Influence of vacuum annealing on the dispersion of thin double niobium-palladium films deposited onto oxide ceramic materials

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Introduction

Since ceramic materials, particularly oxide ones, are usually poorly wetted by molten metals, metal coatings are often used applied in various ways and adhesive-active metals such as Ti, Cr, Nb, Hf and others are the most commonly used to make such coatings. Then, thus metallized ceramic parts are brazed in vacuum or an inert medium by molten metallic solders on the basis of tin, silver, copper, nickel etc. In this case, the thickness of the solder seam is from 50 - 100 μm up to several millimeters. Sometimes, multilayer metal coatings are used, but the thickness of the layers and brazed seams remains high. At the same time, there is information that the reduction in the thickness of the brazed seam leads to a significant increase in the strength of the brazed joint. Obtaining brazed or welded joints of metallized ceramics is possible through reducing the thickness of both the metallization coating on the ceramics and the brazing layer by itself. This can be achieved if the soldering metal or alloy is also applied in the form of a rather thin film, the thickness of which does not exceed several μm.

Permanent vacuum-tight precision ceramic joints with an ultra-thin seam can be made by brazing or pressure welding of metallized ceramic parts. This purpose can be achieved by application onto ceramic surfaces of double metal films, one part of which is 100–200 nm thick and consists of an adhesion-active metal (e.g. Hf, Nb etc.), and the other layer is slightly thicker (1–3 μm) and serves as a solder (e.g. Pd, Ag, Ni etc.) ensuring joining of metallized ceramic materials during brazing or pressure welding with a fine brazed seam (2–4 μm thickness).

The study of two-layer niobium-palladium coatings (films) on oxide materials during annealing in vacuum and the creation of brazed and welded oxide ceramics joints based on them with super-thin brazed seam, the thickness of which does not exceed 5 μm, is the main task of the present work.

Materials and Experimental Procedure

In this paper an electron-beam method for sputtering of metal (Nb, Pd) films was used.

Solid non-metallic substrates were made of leucosapphire, alumina and zirconia ceramics as small thin plates 4 x 3 x 2 mm in size. One of the flat surfaces of each specimen was well polished to a roughness R_z=0.03 ÷ 0.05 μm. After polishing, all specimens were thoroughly defatted and burned in air at 1100 °C for one hour. Specimens made of leucosapphire, alumina and dioxide-zirconium ceramics were used metallized with a 150 nm Nb nanofilm and then by 1,5 μm Pd film atop serving as a soldering metal for ceramic samples joining. The quality of all applied nanofilms was controlled using a XJL-17 metallographic microscope. The specimens with deposited onto them metal films were annealed in a vacuum chamber for various periods of time (from 5 up to 20 min) and at different temperatures (from 1000 °C up to 1200 °C) in the vacuum not less than 2 x 10^{-3} Pa.

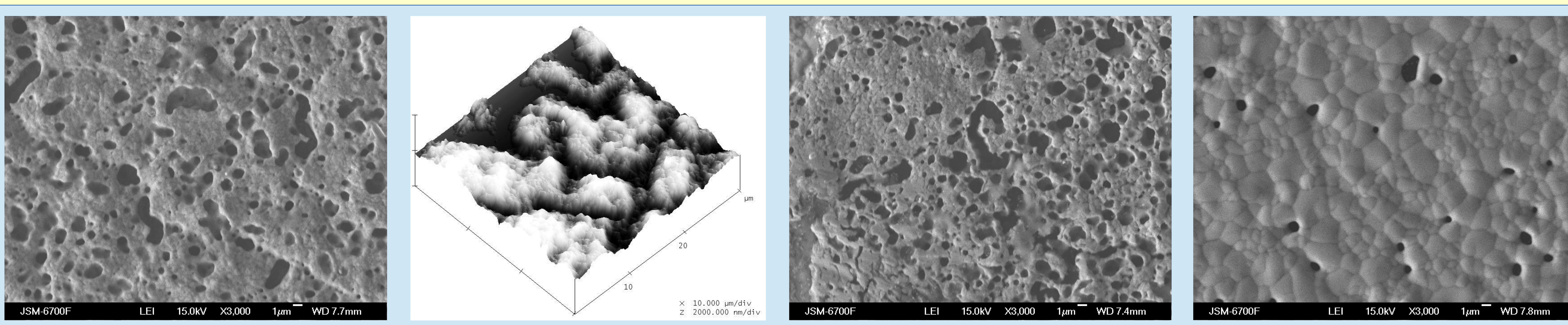
Annealed specimens were investigated using SEM and AFM microscopy with microphotographs storing. Using these microphotographs, the areas of metal islets on the surface of non-metallic samples were determined by the planimetric method. The experimental data obtained were processed in the form of graphs showing the dependence of the surface area of the samples covered with metal films on the annealing parameters (temperature, time).

Results and Discussion

The niobium metallization layer thickness at the leucosapphire, alumina, and zirconium ceramics substrates was 150 nm, whereas the palladium layer thickness reached 1.5 μm. Palladium film serving as a soldering metal for ceramic samples joining.

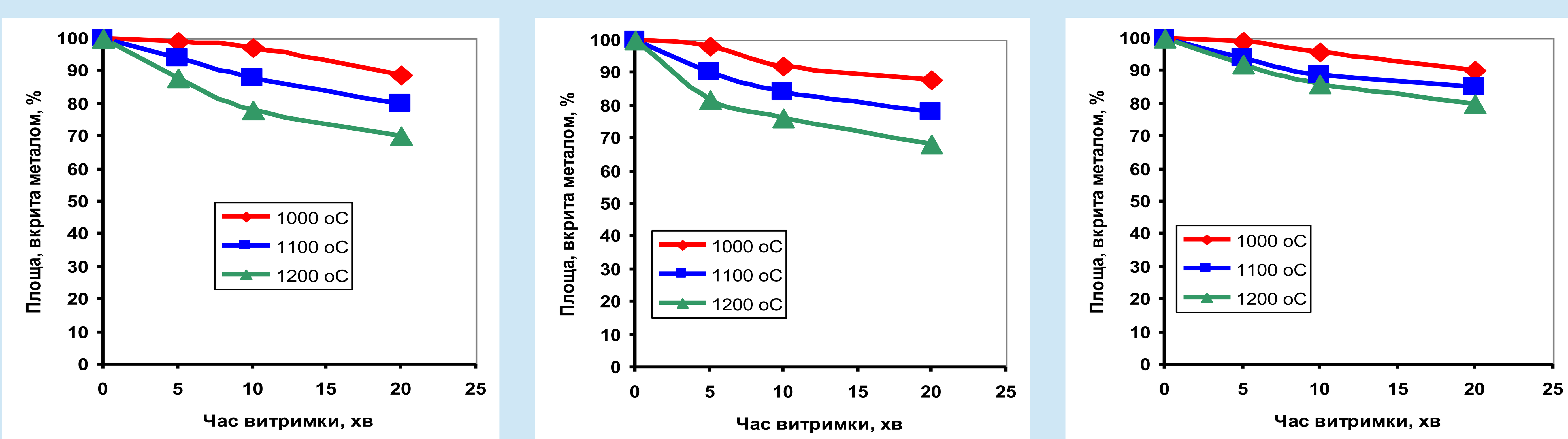
As a result of annealing at 1000 °C for 5 min, the films remained almost unchanged; after 10 min, the number of defects in the film on all oxides increased, the film began to swell and even crack; and after 20 min, the number of defects in the film on all oxides increased even more. With annealing temperature up to 1100 °C, the film underwent significant changes after 5 min of exposure; after 20 min, the number of defects on all oxides increased (Fig. 1). Figure 1, b, shows the image of the niobium-palladium double film at the alumina ceramics at high magnification of the atomic force microscope. This photo shows that a large number of defects (swelling, tears, cracks) in the film, and the height of the swelling of the film reached 500 nm.

Annealing at 1200 °C accelerated sharply the dispersion of the film which began to deteriorate significantly after 10 min of exposure; 20 min of annealing led not only to significant decay of the film, but also to the interaction of Pd with Nb, with film residues still covering 70-80% of the substrate surface area, which made it possible to use double Nb-Pd films to join oxide ceramics at temperatures up to 1200 °C. Figure 2 shows the kinetic curves of dispersion of double Nb-Pd films at the oxides surface depending on the annealing temperature and time. It follows from these dependences that these films can be used to join the oxides under investigation by pressure welding at these temperatures.



a b c d

Fig. 1. SEM (a, c, d) and AFM (b) images of double niobium-palladium film deposited onto oxide materials and further annealed at temperature 1100 °C during 20 min in vacuum: a, b – alumina ceramics; c – leucosapphire; d – zirconia ceramics



a b c

Fig. 2. Dependence of oxide materials area covered by double niobium-palladium film on annealing time at various temperatures (1000 – 1200 °C): a – alumina ceramics; b – leucosapphire; c – zirconia ceramics.

Conclusions

According to our study results, kinetic curves of the investigated thin double metallic films decomposition have been built, from which the basic process parameters (temperature and exposition time at this temperature) for brazing or pressure welding of ceramic materials can be determined. Using the data obtained, process regimes for joining by brazing and pressure welding were selected.

According to these regimes, joints' prototypes with seams up to 2 μm thick were made, which strength reached 150 MPa