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FABRICATION OF HEATER TUBE FUSES

By

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TABLE OF CONTENTS

	<u>Page No.</u>
ABSTRACT.....	4
I. INTRODUCTION.....	5
II. MATERIALS USED.....	7
III. INVESTIGATION OF FABRICATION METHODS....	8
A. Compacting.....	8
B. Sintering and Sinter-Shrink Bonding.	9
C. Coining and Assembly.....	11
D. Bonding.....	11
IV. METHODS UTILIZED FOR FABRICATING TEST SAMPLES.....	15
V. INVESTIGATION OF METHODS OF BRAZING HEATER TUBES TO PAD AND BLOCK.....	15
VI. CONCLUSIONS.....	17

ABSTRACT

Various methods were investigated to produce a heater tube fuse. Utilizing powder metallurgy techniques, fuel tubes with an I.D. of 0.045 inch, O.D. of 0.075 inch and length of 11/16 inch, containing various compositions of copper + UO_2 , were fabricated.

Fabrication methods consisted of compacting the tubes in quarter-length units, sintering under optimum conditions, coining on a tin-plated inner tube with tin washers between units, and then heat treating to obtain a proper bond. By these techniques, 33 units containing various compositions of copper + $U^{235}O_2$ were fabricated and shipped to Atomics International for testing.

In addition, work was performed on the brazing facet of the trigger element assembly, joining the copper + UO_2 fuel tube to a type 347 stainless steel thermal resistance pad and block.

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I. INTRODUCTION

In a reactor, an "inherently reliable" shut-down device which does not depend upon electrical and/or mechanical motivation to shut down the pile would be desirable as a final safety measure. Activation should occur as a result of the flux or heating effect of the runaway reactor, but design must be such as to permit operation under normal fluxes.

Atomics International, in developing such an "inherently safe" reactor shut-down device, designed a heater tube fuse as part of the triggering mechanism. This device is composed of an outer fuel tube containing various amounts of $U^{235}O_2$ uniformly distributed in a metallic matrix and metallurgically bonded to an inner tube. The inner tube contains a plug of low melting solder which, when overheated from the reactivity of the fuel tube, melts and allows a gas poison under pressure to rupture the container and poison the reactor⁽¹⁻¹²⁾ (Fig. 1).

Sylvania Electric Products Inc., in April 1954, contracted to produce 33 of these heater tube fuses
with the following requirements:

1. The fuse must be gas-tight with a maximum permissible loss of one percent over a six-month period.
2. The fuse must withstand an internal pressure of 2000 psi at temperatures up to 150°C.
3. The fuel tube matrix and the inner tube are to be made of copper, but nickel or iron can possibly be substituted.
4. The fuel tube is to be 11/16 $\begin{smallmatrix} +0.0313 \\ -0.0000 \end{smallmatrix}$ inch long with an I.D. of 0.045 ± 0.001 inch and an O.D. of 0.075 ± 0.001 inch.
5. The inner tube is to have an I.D. of 0.035 ± 0.001 inch and an O.D. of 0.045 ± 0.001 inch. It is to extend past the fuel tube at one end.
6. The amount of fuel and the number of heater tube fuses required of each composition are as follows:

<u>Number Required</u>	<u>Milligrams of Enriched UO_2 per Centimeter of Length (Nominal en- richment)</u>
9	37.5
9	22.0
3	12.5
12	2.0

6

Fig. 1 HEATER TUBE FUSE

7. Tubes must withstand at least ten percent burn-up before replacement is required.
8. A good heat transfer bond is required between fuel tube and inner tube.
9. Fuses must possess adequate ductility for flattening, and high density for thermal conductivity and favorable brazing properties.
10. Ends of the fuel tubes must either be closed or sufficiently dense to be impervious to gas flow.
11. If the fuel tube is comprised of more than one piece, the separation between pieces must be extremely small.

II. MATERIALS USED

A -325 mesh copper powder received from Charles Hardy was used in all of the investigations. This powder, which is made by electrolytic deposition, is irregular in shape and has an average particle size of between 20 and 30 microns.

The $U^{238}O_2$ powder used in developmental work was received from Mallinckrodt. This powder was hydrogen fired for three hours at $1300^\circ C$ and then hand-ground through -325 mesh. It is irregular in shape and has an average particle size of between two and three microns.

The original batch of enriched material was received from Atomic International in the form of U_3O_8 . This material was processed in the same manner as the natural UO_2 used in the developmental investigations i.e. by hydrogen reduction for three hours at $1300^\circ C$. In attempting to use the enriched material to fabricate the heater tubes, however, vast differences were encountered. The UO_2 appeared to agglomerate during firing, and the $Cu + UO_2$ mixture compacted poorly. Sintered fuel tubes made with this oxide mixture showed radial cracking and poor ductility. Electron microscopy studies of this material showed vast differences between it and the natural UO_2 used. The enriched material appeared to have large particles and extremely fine particles which tended to flocculate, as compared to a normal size distribution in the natural UO_2 . Differences were noted in the particle shape of the two oxides in addition to the presence of small quantities of unidentified foreign material noted in the enriched UO_2 .

A second batch of enriched material was received from Oak Ridge National Laboratory in the form of UO_2 . This material was made from calcined ammonium diuranate whereas the natural UO_2 was made from decomposed and reduced uranyl nitrate. After this batch of

enriched material was treated in the aforementioned manner, it was used in the fabrication of the heater tube fuses, and no difficulties were encountered.

III. INVESTIGATION OF FABRICATION METHODS

A. Compacting

1. Full Length Tubes

Because of the ease in pressing and non-magnetic nature, copper was selected as the matrix material. Initial work was done with a mixture of copper plus highest UO_2 composition, plus lubricant (either mixed in the powder or coated on the die). It was felt that if successful compacts could be made with the highest UO_2 composition, little difficulty could be expected with the other compositions.

Compacting fuel tubes to the specified length-to-diameter ratio presented considerable difficulties. Friction of the metal powder compact against die wall and core rod caused great difficulty in ejection of the compact, even with lubrication. Furthermore, friction that occurred in the pressing of a piece with large length-to-diameter ratio caused a density gradient along the length of the pressed compact even when double-action type pressing was employed. It is the low density area which makes the compact extremely fragile and difficult to handle. This density gradient becomes quite apparent after sintering, with large variations in shrinkage and thus variations in both the inner and outer diameters of the tube.

Compacts of the desired length were successfully pressed at pressures of 20 to 30 tsi and strung on the inner tube in the as-pressed or pre-sintered condition and subsequently sintered. Attempts at pressing directly on the inner tube in place of the core rod proved unsuccessful due to crimping of the tube during compacting, and subsequent cracking of the compact during ejection.

Although tubes of the correct length were made successfully, difficulties associated with pressing and sintering (to be discussed subsequently) led to fabrication of these tubes in half-length and quarter-length units.

2. Half-and Quarter-Length Units

Compacting of the fuel tubes in half-and quarter-length units which could then be assembled on the inner tube, was found to be quite successful and applicable to high volume production. Virtually no difficulties in pressing or ejection of these compacts were experienced. Too, be-

cause of the reduced friction in ejection of these shorter lengths, it was possible to compact them to higher densities at pressures of 35 tsi.

Shrinkage variations occurring during the sintering of the high and low UO_2 compositions, necessitated the use of a larger die cavity and core rod, when working with the lower UO_2 compositions, to keep the necessary dimensional tolerances.

B. Sintering and Sinter-Shrink Bonding

Correct sintering conditions for the $\text{Cu} + \text{UO}_2$ mixtures were investigated. Sintering for 16 hours at 1050°C in a wet hydrogen atmosphere resulted in optimum sintered densities for all of the $\text{Cu} + \text{UO}_2$ compositions.

One of the necessary requirements for the heater tubes is that of a good heat transfer bond between fuel tube and inner tube. An attempt was made to obtain this bond with the sinter-shrink technique. For this method, pre-sintered fuel tubes (heated in hydrogen at 900°C for 10 minutes to impart strength for handling) were strung on the inner tube and the assembled unit sintered. During sintering, the outer tube shrinks down tightly onto the inner tube and diffusion takes place at the interface. This method was employed using a copper inner tube, nickel inner tube and a copper inner tube which had been electroless nickel plated.

With the nickel inner tube, no diffusion appeared to take place and a wide boundary existed between tubes. The electroless nickel plated copper tube showed complete diffusion of the nickel plate into the porous compact during the sintering and almost no bond was present. Apparently, optimum sintering conditions for the $\text{Cu} + \text{UO}_2$ fuel tube were incorrect to obtain a good diffusion bond between the electroless nickel and copper. Thus, this latter idea was discarded as a possibility for sinter-shrink bonding for two reasons. First, the Ni-Cu alloy obtained at the interface has poor thermal conductivity. Secondly, it would necessitate the use of a fully sintered fuel tube to control diffusion of the electroless nickel into the compact.

Use of an inner copper tube was then investigated. This method appeared to have promise, as metallographic examination showed a diffusion bond was obtained at the interface (Fig. 2). However, restraining the shrinkage of the fuel tube in one direction (onto the inner tube) led to excessive shrinkage in the other direction. This caused separation of the individual units in the multi-unit fuel tube and radial cracking of the fuel tube itself. It was felt that obtaining a balance in dimensions necessary to insure a sinter-bond without cracking and separation, would be too difficult with the

Fig. 2(a)
Sinter-shrink bonding of
Cu + UO_2 fuel tube to
inner copper tube. Heat
treatment 1050°C for 16
hours in wet hydrogen.
Inner rod necessary to
keep concentricity dur-
ing metallographic mount-
ing.

Plate No. 12085

50X

Fig. 2(b)
Sinter-shrink bonding of Cu +
 UO_2 fuel tube to inner copper
tube. Heat treatment 1050°C
for 16 hours in wet hydrogen.

Plate No. 12086

500X

four different Cu + UO₂ compositions used (and thus four different degrees of shrinkage) and this method was discarded.

C. Coining and Assembly

Fuel tubes made with the sinter-shrink technique and even ordinary sintered tubes were of insufficient density to yield the required ductility for flattening and had poor wettability for brazing purposes.

Elimination of a majority of the shortcomings listed above was accomplished by coining of the half-or quarter-length units following the pressing and sintering operations.

Three variations in assembly were employed. The first, to coin these units at 35 tsi directly onto the inner tube with the aid of a mandrel, to prevent distortion and attempt to obtain intimate end-to-end contact. The second method was essentially the same as the first, but one-mil washers of braze material coined between units were utilized. The third method was an attempt to make the heater tube completely impervious to gas flow as well as improve its brazing properties. This could be accomplished by coining the individual units between an inner and outer copper tube with thin copper plugs at both ends, in essence, completely jacketing the fuel tube in a copper container. The results of investigations on these methods is described under the section on bonding which follows.

D. Bonding

1. End-to-End

Attempts were made to obtain intimate end-to-end contact by coining prior to assembly or by coining the fuel tubes during assembly onto the inner tube. This was unsuccessful, especially with half-length units, and was due possibly to variations in shrinkage of the diameter of the fuel tube during sintering, causing slight seizing on the inner tube during coining. The heater tubes assembled in this manner showed too much end-to-end separation to be acceptable.

Use of braze material washers coined between fuel tube units were investigated and proved successful. Investigations were made employing one-mil washers of both brass and tin, coined between fuel tube units and heat treated for bonding purposes.

Results of investigations employing brass washers to obtain end-to-end bonds showed this method to be unapplicable for this type of assembly. Temperatures necessary for bonding proved excessively high and thus detrimental to the braze bond necessary between tubes.

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Use of tin washers coined between fuel tube units did prove successful. Heat treatment necessary for the bond was one hour at 600°C in dry hydrogen. This method proved to be the most acceptable and desirable due to the fact that as shown in (Fig. 3) the braze between tubes could be accomplished during the same heat treatment.

2. Tube-to-Tube

Although the excessive shrinkage during sintering had eliminated the sinter-shrink technique, it had shown distinct possibilities of forming a good diffusion bond between fuel tube and inner copper tube. Investigations were made to determine if an ordinary diffusion bond could be obtained with the fully sintered and coined fuel tube assembly. Maximum heat treatment, however, i.e. 1050°C for 16 hours in hydrogen, did not promote any self-diffusion of copper across the interface to form a bond and the method proved unfeasible.

The technique next tried was that of plating one-mil of tin on the inner tube upon which the fuel tube units were coined. Heat treatment at 600°C for one hour in hydrogen resulted in a good heat transfer bond between tubes as shown in Fig. 4 simultaneous with the end-to-end bond between tubes utilizing the tin washers. This technique was deemed successful when ductility tests of the heater tube assembly showed the unit could be flattened to 0.064 inch without cracking or deterioration of the bond.

Investigations on attempting to jacket the heater tube completely in copper were limited, and met with little success. The assembly method was as follows: Fuel tube units were coined between an inner and outer copper tube with tin washers between units, and a five mil copper plug was coined at both ends of the assembly. All surfaces in contact with the fuel tube were plated with one mil of tin. The assembly was then subjected to heat treatment at 600°C for one hour in hydrogen. Metallographic examination of the assembly following heat treatment revealed a wide boundary still existing between fuel tube and outer tube. Further efforts to improve on the technique were discontinued due to initial success in brazing the fuel tube directly to its stainless steel pad and block.

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Fig. 3 End-to-end bonding of a multi-unit fuel tube through use of a tin washer coined between units and heat treated at 600°C for one hour in dry hydrogen.

Plate No. 15848

250X

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Fig. 4(a)

Transverse Section

Plate No. 12732

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Fig. 4(b)

Longitudinal Section

Bonding of fuel tube to inner copper tube by coining fuel tube over inner copper tube plated with one mil of tin and heat treated for one hour at 600°C in dry hydrogen.

Plate No. 12733

250X

IV. METHODS UTILIZED FOR FABRICATING TEST SAMPLES

From the various investigations, the most suitable techniques were arrived at for fabrication and assembly of heater tube units. Various predetermined compositions of copper and hydrogen-fired UO_2 were weighed out and tumble-mixed together with 1/2 w/o zinc stearate. Individual charges of the mix were compacted in the die at 35 tsi through double action pressing to a length of about 9/32". These individual quarter-length units were then sintered for 16 hours at 1050°C in wet hydrogen, to approximately their final diameter and a length of 15/64". Four of these individual units were then coined with one mil tin washers between units at 35 tsi onto the inner copper tube, which had been plated with one mil of tin on the outer diameter. The method used was such that the fuel tube and inner tube were flush at one end, while the inner tube projected from the other end. The projecting inner copper tube was cut off to the correct length, and the accurately sized assembly heat treated at 600°C for one hour to bond the fuel units to each other and to the inner copper tube. A typical completed heater tube unit is shown in Fig. 5.

V. INVESTIGATION OF METHODS OF BRAZING HEATER TUBES TO PAD AND BLOCK

Ability to braze the Cu + UO_2 fuel tube surface to a type 347 stainless steel pad and block was one of the listed requirements of the program.

It was first thought possible to plate the Cu + UO_2 fuel tube and use the plate material as a braze directly to the stainless steel pad. Several units were made up with plates of copper, tin and silver and furnace heat treated in an attempt to use the plated material as a braze. No bonding took place because of a lack of diffusion with the copper plate and to the removal of all the molten tin or silver by capillary action into the surface pores of the fuel tube.

Commercial braze materials for joining the dissimilar metals copper and stainless steel, such as Handy and Harmon's B T and Silphos, were tried with good results. A Sylvania laboratory brazing alloy containing 10% TiH_2 , 28% Cu, 62% Ag proved to be outstanding because of its ability to wet the UO_2 . Because of the high cross-section of the silver constituent in these alloys, however, an attempt was made to substitute some other type of braze metals.

Promising results were obtained with ternary alloys of copper, tin and nickel. These were made with a mixture of the powders suspended in nitrocellulose and brushed onto the pieces to be brazed. Brazing was conducted in a furnace with dry hydrogen atmosphere at 1025°C for 10 minutes. Initial work resulted in a tightly adherent braze metal which wet both the stainless steel and the Cu + UO_2 tube. This work, however, was curtailed before proper

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Fig. 5 Fabricated Heater Tube Fuse Enlarged 6.4 Times
Plate No. 10,252

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investigation of composition and heat treatment could be completed.

The brazing method finally used by Atomics International consisted of nickel plating (0.0005 - 0.00075 inch thick) the fuel tube, diffusion heat treating at 950°C for one hour and brazing in a hydrogen atmosphere furnace at 850°C using Handy and Harmon grade SS silver solder. (9,10)

VI. CONCLUSIONS

The fabrication by powder metallurgy techniques of heater tube fuses has been proven feasible.

Best results have been obtained when these assemblies are produced with multi-unit fuel tubes. This insures better dimensional tolerance when shrinking during sintering, higher densities and thus greater ductility for flattening. Producing fuel tubes in single lengths has been shown to be possible. However, a multiple piece die body is deemed necessary to eliminate difficulties in ejection of such a piece as well as obtaining a high volume of production.

A coining operation has been incorporated into the assembly phase of producing these heater tubes, resulting in high-density fuel tubes which are critically sized.

Using the sinter-shrink technique for bonding the fuel tube to the inner tube has proven unfeasible in the production of these heater tube fuses. Restraining shrinkage in one direction causes excessive shrinkage in the other, leading to radial cracking of the full-length fuel tube and excessive separation between units of the multi-unit fuel tube. Attempts at obtaining a bond between fully sintered and coined fuel tubes and the inner copper tube by self-diffusion of copper during heat treatment were also unsuccessful.

The required bond between fuel tube and inner tube was accomplished by use of a one-mil tin plate on the inner copper tube, upon which the multi-unit fuel tube was coined. Heat treatment at 600°C for one hour in hydrogen produced a good heat transfer bond between fuel tube and inner tube. During this same heat treatment, one-mil tin washers coined between the multi-unit fuel tubes melted and insured an end-to-end metallurgical bond.

The objectives listed in the introduction were met with delivery of the thirty-three enriched heater tube fuses of the required dimensions. In addition experiments with these units at Atomics International show that some of the requirements listed in the introduction were clearly exceeded, as the units withstood an internal pressure of 15000 psi when under test. (12)

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