

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

Gab I.I.¹, Stetsyuk T.V.¹, Fesenko O.M.², Shakhnin D.B.³

¹ Frantsevich Institute for Materials Science Problems of NAS of Ukraine,
3 Krzhyzhanovskogo st., Kyiv, 03142, Ukraine.

E-mail: tvst@ukr.net, gab@ipms.kiev.ua

² Transfer Technology Department, Institute of Physics of the NAS of Ukraine, Prospect Nauki, 46, Kyiv-03039, Ukraine.

³ University "Ukraine", 23 L'vivska st., Kyiv, 03115, Ukraine.

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 titanium, chromium, niobium and others are the most commonly used to make such coatings. Then, thus metallized ceramic parts are brazed in vacuum or an inert medium (argon, helium etc.) 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 . In addition, a thin soldering seam allows you to obtain precision welded or brazed ceramic and ceramic-metal units which can be used in microelectronics, radio engineering, microwave engineering etc.

This objective can be achieved by application onto the ceramic surfaces of double metal films, one of which is 100 - 200 nm thick and consists of an adhesion-active metal such as Ti, Cr etc., and the other is slightly thicker (2 - 3 μm) and serves as a solder, e.g. Cu, Ag etc., which will ensure joining of metallized ceramic materials during brazing or welding with fine (2 - 4 μm thick) solder seam.

The structure of such two- or multiphase coatings, determination of the optimal ratio of the thickness of each layer, processes of interaction at the phases interface, including the ceramic phase, is also an important area of research.

The study of two-layer niobium-nickel 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, Ni) nanofilms 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 $R_z=0.03 \pm 0.05 \mu\text{m}$. After polishing, all specimens were thoroughly defatted and burned in air at 1100 °C for one hour. As a metal deposited directly onto non-metallic surfaces, niobium was used. Metallized samples were then coated by nickel films serving as a solder when joining the samples. 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 900 °C up to 1100 °C) in the vacuum not less than 2×10^{-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 nanofilms 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 nickel layer thickness reached 1.5 μm .

The starting niobium-nickel films all the three oxide materials were continuous and remaining unchanged after five minutes of annealing at 1000 °C. Noticeable changes in the films appeared only after ten minutes at 1000 °C, but their integrity was also almost not violated; and only after twenty minutes of annealing at 1000 °C, the films surface changed significantly, and even cracks have appeared, which are especially noticeable for films deposited onto leucosapphire. The annealing temperature increase up to 1100 °C have naturally accelerated the changes in the films morphology occurred already after five minutes, and these changes became even more noticeable after ten minutes of annealing, but they did not violate the integrity of the films as well; and only after twenty minutes at 1100 °C, cracks and tears appeared in films (Fig. 1, a), but still the film covered about 90% of the substrate surface. The films annealing at 1200 °C have further accelerated their dispersion process which was noticeable even after five, and especially after ten, minutes of annealing.

With high magnification of an atomic force microscope, niobium-nickel double film deposited onto leucosapphire showed start of swelling and intense disintegration. The height of the swellings reached almost 200 nm. Twenty-minute annealing at 1200 °C led not only to the significant films dispersion, but also to the nickel interaction with niobium metallization layer at the oxides surface, because in the Nb-Ni system, there is a eutectic at 1175 °C (Fig. 1, b).

Figure 1, c, shows the image of the niobium-nickel double film at the dioxide-zirconium ceramics surface, and Figure 1, d, shows the same at high magnification of the atomic force microscope. These photos show that the start of the interaction between niobium and nickel inside the film led to a significant change in the primary relief of the film, and the height of the swelling of the film reached 350 nm.

Figure 2 shows the kinetic curves of dispersion of double Nb-Ni 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.

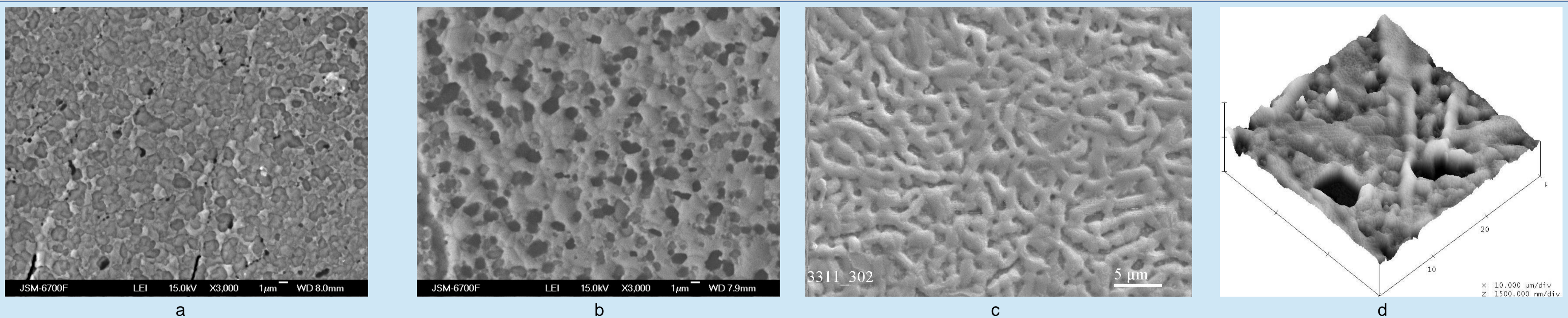


Fig. 1. SEM (a-c) and AFM (d) images of double niobium-nickel film deposited onto oxide materials and further annealed at various temperatures during 20 min in vacuum: a – leucosapphire, 1100 °C; b – leucosapphire, 1200 °C; c, d – zirconia ceramics, 1200 °C.

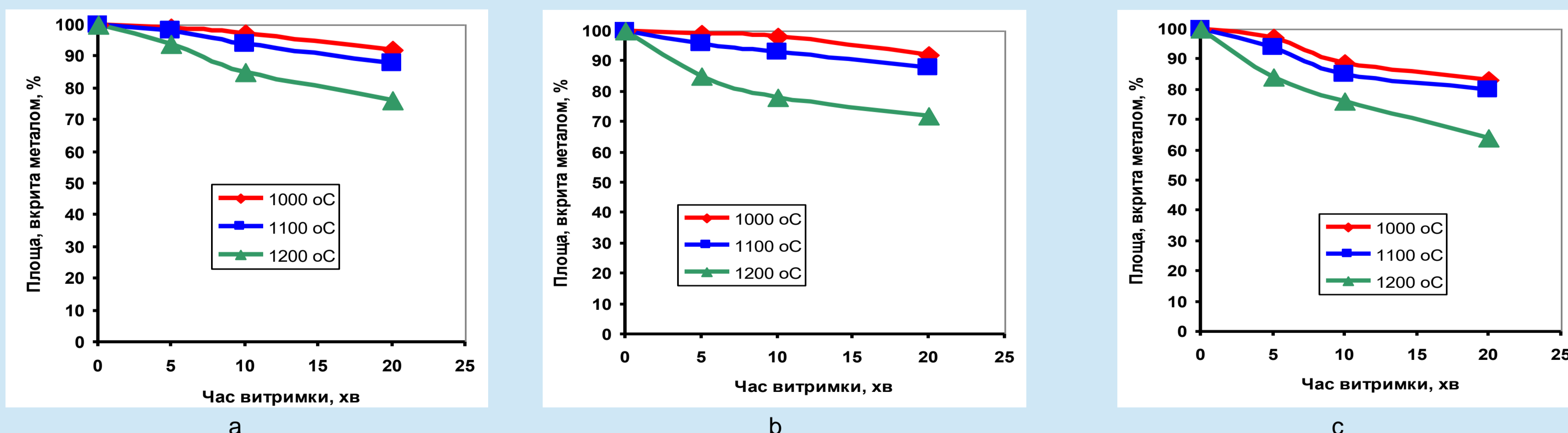


Fig. 2. Dependence of oxide materials area covered by double niobium-nickel film on annealing time at various temperatures (1000 – 1200 °C): a – alumina ceramics; b – leucosapphire; c – zirconia ceramics.

Conclusions

According to the study results, the dispersion during annealing in vacuum of double niobium-nickel films at all three oxides surfaces is practically the same, and all these films have good adhesion to the oxide substrates covering almost 90% of the substrate area after twenty minutes of annealing at 1050 °C. Even after annealing at 1100 °C, nickel film fragments are covering about 80% of the substrate area. This makes it possible to recommend these double films for further joining of all three studied oxide substrates by pressure welding or brazing.

The kinetic curves of dispersion during annealing of double niobium-nickel films on leucosapphire, alumina and zirconia ceramics were plotted, using which it is possible to determine the basic technological parameters (temperature and exposition time at this temperature) of the processes of joining of ceramic materials by brazing or pressure welding.

According to the data obtained, it is also possible to join oxide materials using dual niobium-nickel films by brazing at a temperature 1100 °C. Using both of these methods, brazed ceramics joints with a seam thickness not exceeding 5 microns were obtained having shear strength 150 MPa.