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EFFECT OF GAS-PHASE ALUMINUM-NITRIDE LAYER MORPHOLOGY ON THE THERMAL RESISTANCE OF Mo-AlN-Mo CYLINDRIC PACKAGES

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Results of measurements of the thermal resistance of Mo-AlN-Mo cylindric packages are related to the microstructure of the ceramic and intermediate layers. It is shown that only a special morphology of the nitride ceramic layer makes it possible to obtain a high-density contact between the ceramic and metal layers which is not destroyed upon subsequent heating of the package and upon thermocycling. The required so-called "composite" morphology of nitride ceramics can be obtained only within a narrow range of synthesis temperatures.

1. Multilayered ceramic-metal electrical-insulating compositions are widely used in technology. Most frequently, oxide ceramics are used in such compositions as an electrical-insulating layer. In a number of applications, the heat conductivity (thermal resistance) of ceramic-metal compositions plays an important role [1, 2]. In this connection, attempts have been made to use in such compositions ceramics with enhanced heat conductivity, in particular, those based on aluminum nitride [3, 4]. However, the microstructure of such compositions has received little attention. In the present work we study the microstructure of ceramics in cylindric multilayer packages with the electrical insulation from aluminum nitride produced by the gas-phase technology, the relationship between the ceramic-layer morphology and the state of the ceramic-to-metal transition zone, and the thermal transport characteristic of such packages. In the present work, the thermal resistivity of the package – the reciprocal of the heat transfer coefficient $R_t = 1/k$ $[\mathrm{K \cdot m^2 \cdot W^{-1}}]$ – is used as a thermal transport characteristic.

2. The gas-phase technology of aluminum nitride crystallization [5] in a tubular MChVP molybdenum substrate 20 mm in diameter was used to form the packages. Aluminum monoammoniate produced at the Podolsk Experimental Plant of the State Institute of Rare Metals was used as a raw material. The temperature of the evaporator with the raw material was maintained at 600 ± 10 K. Monoammoniate vapor was transported by a helium flow with a rate of 1 liter/min. Thermal dissociation of the monoammoniate proceeded on the substrate, which was heated to a certain temperature within the interval of 1500–1700 K. During the layer-deposition process the substrate rotated and reciprocated with respect to the nozzle from which the raw-material vapor flowed.

The subsequent encasing of the ceramic with a molybdenum layer was also performed using the gas-phase technology. The pressure of the vapor of molybdenum pentachloride used as a raw material was set to a level of 150–200 Pa. Pentachloride vapor was transported by dehydrated and purified hydrogen, whose partial pressure was maintained within the limits of 1.5–1.8 kPa. The chloride-hydrogen mixture was mixed in a mixer with a flow of dehydrated and purified helium, heated additionally, and flowed from a nozzle on to the surface of the cylindric substrate (AlN-coated molybdenum) with a temperature of 1200–1400 K. Reduction of molybdenum pentachloride by hydrogen and crystallization of the molybdenum coating proceeded immediately on the substrate surface. The total pressure in the crystallization reactor was maintained at a level of 2.5–3.0 kPa by means of evacuation of the system using a roughing pump. The deposition rate of the quality molybdenum coating on the aluminum nitride layer reached under these conditions 100–200 μm/h.

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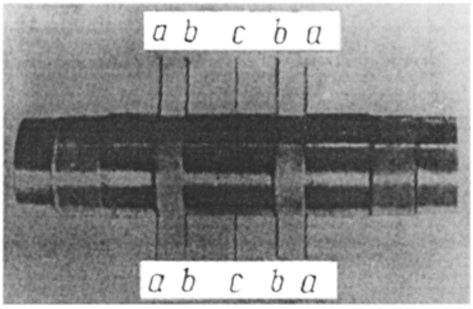

Fig. 1. The appearance of Mo-AlN-Mo package prepared for tests. The package was cut after tests along the lines a-a, b-b, and c-c to carry out the metallographic investigation.

Two control rings were cut from both ends of the finished three-layered package. After grinding and polishing the faces of the rings, the microstructure of the initial ceramic and ceramic-to-metal transition zones was studied. A sample for investigations was fabricated from the middle portion of the package, which consisted of basic and guard electrodes. In order to form electrodes, channels were machined in the molybdenum case to reveal the ceramic, as is seen in Fig. 1. The packages that passed the tests were divided into rings (cross sections are shown in Fig. 1). In all cases, the ring faces were grounded and polished to obtain metallographic sections. Microstructural investigations were performed on a MIM-7 metallographic microscope.

A cylindrical version of the stationary method with internal absorption of heat [6] was used to measure the thermal conductivity. The heat flux was produced by a cylindrical heater of thin tungsten foil. The temperature jump between the molybdenum layers of the three-layer sample under investigation was measured by Chromel-Alumel thermocouples precalibrated against the melting points of pure substances. Measurements were carried out in the temperature region of 700–1100 K. The setup made it possible to remove a heat flux up to 18 $W \cdot cm^{-2}$ from the sample. The relative error of measurement of thermal resistance did not exceed 8%.

3. Results of microstructural investigations are presented in Figs. 2 and 3. Figure 2 presents characteristic morphology types of ceramic coatings of aluminum nitride formed at temperatures of 1370, 1570, and 1670 K: layered, "composite", and coarse-grained morphological types, respectively [7].

Figure 3a presents the structure of the layered-type coating (see Fig. 2a) after formation of the molybdenum case on its surface. It is evident that after the continuous thermal treatment connected with the operation, porosity developed in the darker layers. The porosity at the metal interface remained virtually the same as after applying the ceramic layer. Figures 3c and 3d present the structure of the "composite" coating (see Fig. 2b) after formation of the case. It is evident that after additional thermal treatment, fragmentation takes place in the threadlike aluminum nitride crystals that penetrate the microcrystalline aluminum nitride matrix in the initial ceramic of this morphological type. However, in this case porosity is not developed either within the layer or at the interface between the ceramic layer and metal layers. The structure of the ceramic after formation of a package of the coarse-grained morphological type of nitride ceramic differs little from the initial state (Fig. 2c) and is characterized by coarse porosity both within the ceramic layer and on its interfaces with the metal layers.

After measurements of the thermal resistance of the package with a nitride ceramic of the layered type, the porosity in the ceramic layer increased, and coarse cracks stretching over both the ceramic and the interface of the ceramic and metal layers appeared (see Fig. 3b). In the case of the package with a nitride ceramic of the "composite" type, caking and substantial coarsening of fragments took place after the tests (Fig. 3e) compared with the state of the structure presented in Figs. 3c and 3d. However, no porosity was created in the ceramic layer or on its interface with the metal layers. These results correspond well to data from [8].

4. Results of measurements of the thermal resistance of packages with nitride ceramics are presented in Figs. 4 and 5. The dependences in Fig. 4 correspond to the three morphological types of ceramics. One can see that the packages with the nitride ceramics of the layered (curve 4) and coarse-grained (curve 2) types have a thermal resistance exceeding 2.0 $K \cdot cm^{-2} \cdot W^{-1}$ in the temperature range investigated. Most likely, the effect in this case is caused by both the weak cohesion of the package layers and the structure of ceramic itself, in which in the

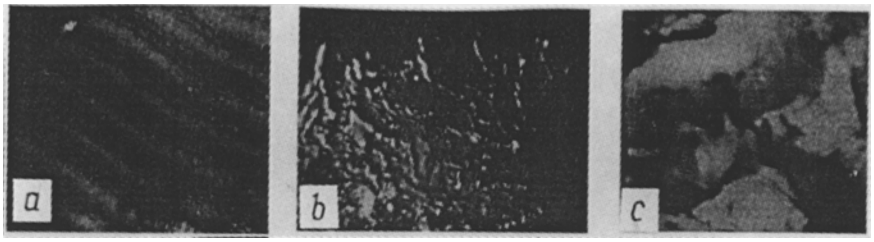

Fig. 2. Microstructure of gas-phase aluminum nitride layers obtained at the following temperatures of the molybdenum substrate: 1370 (a), 1570 (b), and 1670 K (c). ×1350.

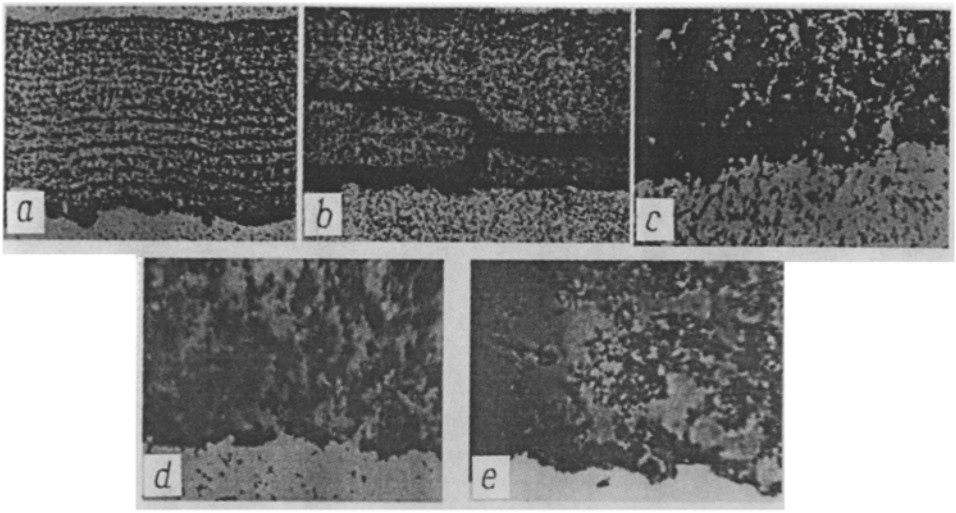

Fig. 3. Microstructure of the Mo-AlN-Mo package: a, b, the ceramic layer is obtained using the gas-phase technology at the temperature 1370 K: a, before tests; b, after the measurement of R_t (×170); c, d, e, 1570 K: c, the interface of the ceramics and the substrate before tests; d, the interface of the ceramics and the case before tests; e, the same after the measurement of R_t ×960.

former case a system of layered pores exists, whereas in the latter case the pores, cracks, and hollows are situated at random. These curves (1 and 2) have an anomalous temperature dependence, which is evidence of the appreciable contribution of the contact resistance to the total thermal resistance of the packages.

Packages with dense "composite" nitride ceramics make it possible to obtain lower values of thermal resistance (curve 3 in Fig. 4) – lower than 1.0 $\mathrm{K \cdot cm^2 \cdot W^{-1}}$. In this case the normal slope of curve 3 bears witness to the negligible contribution of the resistance of the ceramic-to-metal transition zones to the total thermal resistance of packages. The run of curve 3 is most likely determined by the temperature dependence of the heat conductivity of aluminum nitride. According to our estimates, the levels of heat conductivity of the materials of the metal and ceramic layers in the case of "composite" nitride ceramics are approximately equal within the temperature range under investigation.

In the course of the investigations we paid attention to the poor reproducibility of the results of measurements of thermal resistance when a series of packages was fabricated without strict temperature monitoring (pyrometric measurements were not carried out during the fabrication of this series of packages; the temperature of the process was estimated indirectly from the input power of the heater). This is illustrated by Fig. 5, from which is evident that the thermal resistance of the packages varies from 0.6 to 2.0 $\mathrm{K \cdot cm^2 \cdot W^{-1}}$. Subsequent preparation and metallographic investigations of packages have shown that in packages with a high thermal resistance, the ceramic layers are close in their structure to either layered or coarse-grained types. Low values of thermal resistance were found only in packages with "composite" ceramics. It was found later that temperature stabilization of the process based on the current consumed is not reliable, since substantial deviations of the true temperature from

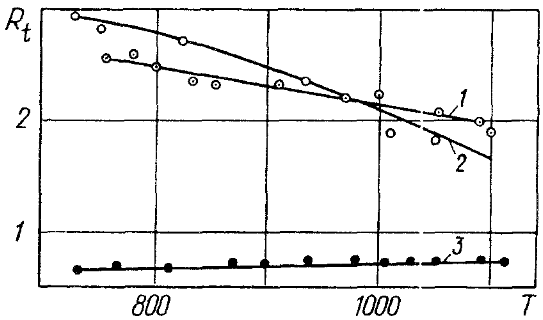

Fig. 4. Temperature dependence of the thermal resistance of packages with electrical-insulating layer of aluminum nitride of different morphological types: 1) layered (synthesis at 1370 K); 2) coarse-grained (at 1670 K); "composite" (at 1570 K). R_t, K·cm^2·W^{-1}.

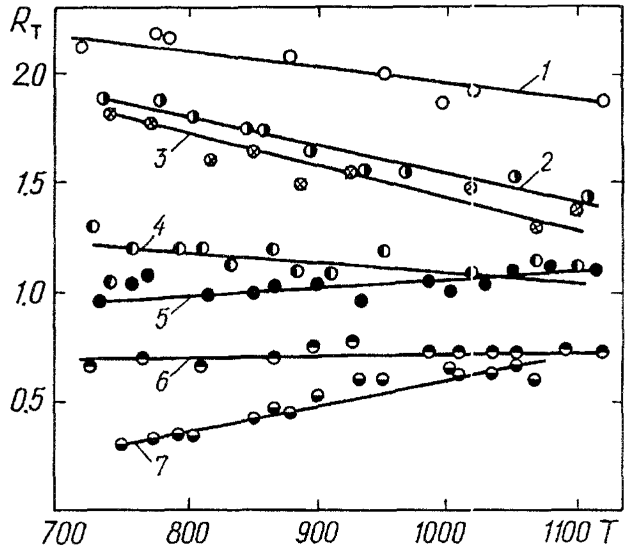

Fig. 5. Temperature dependence of the thermal resistance of a series of packages with electrical-insulating aluminum nitride coating (during the fabrication of the packages, the ceramic layer was formed at a stabilized heater power corresponding nominally to the a temperature of 1550 K): 1) layered type; 2, 3) coarse-grained; 4, 6) transition (from "composite" to coarse-grained); 5) transition (from "composite" to layered); 7) "composite" type.

the preset value are observed as a result of substantial variations in the blackness of the surface of the items [7]. It was found that under conditions of stabilized power at a preset temperature of 1550 K, in fact, it can take values within the range of 1500–1600 K. As a result, the more precise temperature range within which "composite" ceramic layers and, consequently, packages with low values of thermal resistance can be obtained appear to be rather narrow (1530–1570 K).

5. In conclusion, it can be noted that the gas-phase method makes it possible to form multilayer cylindric ceramic-metal packages with molybdenum current-carrying layers and with an electrical-insulating layer of aluminum nitride. Contact heat losses can be almost completely eliminated in these packages when proper temperature control is provided, and the total heat resistance can be lowered to 0.6 K·cm^2·W^{-1}.

The necessity of strict temperature control during the formation of the ceramic layer is determined by the fact that the formation of aluminum nitride of the "composite" morphological type, which provides high characteristics, takes place in the narrow temperature range of 1550±20 K. Only in this case extended defects (pores, cracks) are not formed in the ceramics.

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