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The Effect of Temperature on Electron Beam Brazing of Titanium to Alumina Ceramic

This work studies how the temperature regime used significantly influences the structure of the brazed joint metal

BY A. A. ZENIN, I. Y. BAKEEV, A. S. KLIMOV, E. M. OKS, AND A. V. TYUNKOV

Abstract

The authors have investigated the effect of temperature on the composition and structure of the joint metal produced by electron beam brazing of titanium and alumina ceramic using a fore-vacuum-pressure, plasma-cathode electron source. We found that for brazing temperatures below the titanium polymorphic α-β transition temperature, conditions favorably affect the performance characteristics of the metal-ceramic joint produced. Based on these studies, a strong, tight metal-ceramic joint between titanium and alumina ceramic was obtained.

Keywords

- Metal-Ceramic Brazing
- Aluminum Oxide Ceramic
- Electron Beam
- Fore-Vaccum Pressure Range

Introduction

Metal-ceramic joints are difficult to make due to considerable differences between the physical parameters and properties of the two materials. Thus, to obtain quality vacuum-tight joints, the materials to be joined should have a minimal difference in thermal expansion coefficients and good chemical compatibility (Ref. 1). Most current methods for obtaining vacuum-tight metal-ceramic joints are based on a soldering technique (Refs. 2, 3) in which a preprepared surface of the ceramic item is first metallized by applying a metallized paste or rubbing the paste into the surface. The ceramic is then directly soldered to metal through a binding intermediate solder additive (Refs. 4–6). The main shortcoming of this technology is the complex and time-consuming operation of ceramic metallization with subsequent high-temperature firing. Further, the metallization operation requires the ceramic surface to be specially prepared to have a certain degree of roughness.

Metal-ceramic brazing is conventionally performed in a vacuum furnace (Ref. 7), although alternative heating sources, such as laser radiation (Ref. 8) and electron beams, can also be used for this purpose. Effective electron beam heating of electrically nonconducting ceramic surfaces can be done at somewhat elevated pressures using a fore-vacuum-pressure, plasma-cathode electron source (Ref. 9). We have demonstrated in prior work the possibility of producing metal-ceramic joints with this type of electron source (Refs. 9, 10). However, in this prior work, appropriate consideration was not paid to temperature control of the brazing process or the composition of the gas atmosphere. Also, the question of the influence of temperature on the composition and structure of the brazed joint was not raised. In the work described here, we have extended our investigations on the capabilities and peculiarities of metal-ceramic electron beam brazing performed in various temperature regimes.

Experimental Setup

The experimental setup is shown schematically in Fig. 1. We used alumina ceramic Al_2O_3 samples in the form of 10 × 10 × 5-mm^3 bars. Commercially pure titanium plates 10 × 10 × 2-mm^3 in size were used for the base metal. Aluminum of similar dimensions with a thickness of 1 mm was brazed. The samples were cleansed of impurities and oxides, rinsed in acetone, and placed in a graphite crucible in the vacuum chamber.

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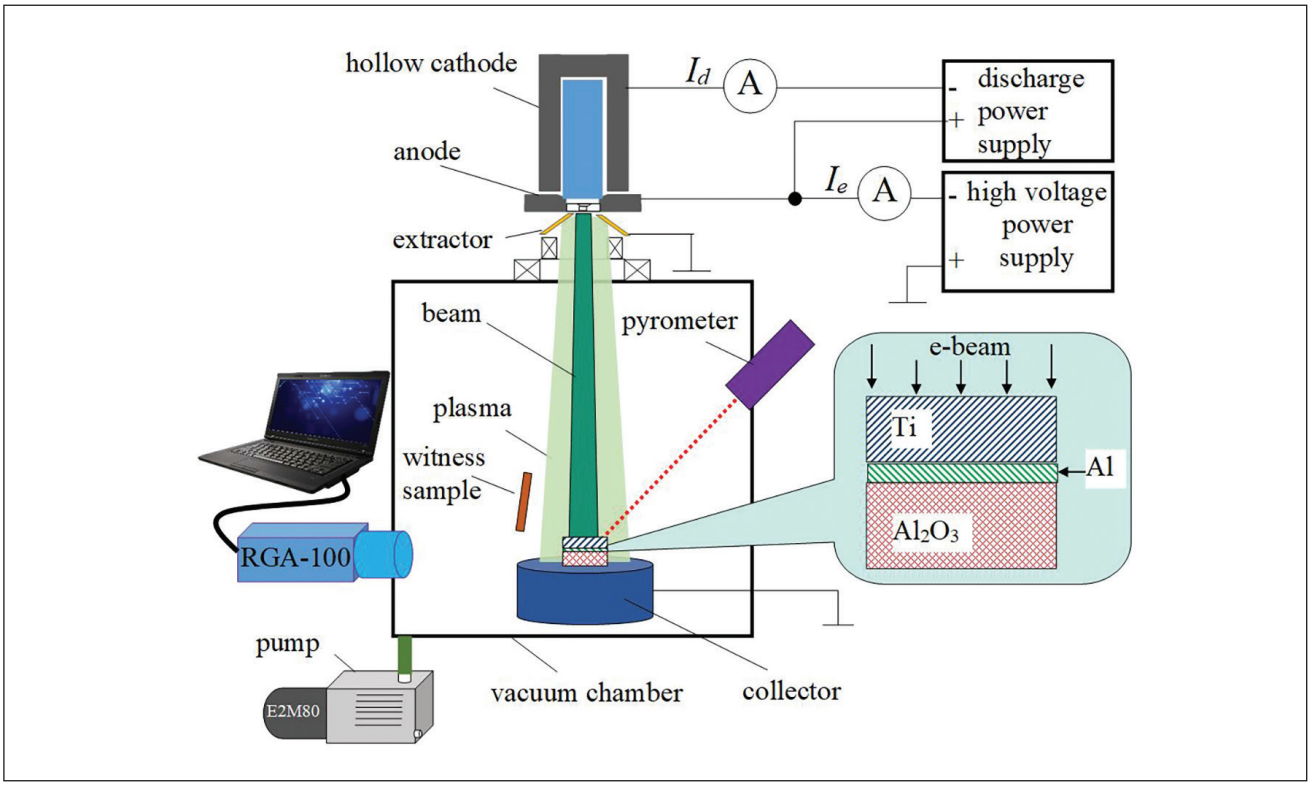

Fig. 1 — Experimental setup.

A fore-vacuum-pressure, plasma-cathode electron source employing a hollow-cathode discharge (Ref. 11) was mounted on the upper flange of the vacuum chamber. The chamber was evacuated using a BOC Edwards E2M80 mechanical rotary vane pump. The pumping rate was sufficient to maintain the required pressure during intense outgassing in the course of brazing. After the pressure reached 2–3 Pa, high-purity (99.99%) helium was admitted into the chamber to a working pressure of 20 Pa. Fore-vacuum plasma electron sources typically operate in an isobaric mode when the pressures in the discharge system, the acceleration gap, and the electron beam transport region are virtually the same. Being an inert gas, helium ensures the purity of brazing and maximum electrical hold-off strength of the source acceleration gap, thereby improving the source's operational stability. Note that using an inert gas during heating of titanium prevents the formation of unwanted brittle oxide, hydride films, and alpha layers on its surface. After the pressure was stabilized, the samples were gradually heated to 750°–1100°C by the electron beam with 8 keV energy and a beam current of 50 to 120 mA. The temperature at the brazing joint was monitored remotely using a Raytec® Marathon MM infrared pyrometer with a measurement range of 540°–3000°C. The entire brazing process took no more than 30 min while the smooth heating of the samples to the melting temperature was carried out for 20 min. After reaching the melting temperature of aluminum, isostatic exposure was carried out 1–2 min then gradually cooled. The thickness of the layer during the experiment was provided by pressing under the action of gravity of the sample. Additional means of controlling the thickness of the brazing joint at this stage of the experiments were not used. The retention of liquid aluminum in the gap was ensured by the forces of surface tension and the geometry of the samples being joined.

The composition of the gas atmosphere during brazing was analyzed using an industrial 100 amu residual gas analyzer (RGA) by Stanford Research Systems. The analyzer sensor was evacuated using a separate pumping station that included membrane and turbomolecular pumps. To provide the required pressure drop between the vacuum chamber and the sensor, the diameter of the analyzer input aperture was less than 1 mm. Since the brazing temperature was greater than the braze alloy melting temperature (for aluminum, about 660°C), the chamber atmosphere's composition was additionally monitored by placing copper witness samples at a distance of 3 cm from the joining samples to indicate the evaporation process. The surface microstructure, elemental composition of the brazed sample cross section, and elemental composition of the film on the witness samples were examined with a Hitachi S3400N scanning electron microscope equipped with a Bruker XFlash® 5010 energy dispersive microanalyzer.

The mechanical properties of the metal-ceramic joints produced were studied using thermal cycling and tightness testing in the atmosphere. Thermal cycling was performed by placing samples in a muffle furnace heated to 350°C for 15 min. After this exposure, the samples were removed from the furnace and allowed to cool to room temperature for 15 min. The thermal cycle was repeated ten times. The tightness was tested by immersing the metal-ceramic samples installed in a fitting into water and admitting compressed air

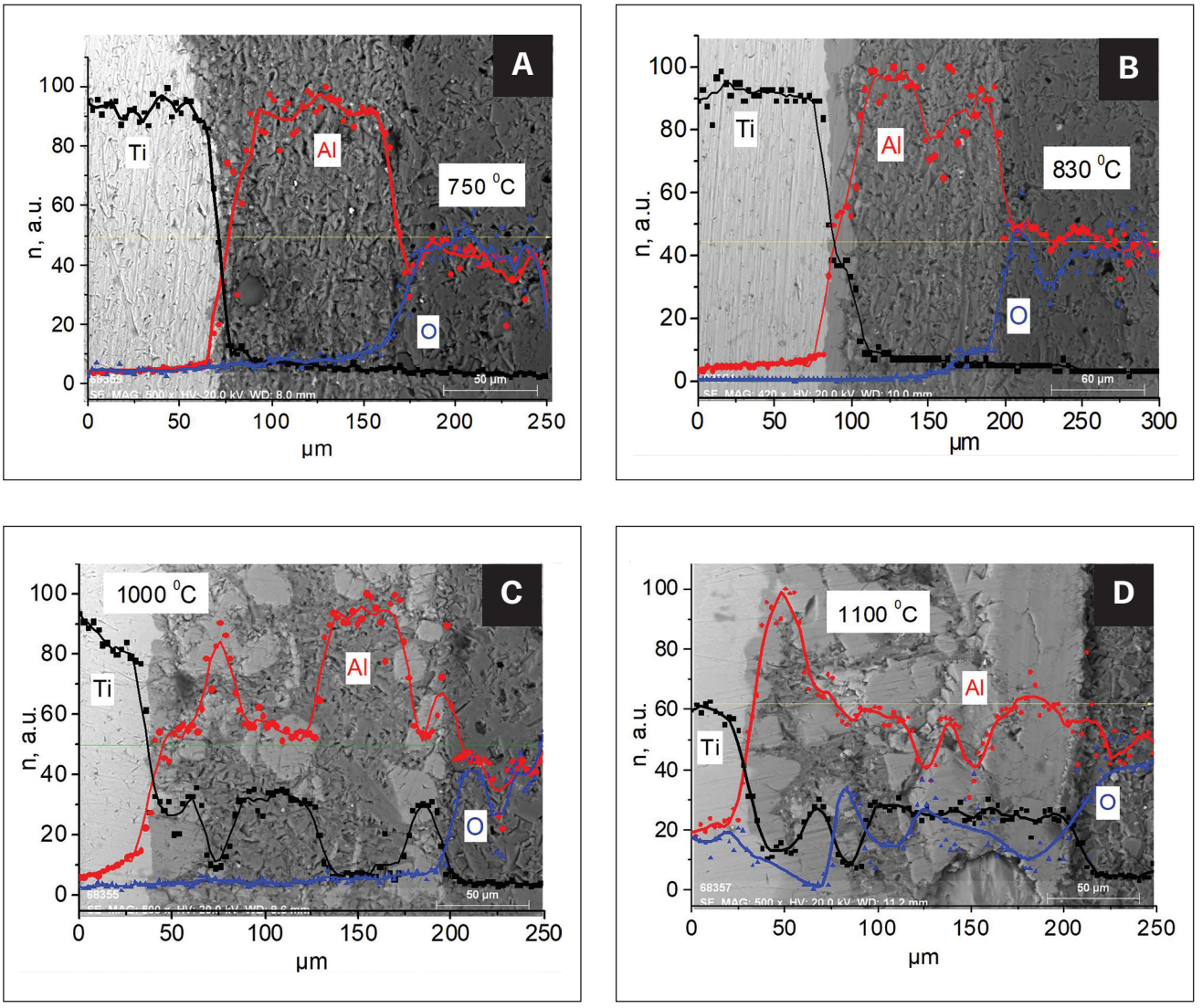

Fig. 2 — Scanning electron microscope images of brazed joints.

at a pressure of 6 bar. The connection was considered tight when there were no air bubbles.

Results and Discussion

Figure 2 shows microphotographs of the cross section of brazed joints obtained at different brazing temperatures from 750° to 1100°C (in all photographs, titanium is on the left, aluminum is in the middle, and alumina ceramic is on the right). The lower boundary of the brazing temperature of 750°C is due to the aluminum melting temperature. The upper temperature limit was selected taking into account that at high temperatures, metals can actively evaporate, which may not only considerably degrade the brazed joint but also require further cleaning of metal condensed on the ceramic's surface.

The microsection images show that an increase in the brazing temperature fundamentally changed the structure of the joint metal. At a brazing temperature of 700° to 750°C, the thickness of the transition layer between titanium and aluminum was relatively small at about 10–15 μm (Fig. 2A) with a distinct boundary. Increasing the brazing temperature to 800°–850°C brought about a more active interaction between titanium and aluminum, and the transition layer in this case was uniform and larger, 20–25 μm (Fig. 2B). On the aluminum–alumina ceramic interface, the transition layer was fairly thin and almost invisible. A further increase in temperature to 1000°–1100°C resulted in an increased region where intermetallic compounds of the Al_nTi_m type (Fig. 2C) were formed, whereas the structure of the diffusion layer was nonuniform with many pores and cracks. At a brazing temperature of 1100°C, the major part of the brazed joint consisted of intermetallic Al_nTi_m compounds (Fig. 2D).

At a brazing temperature of 1100°C, the surface layer of alumina ceramic (Fig. 3, region 5) registered titanium, and the surface layer of titanium contained oxygen (Fig. 3, region 1). This indicated the replacement of oxygen atoms in the ceramic surface layer by titanium atoms, which can improve adhesive properties (Ref. 14). After the replacement, the

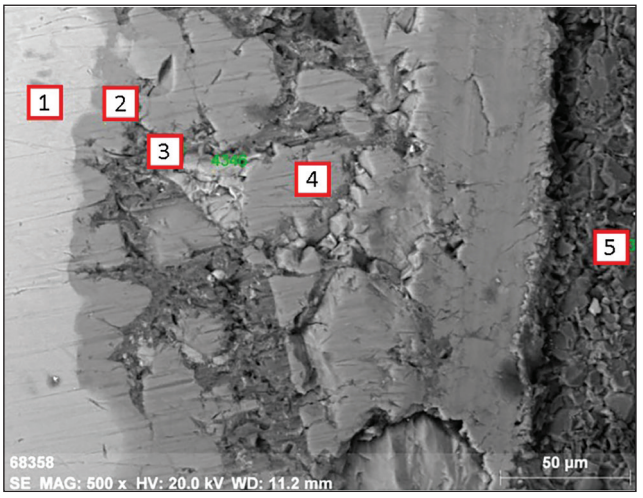

Fig. 3 — Microphotograph of the brazed joint metal.

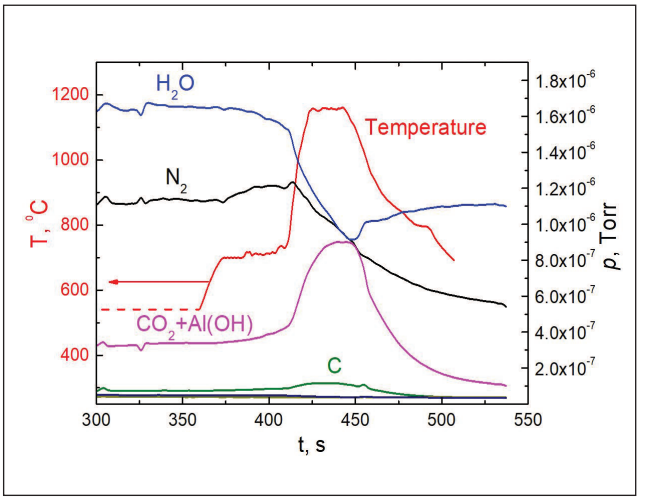

Fig. 4 — Heating dynamics of the brazing samples (red) and composition change in the vacuum chamber's gas atmosphere.

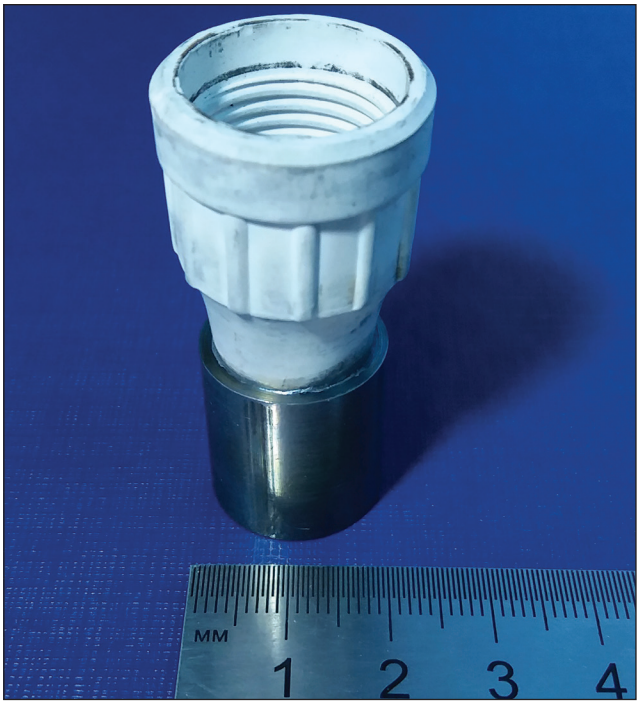

Fig. 5 — Example of electron beam brazing.

oxygen atoms diffused through the aluminum brazing filler metal into the surface layers of the titanium. This was undesirable. The formation of titanium oxide at the titanium-brazing interface contributed to embrittlement upon heating (Ref. 13) since there were significant compressive stresses resulting from a significant difference in the volumes of titanium and its oxides. Regions 2–4 had a similar elemental composition to $Al_xTi_yO_z$ in stoichiometry.

Monitoring of the gas atmosphere when the sample was heated close to the brazing temperature showed (Fig. 4) that at a sample temperature of up to 700°C, the vacuum chamber atmosphere contained, along with atoms of admitted inert gas, atoms of the residual atmosphere (water vapor and nitrogen). Increasing the temperature of the electron beam brazing process above 700°C led to a significant change in the composition of the gas atmosphere in the vacuum chamber.

With an increase in brazing temperature, the energy of the surrounding gas atoms increased, which implied a decrease in the interaction time between hot residual atmosphere atoms and electrons emitted from the analyzer filament. As a consequence, a decrease in the number of ions of the residual atmosphere was observed. Conversely, the number of carbon dioxide ions increased. This was due to the active evaporation of aluminum brazing filler metal at temperatures above 700°C. During evaporation, the brazed atoms interacted with atoms of the residual atmosphere and formed AlOH molecules, which were subsequently ionized by electrons emitted by the RGA filament. The generation of molecular ions of this kind has been observed in prior work (Ref. 13) during evaporation of alumina ceramic by an electron beam. Since ions of CO_2 and AlOH molecules were indistinguishable in mass by our device ($m/z = 46$ in both cases), before active evaporation (700°C) of the brazing filler metal, the curve reflected the number of carbon dioxide molecular ions, and, after that, the total number of molecular ions of CO_2 and AlOH. This assumption was confirmed by elemental analysis of witness samples placed in the immediate vicinity of the brazed samples, revealing that increasing the brazing temperature resulted not only in changing the gas's atmosphere composition but also in the evaporation of brazing filler metal (aluminum) (Table 2). In the brazing temperature range 700°–900°C, the witness sample surface contained the base element with insignificant (error level was about 1%) impurity content. For brazing performed in the temperature range 1000°–1100°C, considerable growth of the aluminum and oxygen content on the sample occurred, indicative of intense evaporation of the brazing filler metal (aluminum) and its binding with oxygen-based molecules.

Our studies of the composition and structure of the brazed joint and gas atmosphere allowed us to conclude that the optimal temperature range for brazing metal-ceramic joints

with aluminum as the brazing filler metal is 800°–900°C. In this temperature range, a uniform transition layer between titanium and the brazing filler metal was formed. There were practically no oxides or hydrides in the titanium surface layer. Only a small outgassing and a negligibly small number of brazing filler metal atoms near the ceramic registered. Further increase in temperature contributed to the formation of a conducting film on the ceramic surface and nonuniform transition layers of titanium oxide, which negatively affected the performance characteristics of the metal-ceramic parts so produced.

Based on this work, we brazed a series of six metal-ceramic joints at a temperature of 850°C (Fig. 5). For the ceramic part, we used a standard argon arc welding machine nozzle

Table 1 — Elemental Composition of Different Regions for a Brazing Temperature of 1100°C

	Elemental Composition (at.-%)			
No.	O	Al	Ti	Other Elements
1	18.4	26.7	54.5	0.4
2	12.8	53.6	33.4	0.2
3	12.5	77.3	9.6	0.6
4	14.1	57.7	27.7	0.5
5	58.6	38.4	2.7	0.3

Table 2 — Composition of Witness Sample Coatings vs. Brazing Temperature

Element	Temperature				
	700°C	830°C	900°C	1000°C	1100°C
Copper	100	96.8	96.68	16.37	2.36
Aluminum	—	1.32	1.99	39.83	47.32
Oxygen	—	1.84	1.34	42.92	49.49
Titanium	—	—	—	0.88	0.83

and a titanium tube with a 16 mm inner diameter and a wall thickness of 2 mm for the counterpart; the brazing filler metal was aluminum.

After running a series of thermal cycle tests of the produced metal-ceramic parts, we found that they all preserved their tightness. The breakage load in the shear stress state was determined by the standard method: the contact area was 200 mm^2, and the breaking force varied from 6 to 8 kN. Thus, the breakage load was from 30 to 40 MPa.

Conclusion

We have explored the electron beam brazing of metal-ceramic parts and found that the temperature regime used significantly influenced the structure of the brazed joint metal. Increasing the brazing temperature from 700° to 1100°C led to a noticeable redistribution of elements in the joint metal, the appearance of an intermetallic phase, and a change in the gas atmosphere near the processed part. We found that at a brazing temperature of 750°–850°C, there was no noticeable mixing of the base material and the brazing filler metal in the joint metal, and a uniform transition layer tens of microns thick was formed. Outgassing at these temperatures was relatively small and brazing filler metal evaporation was insignificant. Increasing the brazing temperature to 1000°–1100°C led to a large outgassing from the surface samples and partial evaporation of the brazing filler metal, which fundamentally affected the electrical characteristics of the ceramic and the formation of aluminum and titanium intermetallic compounds along the entire length of the joint metal; the formed joint was nonuniform with many pores and cracks. Based on the present studies, we produced a tight metal-ceramic joint with a tensile strength up to 40 MPa. The results of the research indicate that electron beam devices can be used as effective production tools for joining metal-ceramic parts.

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ALEKSEY A. ZENIN (*zenin1988@gmail.com*), **I. Y. BAKEEV**, **A. S. KLIMOV**, **E. M. OKS**, and **A. V. TYUNKOV** are with Tomsk State University of Control Systems and Radioelectronics, Tomsk, Russia. **E. M. OKS** is also with the Institute of High Current Electronics SB RAS, Tomsk, Russia.