PLASMA PHYSICS AND PLASMA METHODS

UDC 533.9.08 PACS: 52.70.-m
EDN: ESIXGW

The influence of the working gas composition on the mass-to-charge composition of ions in the beam plasma during the evaporation of an YSZ target by an electron beam

A. V. Tyunkov, A. A. Andronov, D. B. Zolotukhin, T. O. Klimkin, A. K. Nesterenko, S. A. Salnikov, Yu. G. Yushkov

The paper presents the results of a study on the mass-to-charge composition of ions in the beam plasma generated during the evaporation of a solid-state ceramic target made of yttria-partially stabilized zirconia (YSZ) in an environment of inert and reactive gases by an electron beam in the forevacuum pressure range. Monitoring of the mass-to-charge composition of beam plasma ions was carried out using a modernized RGA-300 residual atmosphere mass analyzer. The influence of the working gas composition on the mass spectra of ions of the evaporated target material is shown.

Keywords: type of working gas; mass-to-charge composition; beam plasma; YSZ.

DOI: 10.51368/1996-0948-2025-1-24-30

Introduction

A distinctive feature of forevacuum plasma electron sources is that they generate beam plasma with a concentration of up to 10^{11} cm^{-3} during beam transport through the volume of a vacuum chamber [1]. The electron beam formed at forevacuum pressures is an effective tool to modify the surface of non-conductive materials [2, 3], and the generated plasma is used, for example, to initiate plasma-chemical reactions [4].

In addition to assisting and initiating any processes in the plasma, the generated ions are used directly for surface modification [5]. When a negative potential is applied to the substrate, the beam plasma ions accelerate

Andrey V. Tyunkov, Senior Staff Scientist, Candidate of Technical Sciences
Artem A. Andronov, Junior Staff Scientist
E-mail: artem.andronov.98@bk.ru
Denis B. Zolotukhin, Leading Staff Scientist, Doctor of Physical and Mathematical Sciences
Timur O. Klimkin, Junior Staff Scientist
Alexey K. Nesterenko, Junior Staff Scientist
Sergey A. Salnikov, Junior Staff Scientist
Yury G. Yushkov, Head of the Laboratory, Doctor of Technical Sciences

Tomsk State University of Control Systems and Radioelectronics
40, Lenina Ave., Tomsk, 634050 Russia

Received September 17, 2024
Revised October 16, 2024
Accepted December 8, 2024
Scientific specialty code: 1.3.5

and bombard it. By changing the magnitude of the negative potential, it allows to control the magnitude of the ion energy, and, consequently, manage the processes of activation, cleaning or sputtering of the treated surface. Accordingly, knowledge of the ion composition of the beam plasma is important when forming coatings or performing other types of surface modification of products.

Today, mass spectrometric studies of beam plasma in the forevacuum range are complicated, since the ion path lengths do not exceed units of centimeters. To solve this problem, the authors [6] modernized the RGA-100 quadrupole residual atmosphere analyzer and also conducted research on the analysis of the ion composition of beam plasma of various gases.

The study of the influence of the type of working gas on the mass-to-charge composition of ions in the process of evaporation of a target partially stabilized with yttrium oxide zirconium dioxide (YSZ) plays a key role in improving the characteristics of the resulting coating. The choice of working gas directly affects the temperature of the beam plasma electrons, generation, transportation, and focusing of the electron beam, as well as other parameters of the beam and plasma [7], which, in turn, determines the efficiency of evaporation and the uniformity of the distribution of the material over the surface of the product. Different gases can result in the formation of ceramics with different physical and performance properties.

Thus, the study of the influence of the type of working gas on the parameters of beam plasma in the context of electron beam evaporation with subsequent ionization of YSZ ceramic vapors is an important step towards the creation of high-quality materials with the necessary properties and the improvement of their production technologies. This paper presents the results of a study of the mass-to-charge composition of ions generated as a result of evaporation of a solid YSZ target in an atmosphere of various gases.

Experimental technique and methodology

The scheme of experiments for studying the mass-charge composition during the evaporation of a solid target is presented in Figure 1. A fragment of zirconium ceramics partially stabilized with yttrium oxide YSZ (target) was placed on a crucible made of carbon. The electron beam was formed by a forevacuum source with a plasma cathode, after which it passed through an atmosphere of working gas (helium, a mixture of helium with argon, a mixture of helium with oxygen) to the target. During transportation, the beam electrons ionized the working and residual gases. Using a focusing and deflection system, a low-power electron beam ($U_{acc.}$= 3 kV, $I_{em.}$= 23 mA) was positioned on a ceramic target and preheated it to prevent explosive cracking. The electrode system and the operating principle of the electron source are described in detail in [8].

At the same time, monitoring of the gas atmosphere was carried out using a modernized RGA-300 residual atmosphere analyzer (spectrometer) [9] according to the method [6]. The appearance of the upgraded analyzer is shown in Figure 2.

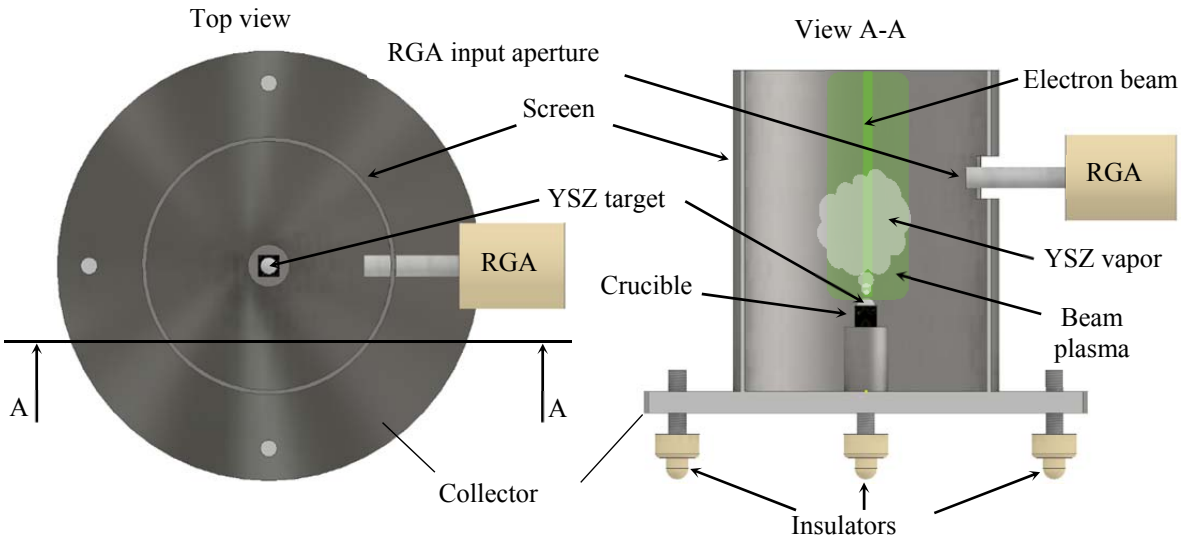


Fig. 1. Experimental scheme

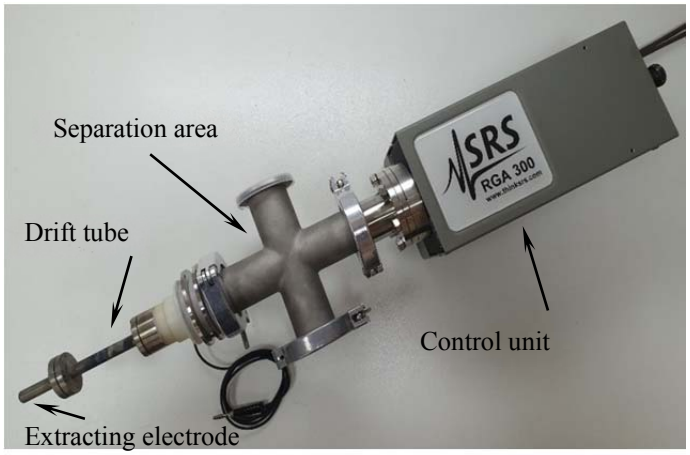


Fig. 2. Upgraded quadruple mass analyzer RGA-300

To control the ion flow into the ion separation area, the crucible and target were placed on the electron beam collector. The collector is made in the form of a massive stainless-steel disk, 10 mm thick and 200 mm in diameter. The disk was electrically connected to a thin-walled cylinder with a diameter of 130 mm. A technological hole was made in the cylinder to introduce the input aperture of the spectrometer. The collector was electrically isolated from the grounded walls of the vacuum chamber and other electrical parts of the unit, and was supplied with a positive potential of 50 V relative to ground. The target to be evaporated was located 4 cm from the input aperture of the mass analyzer, both in the vertical and horizontal directions.

After monitoring the gas atmosphere, the target was gradually heated to a temperature sufficient for its evaporation. At the same time, the mass-to-charge composition of plasma ions was monitored throughout the entire duration. The maximum parameters of the electron beam were as follows: accelerating voltage $U_{acc} = 13$ kV,

emission current I_{em} = 70 mA. The residual pressure in the vacuum chamber was 1 Pa and was provided by a two-stage vacuum unit based on a vane-rotor pump and a Roots pump. The working pressure was maintained at 5.7 Pa. Helium was supplied through the hollow cathode of the electron source to a chamber pressure of 3.92 Pa. Then, additional gas (helium, argon, oxygen) was supplied directly into the chamber to the required value (5.7 Pa).

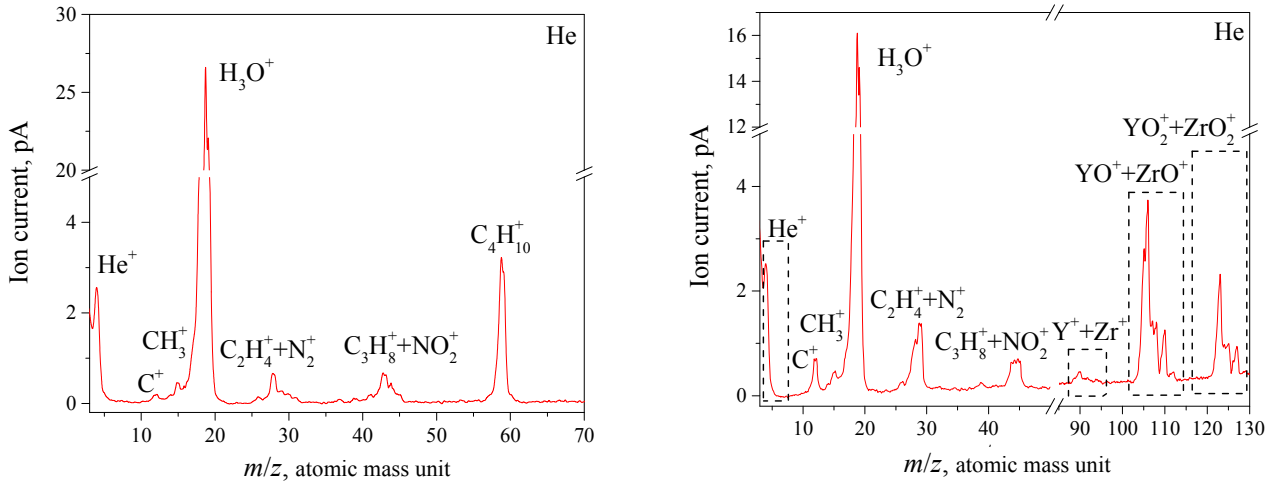
Results of experiments and discussion

Figure 3 shows the ion spectra when using helium plasma gas before evaporation of the YSZ target material (*a*) and during intense evaporation (*b*).

During the heating of the ceramic target up to the evaporation temperature, the spectra (Fig. 3*a*) record ions of the residual atmosphere, mainly ions of water vapor and hydrocarbons, as well as ions of the working gas (helium).

By increasing the power density of the electron beam to the level required for intensive evaporation of the target, ions of all stable isotopes of the target material (Fig. 3*b*), their oxides and dioxides can be detected in the spectra. It should be noted that the amplitude of the water vapor peak decreases as the target evaporation rate increases, while the amplitude of the target material ion peaks increases. The dominant ions of the target material at the initial stage of evaporation, when a melt pool only appears in the beam impact zone, are their dioxides. However, with a further increase in the beam power density, up to maximum values, the peaks of the target material oxides dominate, while the amplitude of the dioxide peaks decreases.

Figure 4 shows the ion spectra when using helium-argon mixture plasma gas before evaporation of the target material (*a*) and during intense evaporation (*b*).

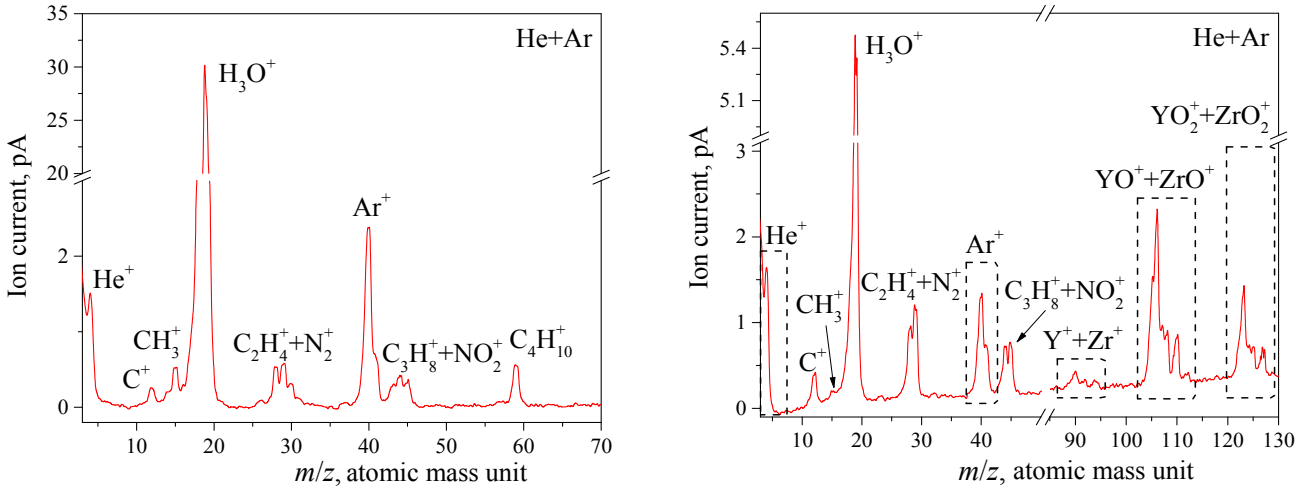


a) *b*)

Fig. 3. Beam plasma ion spectrum before YSZ target evaporation (a) and during YSZ target evaporation (b): working gas – helium

Similar to the spectra where only helium was used as the working gas, the amplitude of the argon ion peaks does not exceed the amplitude of the water vapor peak. A possible reason for the dominance of water vapor ion peaks is the excess of their number in the neutral state over the number of argon atoms. The adsorption of a large number of water

molecules on the walls of the vacuum chamber is facilitated by the low pumping speed of the forevacuum pump and the absence of preliminary heating of the chamber. Since argon is inert and has a significant atomic mass, when it collides with the walls of the vacuum chamber, it desorbs water molecules, which are subsequently ionized by the electron beam. Thus, partial replacement of helium with argon does not lead to significant changes in the ion spectra, except for the appearance of additional argon ions in the spectra. At the same time, an increase in the power of the electron beam and, consequently, the intensity of evaporation of the ceramic target ensures a further increase in the peaks of the target material oxides in the spectra.



a) *b*)

Fig. 4. Beam plasma ion spectrum before YSZ target evaporation (a) and during YSZ target evaporation (b): working gas – mixture of helium and argon

Figure 5 shows the ion spectra when using a mixture of helium and oxygen plasma-forming gas before evaporation of the target material and during intense evaporation. As it shown on Figure 5, the addition of oxygen significantly transforms the ion spectrum. At low electron beam power, the spectrum is dominated by the peak of molecular oxygen ions, which is explained by the large number of neutral oxygen molecules in the vacuum chamber and the close potentials and ionization cross-sections of oxygen and water molecules. Atomic oxygen ions are also recorded, indicating partial dissociation of oxygen molecules and ionization of decay products. However, when the target material evaporates intensively, the amount of oxygen ions decreases significantly, and the peak of water vapor ions becomes dominant. The peaks of target material isotopes (pure Y, Zr) were not detected in the spectra. Only their oxides and dioxides have been recorded. In this case, the peaks of the isotopes of the target material dioxides are predominant, in contrast to the spectra when inert gases are introduced. Increasing the power of the electron beam does not lead to the dominance of oxide ions in the spectra, as when using inert gases. A decrease in the peak of molecular oxygen ions indicates the interaction of target material ions with oxygen ions, which ensures the predominance of dioxide ions at any electron beam power.

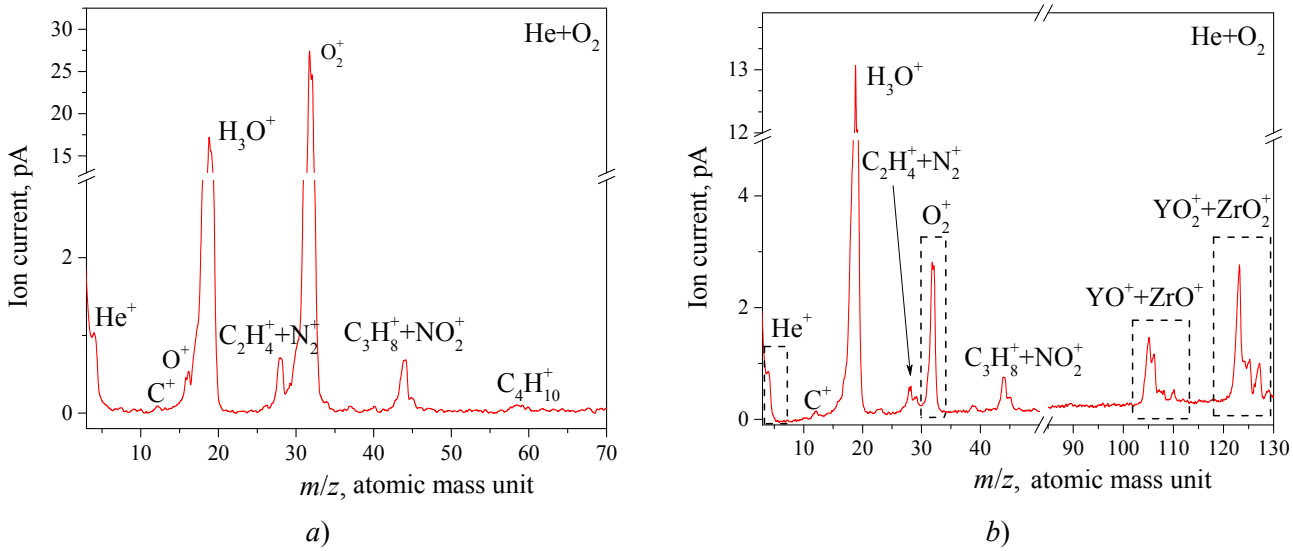


Fig. 5. Beam plasma ion spectrum before YSZ target evaporation (a) and during YSZ target evaporation (b): working gas – mixture of helium and oxygen

A qualitative explanation for these experimental facts may be as follows. When adding inert gases to a vacuum chamber, the dominance of dissociation or oxidation processes is determined by the partial pressure of oxygen and the vapor pressure of the target material [10]. At the initial stage of evaporation, at low beam power, the vapor pressure of the target material is lower than the partial pressure of oxygen, which leads to the dominance of oxidation processes and, consequently, of dioxide ions of the target material. As the evaporation rate increases, the vapor pressure increases as well, which leads to the dominance of dissociation processes and oxide ions. However, when chemically active oxygen is added to the chamber, the total partial pressure of oxygen increases, which prevents the predominance of dissociation and ensures the dominance of dioxide ions in the spectra, regardless of the power of the electron beam.

It is known [11] that after deposition of YSZ coatings by the electron beam method from the vapor phase in a high vacuum, further annealing of the samples is carried out for several hours at a temperature of 700 °C in a furnace in an air atmosphere. This technological operation is carried out to restore the color of the ceramic coating surface lost during oxygen deposition. Therefore, from the point of view of coating deposition, the use of a mixture of helium and oxygen as a plasma-forming gas looks very promising, since the surface of the coatings is subject to additional oxidation directly during deposition. As a result, no additional annealing operation of the samples is required.

Conclusion

Thus, as a result of the experimental studies, it was established that the use of different gases during the evaporation of the YSZ target at the forevacuum pressures changes the ratio of the amplitudes of the peaks of ions of the target materials oxides and dioxides in the mass. When using helium or a mixture of helium and argon as the working gas, the peaks of target material oxides dominate in the target material ion

spectra. Moreover, their signal increases with the increase in the power of the electron beam. The use of helium and oxygen as a working mixture ensures the dominance of target material dioxide ions at any electron beam power.

The work was carried out with the financial support of the Russian Science Foundation, grant 24-79-10026 (https://rscf.ru/en/project/24-79-10026/)

REFERENCES

1. Yushkov Yu. G., Oks E. M., Corbella C., Tyunkov A. V. and Zolotukhin D. B., Ceramics International **46** (4), 4519–4525 (2020).

2. Yushkov Y. G., Oks E. M., Tyunkov A. V., Zolotukhin D. B., Yushenko A. Y. and Yushkov A. Y., Ceramics International **46** (13), 21190–21195 (2020).

3. Golosov D. A., Zolotukhin D. B., Nikonenko A. V., Oks E. M., Tyunkov A. V., Yushkov Y. G. and Yakovlev E. V., Surface and Coatings Technology **383**, 125241-1–125241-8 (2020).

4. Zolotukhin D. B., Lomaev M. I., Oks E. M., Tyunkov A. V. and Yushkov Y. G., Plasma Sources Science and Technology **28** (3), 035018-1–035018-11 (2019).

5. Tyunkov A. V., Zolotukhin D. B., Yushkov Yu. G. and Yakovlev E. V., Vacuum **180**, 109573-1–109573-5 (2020).

6. Zolotukhin D. B., Tyunkov A. V., Yushkov Yu. G. and Oks E. M., Rev. Sci. Instrum. **86**, 123301 (2015).

7. Tyunkov A. V., Yushkov Yu. G. and Klimov A. S. Applied Physics, No. 1, 96–99 (2016) [in Russian].

8. Burdovitsin V. A. and Oks E. M., Laser and Particle Beams **26** (4), 619–635 (2008).

9. Tyunkov A. V., Burdovitsin V. A., Oks E. M., Yushkov Yu. G. and Zolotukhin D. B., Vacuum **163**, 31–36 (2019).

10. Kulikov I. S., Thermal dissociation of compounds. Izd. 2, Moscow, Metallurgy, 1969.

11. Kachanov E. B. and Tamarin Yu. A., Technology of light alloys, No. 2, 16–28 (2010).