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# METHOD OF BONDING ALUMINUM MEMBERS

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12 Claims. (Cl. 29—487)

Our invention relates to a method of joining aluminum members, and more particularly to a diffusion bonding method of joining aluminum members.

Aluminum is widely employed as a cladding material for nuclear reactor fuel elements because of its low thermal neutron absorption cross section and its generally satisfactory mechanical properties. However, the mechanical strength of aluminum is deficient for application in high temperature reactors. Improved strength characteristics are particularly required when the fuel is in a form, such as $UO_2$, which will not bear the load of the fuel assembly. Aluminum alloys having better high temperature strength characteristics than aluminum and satisfactory neutron cross sections are, therefore, being adapted for use in nuclear reactors. Prominent among the aluminum alloys being used for high temperature reactor application, and for many other applications where a greater strength than that of aluminum metal is required, is the class of commercially-available aluminum alloys consisting of about 4–12 wt. percent aluminum oxide ($Al_2O_3$) dispersed in an aluminum matrix. Unless otherwise stated, the word aluminum, as used in this specification and in the appended claims, is defined to generically embrace both aluminum metal and aluminum alloys.

The bonding of aluminum members in strong, leak-tight fashion is a severe requirement in fuel element fabrication. It is essential that closure members be securely bonded to prevent escape of fission products to the interior of the reactor or the environment, and to protect the fuel against corrosive or erosive attack by coolants. For example, a fuel element may consist of a plurality of tubular aluminum alloy members containing $UO_2$, and this assembly must be closed with a leak-tight and strong end cap. Heretofore, the bonding of aluminum metal to aluminum alloy or aluminum alloy to aluminum alloy members has been accomplished by high pressure, solid state bonding, for instance, at 30–80,000 p.s.i. This procedure is time consuming, may result in warpage of aluminum members, and is not adapted to a production process where thousands of fuel rod end closures may be required for a single core loading. Welding techniques tend to destroy the elevated temperature properties of the aluminum-aluminum oxide alloys.

Accordingly, the principal object of our present invention is to provide a method of joining aluminum members.

Another object is to provide such a method in which the resulting joint approaches the strength of the parent material.

Another object is to provide a eutectic diffusion bonding method for joining aluminum alloys.

Another object is to provide a method of forming reactor fuel rod end closures which will remain gas leak-tight at elevated temperatures.

Still another object is to provide such a method which can be rapidly performed using moderate pressures.

A further object is to provide such a method in which the boundary layer between the joined members is extremely thin and free of brittle intermetallic compounds.

A still further object is to provide such a method which is readily suited as a production process.

Further objects and advantages of our invention will become apparent from the following detailed description and the appended claims.

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In accordance with our present invention we have provided a method of joining aluminum members, which comprises providing on at least one mating surface a thin layer of a bonding metal selected from the group consisting of silver, copper, gold, tin, and zinc, bringing the mating surfaces into intimate contact, and heating the resulting assembly at a temperature between the eutectic formation temperature and the melting point of the aluminum members, until said members are joined.

By the use of our method, aluminum members may be readily joined in a bond which approaches the strength of the parent material and which does not diminish in strength at elevated temperatures. End closures on fuel rods have been effected which are gas tight and remain unaffected by temperature cycling at elevated temperatures over a prolonged period of time. The bond formation appears to take place by the formation of a eutectic between the aluminum surfaces and the bonding metal, which after euectic formation diffuses away from the interface to leave an extremely fine juncture line.

In the practice of our invention a very thin layer of at least one bonding metal selected from the group consisting of silver, copper, gold, tin, and zinc, is applied to at least one of the surfaces to be joined, but preferably only to one surface. We find that it is desirable to apply only a very thin layer of the bonding metal. For instance, the film is desirably not thicker than about $10^{-3}$ inches, while a film of about $10^{-4}$ to $10^{-5}$ inches is preferred. While thicker films may be operably employed, such films may result in the formation of intermetallic compounds at the interface which are brittle and hence of decreased strength. The film may be applied on to the surfaces by deposition methods known to the art for the particular metals. For instance, films may be deposited by such convenient, controllable means as vacuum and electrolytic deposition.

After at least one of the surfaces to be joined has been first cleaned by such conventional means as washings with organic reagents and alkaline and acid rinses, following which the bonding film is applied, the surfaces are brought together and maintained in intimate contact. This may be done by use of such conventional means as a collet or other holding means. Only sufficient pressure need be applied to the holding means to maintain the intimate contact during the heating period.

The temperature at which bond formation takes place is dependent upon the particular eutectic bonding agent employed. The temperature will thus range between about 468° F. for eutectic formation between tin and the aluminum member, 720° F. for zinc, 980° F. for gold, through about 1012° F. for copper to about 1030° F. for silver. The maximum temperature will be a function of the melting or distortion points of the aluminum, this being about 1100° F. for aluminum metal and about 1200° F. for the above aluminum alloy. It is desirable to exceed the eutectic formation temperature for the particular eutectic material by about 30–60° F., in order to promote diffusion of the eutectic into the parent material and formation of a thin interface which adds considerably to the quality of the resulting bond. For instance, a highly satisfactory temperature for silver eutectic bonding is about 1060–1090° F., while a temperature of about 1080° F. is preferred. The heating step may be satisfactorily conducted in either air or in an inert gas atmosphere. Maintaining intimate contact between the surfaces being joined is sufficient to prevent oxidation at the interface as the temperature is being raised, and air heating, by such means as an induction coil, is very satisfactory as a production method.

We find that eutectic formation and diffusion into the parent material to form a strong bond takes place at the above-indicated temperatures relatively rapidly, and this permits aluminum members to be joined in a short period of time in a convenient manner. Thus, the members are heated at the above temperatures for a period of about 1½ to 10 minutes, while a period of 2½ to 5 minutes is preferred. After the heating step is concluded, the now joined members are permitted to cool to the ambient temperature, preferably while still held by the holding means. This is typically for a period of about 30 minutes. We find that cooling while still maintaining the members in the holding means, while not critical, further promotes diffusion of the eutectic into the parent material.

The following examples are offered to illustrate our invention in greater detail.

*Example 1*

This example relates to forming a fuel rod end closure on a tube having an I.D. of 300±.003 and a wall thickness of .030 inch. The tube was fabricated of an aluminum—4–8 wt. percent aluminum oxide alloy having the Alcoa commercial designation APM (M–257). The end plug was machined of the same material from a ⅜ inch diameter, swaged rod. The diameter of the portion of the end plug to be inserted in the tube was 0.297±.002 inch.

The specimens were cleaned by degreasing with trichloroethylene liquid and vapor followed by cleaning with an aqueous caustic solution, rinsing with water, treatment with nitric acid (50% by volume), and final water rinsing and drying. The end plug was electroplated with silver from a plating bath comprising silver cyanide, 6 oz. per gallon, and potassium cyanide, 12 oz. per gallon. The sample was electroplated for 30 seconds at 40 amps. per sq. ft., resulting in the deposition of about $10^{-5}$ to $10^{-4}$ inches of silver.

The end plug was put in the tube and placed under pressure in a six-segment collet having an open I.D. of .375 inch. The assembly was then heated by induction heating to a temperature of 1060° F. –1100° F. Pressure was maintained by tightening the collet die at intervals of approximately 300° F. at 930° F., and after reaching temperature, in order to maintain intimate contact upon expansion due to heating. Temperature was maintained for about 2½ minutes, after which the specimen was permitted to cool in the die for about 20 minutes before removal. The tube was similarly bonded on the other end. Both ends were found to be helium leak-tight by drilling one plug through for testing. After checking, this hole was closed by hot knifing (1200° F.) using a copper plated aluminum weld rod inserted in the hole to facilitate closure.

The tube was then subjected to external pressure in an autoclave at 800° F. When the pressure reached 2300 p.s.i., the tube failed but the end closure was still intact and proved helium leak-tight.

The tubes were also subjected to pressure cycling tests at 750° F. One specimen was given 50 cycles from 0–1000 p.s.i.g. and remained helium leak-tight. Another tube failed through the cladding but the end closure remained intact. Specimens also remained leak-tight after 90 days at 1000° F. in an air furnace and after 1,000 hours in a terphenyl solution at 750° F.

*Example 2*

The same as Example 1 except that copper was used as the eutectic bonding agent. The copper was electroplated on to the aluminum end plug from an aqueous plating bath containing copper cyanide, 6–8 oz. per gallon, free KCN, 1.2–1.5 oz. per gallon, and KOH, 3–4 oz. per gallon. About $10^{-5}$–$10^{-4}$ inches of copper was deposited on the sample in 30 seconds at 50 amps. per sq. ft. current density.

Testing of the resulting assembly using the methods of Example 1 showed the resulting end closure to be helium leak-tight, and no leaks were revealed after the pressure cycling, temperature cycling, and organic soaking tests.



*Example 3*

The same as Example 1 except as indicated. In this example the aluminum-aluminum oxide tube was closed with an aluminum metal end plug. The closure was made utilizing a 0.250 inch O.D. tube having a 0.020 inch wall. The end plug had a shoulder 0.250 inch in diameter x 0.050 inch thickness for ease of placement. The area plated was 0.210 inch in diameter x 0.500 inch long.

The aluminum metal end plug was plated by vacuum deposition with silver to a thickness ranging from $9\times10^{-5}$ to $1.8\times10^{-4}$ inches. The members were cleaned prior to plating as in Example 1. The components were then assembled and placed in a collet die which was tightened to apply pressure to the assembly. The die assembly containing the end closure assembly was then centered in an induction coil and heated for 2½ minutes at a temperature of about 1060–1100° F. The die assembly was tightened every 30 seconds to maintain pressure while heating. The closure was allowed to remain in the die assembly until cool, about 25 minutes. After removing the closure from the die, it was checked for helium leak-tightness at room temperature and found to be helium leak-tight.

The closure was subjected to 175 thermal cycles which ranged from 78° to 900° F. and required 30 minutes per cycle. The specimen still remained helium leak-tight.

The closure was then subjected to gas pressure after soaking at 750° F. for 30 minutes. The tube failed at 2300 p.s.i. with the closure still intact. This specimen was then submitted to metallography which showed evidence of total interdiffusion at the bond interface and the bond to be of unquestionable integrity.

The foregoing examples are merely illustrative rather than restrictive of our invention. Variations may be made in assembly techniques by those skilled in the art within the scope of our invention. Therefore, our invention should be understood to be limited only as is indicated in the appended claims.

We claim:

1. A method of joining aluminum members which comprises providing a thin layer of less than $10^{-3}$ inches of silver on at least one of the mating surfaces of the aluminum members, contacting said mating surfaces and, while maintaining said members in intimate contact, heating the resulting assembly at a temperature between about 1030° F. and below the melting point of said aluminum members to form a eutectic of silver and aluminum at the interface of said members, and maintaining said temperature for a sufficient time to form a direct bond between the joined members by diffusion of the formed eutectic away from the interface into said members.

2. The method of claim 1 wherein said heating is conducted at a temperature of about 1060°–1090° F.

3. A method of joining aluminum members which comprises providing a layer of silver of about $10^{-4}$ to $10^{-5}$ inches on one of the mating surfaces of said members, contacting said mating surfaces in intimate contact, and heating the resulting assembly at a temperature of about 1060° F. to 1090° F. for a period of about 1½ to 10 minutes to form a eutectic of silver and aluminum at the interface of said members and to diffuse the formed eutectic away from the interface into said members, thereby to directly bond said members together.

4. The method of claim 3 wherein said members are heated for a period of about 2½ to 5 minutes in air.

5. The method of claim 3 wherein said aluminum members are of an aluminum alloy consisting essentially of about 4–12 weight percent $Al_2O_3$ and the remainder aluminum.

6. The method of claim 3 wherein the resulting assembly subsequent to heating is permitted to air cool to the ambient atmospheric temperature while said intimate contact is being maintained.

7. A method of joining aluminum members which com-

prises providing a layer of less than $10^{-3}$ inches thickness of a metal selected from the group consisting of silver, copper, gold, tin, and zinc on at least one mating surface, bringing the mating surfaces of said members together and, while maintaining said members in intimate contact, heating the resulting assembly at a temperature between the eutectic formation temperature of said bonding metal and aluminum and the melting point of said members to form a eutectic of said bonding metal and aluminum at the interface of said members, and maintaining said temperature for a sufficient time to form a direct bond between the joined surfaces by diffusion of the formed eutectic away from the interface into said members.

8. The method of claim 7 wherein said temperature range is between about 500°–1200° F.

9. The method of claim 7 wherein said assembly is heated for a period of about 1½ to 10 minutes.



10. The method of claim 7 wherein said assembly is heated in air.

11. The method of claim 7 wherein the film of said bonding material is provided on the mating surface of said aluminum member by electrolytic deposition.

12. The method of claim 7 wherein at least one of said aluminum members is an aluminum alloy consisting essentially of about 4–12 weight percent $Al_2O_3$ and the remainder aluminum metal.


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JOHN F. CAMPBELL, *Primary Examiner.*

WHITMORE A. WILTZ, *Examiner.*

UNITED STATES PATENT OFFICE

# CERTIFICATE OF CORRECTION

Patent No. 3,180,022 April 27, 1965

Bill N. Briggs et al.

It is hereby certified that error appears in the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 2, line 15, for "cylcling" read -- cycling --; column 3, line 17, for "300±.003" read -- .300±.003 --; line 40, for "300$^{o}$ F." read -- 300$^{o}$ F., --; column 6, under "UNITED STATES PATENTS" after line 15, add the following:

| | | | |
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| 2,798,843 | 7/57 | Slomin et al. | 29-504 UXR |
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| 540,961 | 11/41 | Great Britain | 29-501 |

Signed and sealed this 3rd day of August 1965.

(SEAL)
Attest:

ERNEST W. SWIDER
Attesting Officer

EDWARD J. BRENNER
Commissioner of Patents