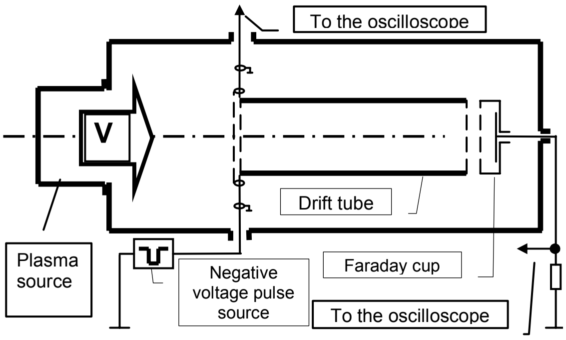

Fig. 1. Block diagram of the installation

Two grids forming an accelerating gap are placed at the input of the PITFS drift tube. One of the grids is fixed on the drift tube, and another is grounded. A negative potential with amplitudes from 500 V to 2000 V and 300-ns duration from a bias pulse source is applied to the drift tube. The Faraday cup is placed at a distance of 20–30 mm from the drift tube output. The transit base length in different experiments was 35, 61, and 100 cm.

The vacuum-arc evaporator was used as a source of metal single- and multi-component plasma. The discharge was supplied from a voltage source including a three-phase transformer and a rectifier operating under voltages up to 70 V and currents up to 200 A (a VD-306 UZ welding rectifier). The current of focusing and stabilizing coils was supplied from a separate source.

A plasma generator "PINK" based on a non-self-maintained arc discharge with the heated cathode was used to ionize nitrogen and argon gases. The discharge current was smoothly adjusted from 15 to 60 A for nitrogen and from 45 to 175 A for argon. In this case, the arcing voltage changed from 46 to 54 V.

Plasma generator provided in the installation working chamber which is a hollow anode for the arc discharge, plasma concentration ~10^9–10^{10} cm^{-3} with a heterogeneity not worse than ±20 % from an average value.

A LeCroy-9354 scope with statistical averaging over 200 waveforms was used to register signals from the Faraday cup PITFS.

3. Experimental results

Fig. 2 shows signals from the Faraday cup for the titanium plasma.

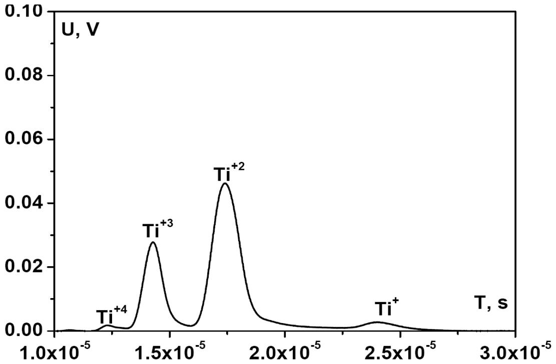

Fig. 2. Signals from the Faraday cup for the titanium plasma

From Fig. 2 it can be seen that peaks of titanium plasma ions are resolved well enough and do not require the application of the method of subtracting waveforms measured with different accelerating pulse durations [6]. The content of singly-charged titanium plasma ions (Ti^{+}) was less than contents of doubly and triply charged titanium ions. The content of the Ti^{+2} ions was maximum, and the content of the Ti^{+3} ions was slightly smaller. Registration of quadruply charged ions in the spectrum demonstrates the high sensitivity of the PITFS. The calculated average titanium plasma ion charge was $\bar{z}=1.94$, which exceeded the value of $\bar{z}$ presented in [7]. This can be explained by the fact that the plasma filter operating in the optimal mode increased the average charge.

The Zr, W, and 50 % Ti – 50 % Zr cathodes were used to produce different metal plasmas.

Fig. 3 shows waveforms obtained with the use of the Zr cathode.

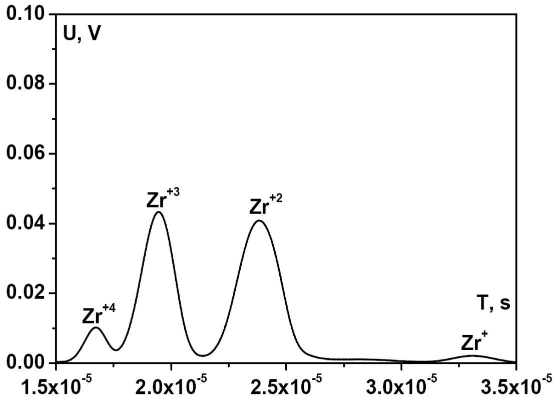

Fig. 3. Ion waveforms for Zr cathode

Since the atomic zirconium mass is by a factor of 1.9 greater than the atomic titanium mass, it can be seen that the transit time for the zirconium ions with different charge states is much greater. The relative peak intensities for the zirconium ions in this case also differed from those for the titanium ions. A much higher content of Zr^{+4} in comparison with Ti^{+4} in the titanium plasma is seen.

The number of singly charged Zr ions in the spectrum is comparable to that of Ti^{+} in the titanium plasma spectrum. The maximum numbers of Zr^{+2} and Zr^{+3} ions in the spectrum are almost identical.

Fig. 4 shows a waveform obtained in the experiment with the composite (50 % Ti – 50 % Zr) cathode.

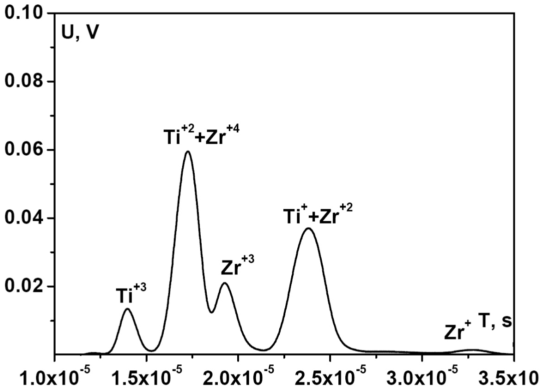

Fig. 4. Ion waveforms for the composite cathode

It can be seen that Ti^{+3} and Zr^{+} peaks are well separated, unlike the Ti^{+2} + Zr^{+4} and Ti^{+} + Zr^{+2} peaks. Since the mass-to-charge ratios for the Ti^{+} + Zr^{+2} and Ti^{+2} and Zr^{+4} ions are not equal but close in values, the Ti^{+} + Zr^{+2} and Ti^{+2} + Zr^{+4} peaks in the spectrum obtained using the Faraday cup are superimposed. To resolve the superimposed peaks, the method of subtracting waveforms measured with different accelerating pulse durations should be used [6]. The peak of quadruply charged titanium is very small, within the limits of a few fractions of percent.

Fig. 5 illustrates the spectrum for the tungsten cathode.

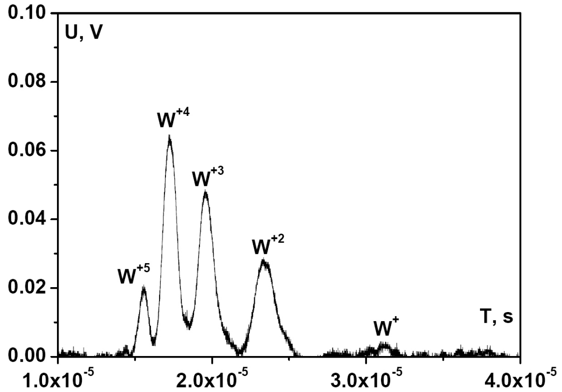

Fig. 5. Ion waveforms for the tungsten cathode

Unlike the spectra for titanium and zirconium plasmas, the tungsten plasma typically comprises the W^{+4} and W^{+5} ions, and probably W^{+6} ions. The number of quadruply charged tungsten ions (W^{+4}) is maximum in comparison with the corresponding numbers of W^{+3} and W^{+2}. The number of singly charged tungsten ions is minimum in comparison with W+5. It can be seen that peaks of W^{+2} and W^{+} ions are well separated, and peaks of W^{+3}, W^{+4}, and W^{+5} ions slightly merge.

Figs. 6 and 7 show the ion waveforms recorded with the "PINK" for N_2 and Ar gases.

The ion current for nitrogen plasma (Fig. 6) has a peak of singly charged molecular nitrogen ion and a peak of singly charged atomic nitrogen.

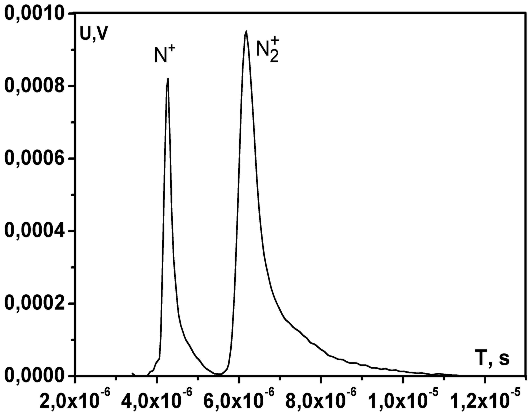

Fig. 6. Typical ion waveforms for the N_2 gas

The argon plasma is mainly used for preliminary cleaning and heating of surfaces before deposition of coatings. Fig. 7 shows the spectrum of the argon plasma. It can be seen that the content of singly charged argon ions is greater than that of the doubly charged ions. The argon peak amplitudes are approximately twice as great as the nitrogen peaks. This is explained by the fact that the discharge current for argon twice exceeded the discharge current for nitrogen. The presence of the molecular nitrogen ion in the spectrum is explained by its presence in the argon container.

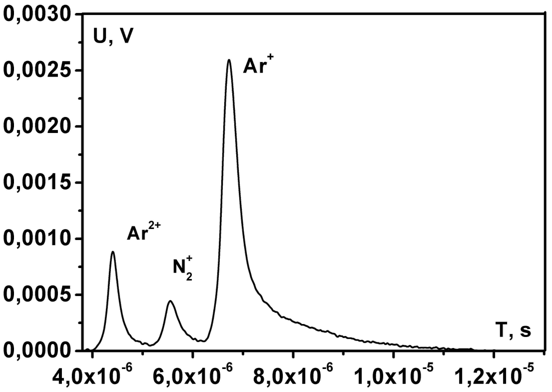

Fig. 7. Typical ion waveforms for the Ar gas

Fig. 8 shows the dependences of change in the integral of gas and metal plasma ion current versus the nitrogen pressure. It is typical that when the vacuum-arc discharge is used for titanium plasma generation, changes of nitrogen pressure in the chamber from 10^{-4} to $2\cdot10^{-2}$ Torr produce no peaks of atomic or molecular nitrogen ions with significant amplitude. This means that no noticeable nitrogen ionization occurs in the Ti plasma and in the gap from the plasma source to the PITFS input.

From Fig. 8 it can be seen that the relative contribution, in percent, of the integral of currents for the titanium plasma ions with different charge states changes with increase in the nitrogen pressure in the chamber. The integral current for Ti^{2+} ions increased by 5 %, and for Ti^{+} ions it increased by 2 % when the pressure increased from 10^{-4} to $2\cdot10^{-2}$ Torr. In this case, the number of Ti^{3+} ions decreased by 7 % when pressure increased from 10^{-4} to $2\cdot10^{-2}$ Torr. The quadruply charged titanium ion peak almost vanished as the nitrogen pressure in the chamber slightly increased.

For the nitrogen plasma, the integral N^{+} ion current decreased by 7 %, whereas for N^{2+} ions it increased by 7 % as the pressure increased from $9\cdot10^{-4}$ to $1.2\cdot10^{-2}$ Torr, at keeping the general discharge current.

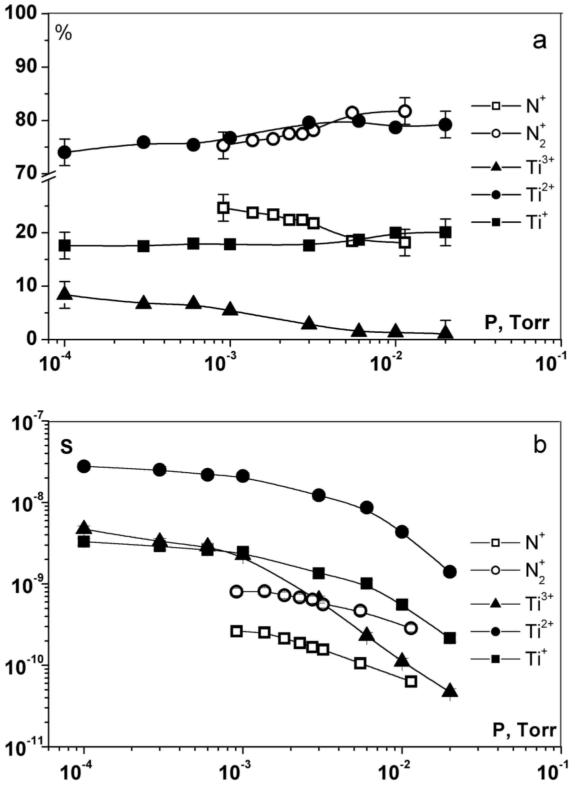

Fig. 8. Dependences of change in the integral of current for ions with different charge states for gas and metal plasmas on the nitrogen pressure in the chamber: a) in percent and b) in absolute values of the current integral

The absolute value of the integral current changed much stronger versus the nitrogen pressure in the chamber. From Fig. 8, *b* it can be seen that when pressure incre ases from 10^{-4} to 10^{-3} Torr, currents for titanium ions with different charge states remain essentially unchanged. With increase in the nitrogen pressure from 10^{-3} to $2\cdot10^{-2}$ Torr, the absolute value of the current integral for Ti^{3+} ion sharply decreased, unlike Ti^{2+} and Ti^{+} ions for which the current integral starts to decrease sharply when pressure increased from $6\cdot10^{-3}$ to $2\cdot10^{-2}$ Torr.

Since the discharge parameters changed with pressure, to ensure stable values of current I_d=50 A and pressure U_d=50 V, they were stabilized using the selection of power discharge supply source and the current of the heated cathode.

In this case with "PINK" application, the current integrals for N^{+} and N^{2+} ions change insignificantly when pressure increases from $9\cdot10^{-4}$ to $1.2\cdot10^{-2}$ Torr, which testifies to stable operation of this source under these pressure variations.

To elucidate the effect of the transition base length on ion charge transfer, experiments were carried out for Ti metal plasma with two transition bases having lengths L=61 cm and L=35 cm. The measured integrals of current for ions with different charge states of metal plasma versus the nitrogen pressure coincided for long and short transition bases. This demonstrates that changes in the transition base length within the above-indicated limits do not affect the charge state of measured ion under conditions of our experiments. Hence, the ionization processes occur in the gap between the cathode and the plasma anode column.

When conducting the experiments to reveal the influence of the discharge burn voltage on the integral current values, the obtained results demonstrated that at changing Ud from 48 V to 54 V there is no significant variation in the current integral.

4. Conclusions

Our investigations have demonstrated that the application of the plasma-immersion transition-time spectrometer for measuring charge and mass composition of plasma ions is possible for different metal and gas plasma sources.

Laws of change in the composition and charge state of gas and metal plasma ions have been obtained versus the pressure of the nitrogen working gas.

The revealed laws are important both for understanding the mechanism of plasma generation in big amounts and for the working parameters optimization of electroarc plasma generators working in technological vacuum ion plasma installations.

References

[1] I.G. Brown, J.E. Galvin, R.A. MacGill, and R.T. Wrigth, Rev. Sci. Instrum. 58, 1589 (1987).

[2] A.I. Ryabchikov, I.A. Ryabchikov, I.B. Stepanov, *Patent RU 2266587 C1*, 2005.

[3] A.I. Ryabchikov, I.A. Ryabchikov, I.B. Stepanov, A.A. Sinebryukhov, S.V. Dektyarev, Rev. Sci. Instrum. 77 (2006).

[4] A.I. Ryabchikov, I.B. Stepanov, S.V. Dektjarev, O.V. Sergeev, Rev. Sci. Instrum. 69 (1998), pp. 893—895.

[5] D.P. Borisov, N.N. Koval, P.M. Schanin, *IV All-Russian Conf. on Modification of Properties of Structural Materials by Charged Particle Beams*, Tomsk, 1996, pp. 34—35.

[6] I.A. Ryabchikov, Candidate's Dissertation in Technical Sciences, Tomsk, 2006, 149 pp.

[7] V.A. Barvinok, *Stress Condition Control and Plasma Coating Properties*, Moscow, Mashinostroenie, 1990, pp. 42—43.