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**Fabrication of
Niobium Fluidic Circuit Components
by Diffusion Bonding**

by

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FABRICATION OF NIOBIUM FLUIDIC CIRCUIT
COMPONENTS BY DIFFUSION BONDING

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ABSTRACT

A technique has been developed for fabricating fluid circuit components by diffusion bonding chemically milled niobium foil laminates. Sections of 0.002-in.-thick niobium foil were chemically milled to provide the correct channel pattern. The foil sections were then vapor coated with titanium and stacked in a diffusion bonding fixture. The laminated assembly was vacuum diffusion bonded at 1700°C by using an axial load of 300 to 500 psi, based upon the total assembly area.

I. INTRODUCTION

Group K-4 of the Los Alamos Scientific Laboratory (LASL) has been working with the Marquardt Corporation of Denver to design and test fluidic circuit assemblies to be used for sensing characteristics of hot flowing gases. Although Marquardt has been designing and testing fluidic circuits for room-temperature applications, they have neither fabricating nor testing facilities to build and evaluate fluidic systems for use in high-temperature environments.

LASL Group CMB-6 has fabricated a limited number of fluidic devices to be evaluated by Group K-4 in the UHTREX (Ultra-High-Temperature Reactor Experiment) reactor. These devices have been designed for use at 1300°C in a methane gas environment.

II. MATERIALS AND EQUIPMENT

A. Materials

A 0.002-in.-thick commercial niobium foil was used for the body of the fluidic circuit. The top and bottom plates of the assembly were made from a 1/16-in.-thick commercial niobium plate. The tubing was drawn to 0.150-in. o.d. by 0.060-in. i.d. from commercial 1/4-in. o.d. by 0.045-in. wall Nb-12Zr tubing.

B. Equipment

The initial diffusion bonding work was done

with a resistance-heated Brew vacuum furnace that utilized a tungsten heating element. Axial pressure was applied by dead-weight loading. Later work was done in a vacuum hot-press unit built by the Powder Metallurgy Section of Group CMB-6. This unit was heated by induction from a 10-kHz motor generator unit. The axial load was applied to the assemblies by a hydraulic ram system.

C. Fixturing

The initial fixturing for diffusion bonding was made from Al_2O_3 ceramic plates that were ultrasonically drilled to the desired configuration. The ceramic plates were tied together by tungsten pins, with the chemically milled niobium plates between the ceramic fixturing plates as shown in Fig. 1. The Al_2O_3 was very reactive with niobium at temperatures in excess of 1700°C. To alleviate this problem, TZM molybdenum was used in place of the Al_2O_3 plates. The faces of the TZM plates were coated with a thin thorium slurry to prevent diffusion between the TZM and the niobium. These fixturing plates were easier to machine than the Al_2O_3 parts and have done the job very well.

III. COMPONENT PREPARATION

A. Chemical Milling

The 0.002-in.-thick niobium sheet was treated with Eastman Kodak Metal Etch Resist (MER) and an

Fig. 1. Fluidic circuit diffusion bonding fixture.

aluminum stearate additive "D," as recommended for glass or ceramic etching in the Eastman Kodak 1966 Applications Data booklet.

The treated and exposed sheet was anodically milled in a 48% HF solution at 2 V. The 0.002-in.-thick sheet was etched from both sides simultaneously for 20 to 30 sec.

B. Surface Preparation

The chemically milled components were cleaned thoroughly and then a coating of approximately 0.000020 in. of titanium was vapor-deposited on both sides of the foil components. Both sides were treated to avoid having two uncoated foils in contact in the stacked assembly.

The treated foils were then placed on clean soft paper and stored in a desiccator. Additional cleaning of the treated components was not attempted because further cleaning of the as-deposited titanium surface might contaminate or remove the vapor-deposited coating.

IV. DIFFUSION BONDING DEVELOPMENT

The design of the first diffusion-bonded assembly was such that the top and bottom cover plates were made from 0.002-in.-thick foil laminates as shown in Fig. 2. The foil sections were all stacked (Fig. 2) and fixtured between two Al_2O_3 ceramic plates. Tungsten pins were located in the four corner holes to align the various components.

Diffusion bonding was accomplished by dead-weight loading the assembled components to approximately 50 psi and holding at 1600°C for 1 h in a vacuum of $< 1 \times 10^{-4}$ Torr.

Fig. 2. Exploded view of laminated fluidic circuit assembly.

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Fig. 3. Cross section of diffusion bonded fluidic circuit assembly showing lack of bonding between the layers located above or below gas channels.

Figure 3 shows a photomicrograph of a section cut through this first assembly. Two problems became immediately evident. First, there was obvious lack of bonding in the top and bottom plate sections immediately over the channel configurations. The reason, of course, is that in these unsupported areas no normal load can be applied to maintain intimate contact between the foils. Second, the misalignment between adjacent foils resulted in irregularly shaped

walls. A second assembly, for which the foils had already been prepared, was fixtured and diffusion-bonded in much the same manner as the first assembly. The main difference in procedure was that the top, bottom, and center sections of the assembly were bonded as separate components, recoated with vapor-deposited titanium, and bonded into a single assembly. This procedure eliminated the problem of inadequate bonding in the top and bottom sections; however, the alignment was still rather poor. The alignment problem was traced to inconsistencies in the original drawings from which the films were made for chemical milling. A photograph of the bonded channel section of this assembly is shown in Fig. 4.

Nb-12Zr tubes were brazed into the inlet and outlet ports of the bonded assembly using a 65V-35Nb brazing alloy. Brazing was done at 1850°C in a vacuum of $< 1 \times 10^{-4}$ Torr.

On the basis of this first work, some design changes were made before the next units were fabricated. The channel configuration of the center section was changed to accommodate the fluid amplification

requirements. The top and bottom plates were made from 1/16-in.-thick niobium stock, rather than the laminated structure, to avoid the intricate two-stage procedure required with the laminated design. The diffusion bonding was done in two stages to check the alignment between the foils in the center section.

The chemically milled foils, as well as the top and bottom plates, were titanium coated as in the previous procedure. The bottom plate and the foil components were fixtured between two Al_2O_3 ceramic plates, dead-weight loaded to approximately 19 psi, and held at 1700°C for 1 h in a vacuum of $< 1 \times 10^{-4}$ Torr. The loading in this case was reduced because of an increase in the assembly size and the load-bearing limitations of the furnace. The temperature was increased to 1700°C to compensate for the decreased loading. The alignment on this assembly was better than for the previous two assemblies; however, the fabrication of this assembly brought other problems to light. The assembly is shown in Fig. 5.

Although the original bonded assembly (bottom to core) was leaktight, the joint opened up when the joint between the top plate and the core was made. Attempts to repair this leak by bonding at higher temperature (that is, 1800°C) resulted in a reaction

Fig. 4. Diffusion bonded channel section consisting of 12 layers of 0.002-in.-thick niobium foil.

Fig. 5. The diffusion bonded channel and bottom sections of the second fluidic amplifier design.

Fig. 6. Plot of the unbonded areas which were detected by ultrasonic inspection. The cross-hatched areas are the areas which showed lack of bonding.

between the niobium assembly and the Al_2O_3 fixturing. Later investigation showed that 1700°C is the maximum temperature attainable without inducing this reaction.

Another problem in the fabrication of this assembly was the excessive braze alloy flow when the Nb-12Zr tubing was brazed to the inlet and outlet ports. The excess alloy flowed into one of the channels and blocked the throat. A second assembly was made by using essentially the same approach except that the entire assembly was made in one diffusion bonding step.

The bonded assembly was ultrasonically inspected to determine the distribution of bonding (or the lack of bonding) between the niobium plates. A trace of the ultrasonic results is shown in Fig. 6 with an outline of the channel and aligning hole configuration superimposed over the ultrasonic trace. The void areas (crosshatched) are not necessarily interconnecting because the composite assembly was made up of approximately 12 layers of 0.002-in.-thick niobium foil. It would be possible to make a three-dimensional ultrasonic trace to determine to what extent the unbonded areas are interconnected.

Niobium tubes were then brazed to the assembly with a very tight-fitting joint to prevent excessive

alloy flow. The braze joints were made at 1850°C in vacuum by using 65V-35Nb brazing alloy. After brazing in the tubes, several small leaks were discovered around the perimeter of the assembly. These leaks may have been opened by the brazing operation or may have been present after diffusion bonding because the assembly had not been leak checked at that time. These leaks were patched by electron beam welding around the perimeter of the assembly.

The fabrication of these assemblies proved several interesting facts.

1. Sufficient axial pressure is imperative in this process and 50 psi may be considered the absolute minimum required.

2. The Al_2O_3 ceramic platens being used for fixturing should be replaced with some other material to reduce distortion and to eliminate the Nb- Al_2O_3 reaction which occurs at temperatures greater than 1700°C .

3. The tight tube joint appears to satisfactorily limit braze alloy flow during brazing of the inlet and outlet tubes.

The third type of assembly to be fabricated was a fluidic oscillator of a design slightly more complex than that of the previous assemblies. The design of this assembly required that the core be made from two separate chemically milled and carefully aligned shapes. The core components were chemically milled by the standard procedure. The core components, as well as the top and bottom plates, were vapor-coated with titanium. Only the core components and the bottom plate were fixtured together for the initial diffusion bond, although the complete assembly could have been bonded in one operation. The parts were stacked and pinned in a TZM alloy fixture coated with ThO_2 to prevent bonding to the fixture. The ThO_2 coating was applied by brushing on a thin layer of a ThO_2 alcohol slurry and then allowed to dry.

Diffusion bonding was accomplished by canning the assembly in a 0.010-in.-thick welded tantalum can and holding at 1700°C in vacuum for 1 h under an axial load of 300 psi. The bonded assembly (Fig. 7) was stripped from the TZM fixture with no difficulty and was leaktight. The bonded assembly was recoated with titanium and bonded to the top plate at 1700°C and an axial load of 500 psi. An induction-heated

Fig. 7. Diffusion bonded channel and bottom sections of the fluidic oscillator assembly.

vacuum hot press was used for both bonding operations. The reason for canning the assembly in each case was to protect it from the carbiding action of the graphite susceptor being used.

The Nb-1Zr tubes were then brazed to the bonded assembly by using pure titanium alloy. Brazing was done at 1800°C in a vacuum of $< 1 \times 10^{-4}$ Torr. The fabricated assembly was leaktight to a helium leak rate of $< 1 \times 10^{-8}$ cm³/sec.

V. CONCLUSIONS

Satisfactory leaktight fluidic circuit assemblies may be fabricated by vacuum diffusion bonding 0.002-in.-thick chemically milled niobium foils. Coating the foils with a thin layer of vapor deposited titanium helps to promote diffusion bonding between the foils. An axial load of 300 to 500 psi and a bonding temperature of 1700°C is adequate to achieve good bonding under the conditions used for this work.

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