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FABRICATION DEVELOPMENT OF APM ALLOYS FOR FUEL ELEMENTS

AEC Research and Development Report

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FABRICATION DEVELOPMENT OF APM ALLOYS FOR FUEL ELEMENTS

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ABSTRACT

The development work being done at Atomics International, to adapt aluminum powder metallurgical products (APM) for organic reactor fuels, is described. An APM alloy (Alcoa M257 alloy). containing 5 to 7 wt % finely dispersed Al_2O_3 in a pure aluminum matrix, was tentatively selected as a cladding for UO_2 fuel pellets, on the basis of its superior creep and tensile properties at temperatures to 900°F. Mechanical properties of M257 alloy, together with its formability, weldability, and adaptability to fuel element requirements, are described.

Details of the following fabrication developments are presented:

1) Description of APM finned fuel cladding tubes formed to close tolerances by impact extrusion. Tube dimensions were 0.305-in. ID by 0.030-in. wall containing eight longitudinal external fins.

2) Extruded APM square box shapes for fuel bundle containment.

3) Silver eutectic diffusion bonding process development for obtaining end closures in M257 aluminum fuel tubes.

4) Special techniques used for assembly and thermocouple instrumentation of M257 clad, uranium dioxide fueled prototype fuel elements.

I. INTRODUCTION

Aluminum powder metallurgy (APM) products are being developed by Atomics International for application in organic moderated and cooled reactor (OMCR) systems. APM alloys are superior to conventional wrought aluminum alloys, in that they possess good high temperature strength which is retained after prolonged periods of heating. This high temperature strength, along with the properties of high thermal conductivity, low neutron absorption, and excellent corrosion resistance in organic coolants, make APM alloys especially applicable as cladding and structural materials for OMCR fuel elements. M257, an APM alloy produced by Alcoa, is being developed as a fuel cladding for ceramic fuels, such as UO_2 or UC, which offer the potential of high fuel burnup and low fuel cycle costs. OMCR fuel elements, utilizing these ceramic fuels clad with M257, can be operated at temperatures up to 900°F, some 200 to 300°F higher than for wrought aluminum clad elements.

Fuel elements, consisting of UO_2 pellets contained in M257 fuel tubes, are being fabricated by Atomics International for testing in the OMRE. The fuel tubes are forward extruded, using the impact extrusion process. Typical tubes are 0.305-in. ID, with 0.030-in. walls, and have eight longitudinal fins, 0.030-in. wide by 0.086-in. high. For a typical element, the tubes are extruded in 5-ft lengths, with a tolerance of ±0.003 in. for both wall thickness and diameter. However, in other fabrication development efforts, tubes with 0.015-in. walls and ten longitudinal fins have been impact extruded in lengths up to 14 ft. Prior to loading the tubes with the UO_2 fuel, the tubes are further processed by twisting, to give a uniform spiral of 45°/ft. End closures in the fuel tubes are made by a eutectic diffusion bonding technique developed at Atomics International.

A structural application for APM alloys in OMCR is the process tube, or fuel box, which contains the fuel tubes and directs the coolant flow. At OMCR conditions, this box must withstand internal pressures of 16 psi, at velocities up to 30 fps and temperatures up to 700°F. Since stainless steel has been used for this application in the past, it is expected that boxes for future OMRE elements, and for other OMCR power reactor designs, will use these alloys, thereby producing a considerable increase in neutron economy.

II. PROPERTIES OF APM

Two types of APM alloy are available commercially from Alcoa: (a) a dispersion of Al_2O_3 in a matrix of aluminum, and (b) a pre-alloyed atomized powder, utilizing insoluble intermetallic compounds as the dispersion strengthening media. Alloy M257, the alloy being evaluated by Atomics International, contains about 5% of Al_2O_3. Other alloys made by Alcoa are M470 and M430, containing 10 and 14% Al_2O_3 respectively. A recent addition, M583 (designed as a replacement for M257) is being produced according to a revised powder preparation method. The main objective is to yield a product with a lower impurity content than M257. Future fuel element fabrication efforts will include the evaluation of this M583 alloy. Alloy M486, one of the currently available pre-alloyed powder products, has the nominal composition of 7.8% Fe with 0.2% each of Cr, Ti, Zr, and V, and contains approximately 0.5% Al_2O_3 as an impurity. Other alloys include M457 and M643, both containing Fe and Ni as alloy additions, and are being developed for their corrosion resistance to high temperature water or steam.

The mechanical properties of APM alloy M257, along with those of the other alloys mentioned above, have been published.[1] These data include tensile properties at temperatures up to 1000°F, creep-rupture properties up to 900°F, and creep properties up to 600°F. The objectives of the mechanical property studies, undertaken in the program at Atomics International, were to confirm published data, to further develop creep and creep-rupture properties in the temperature range above 600°F, and to determine the effect, if any, of fabrication history on mechanical properties.[2,3] The principle alloy to be investigated was M257, as it was most readily available and possessed the most desirable combination of ductility and strength at temperatures above 800°F.

The average tensile properties of M257, for temperatures up to 900°F, are depicted graphically in Figure 1. The values plotted include data for M257 in the as-extruded condition and for as-rolled sheet. Previously published values are also plotted for reference. Good agreement was obtained for room temperature tensile properties for both conditions. At temperatures above 600°F, the ultimate and yield strength values determined in this investigation were some 10 to 15% lower than those claimed for M257 in the as-extruded condition; conversely, however, it was shown that the ductility (elongation) increased at

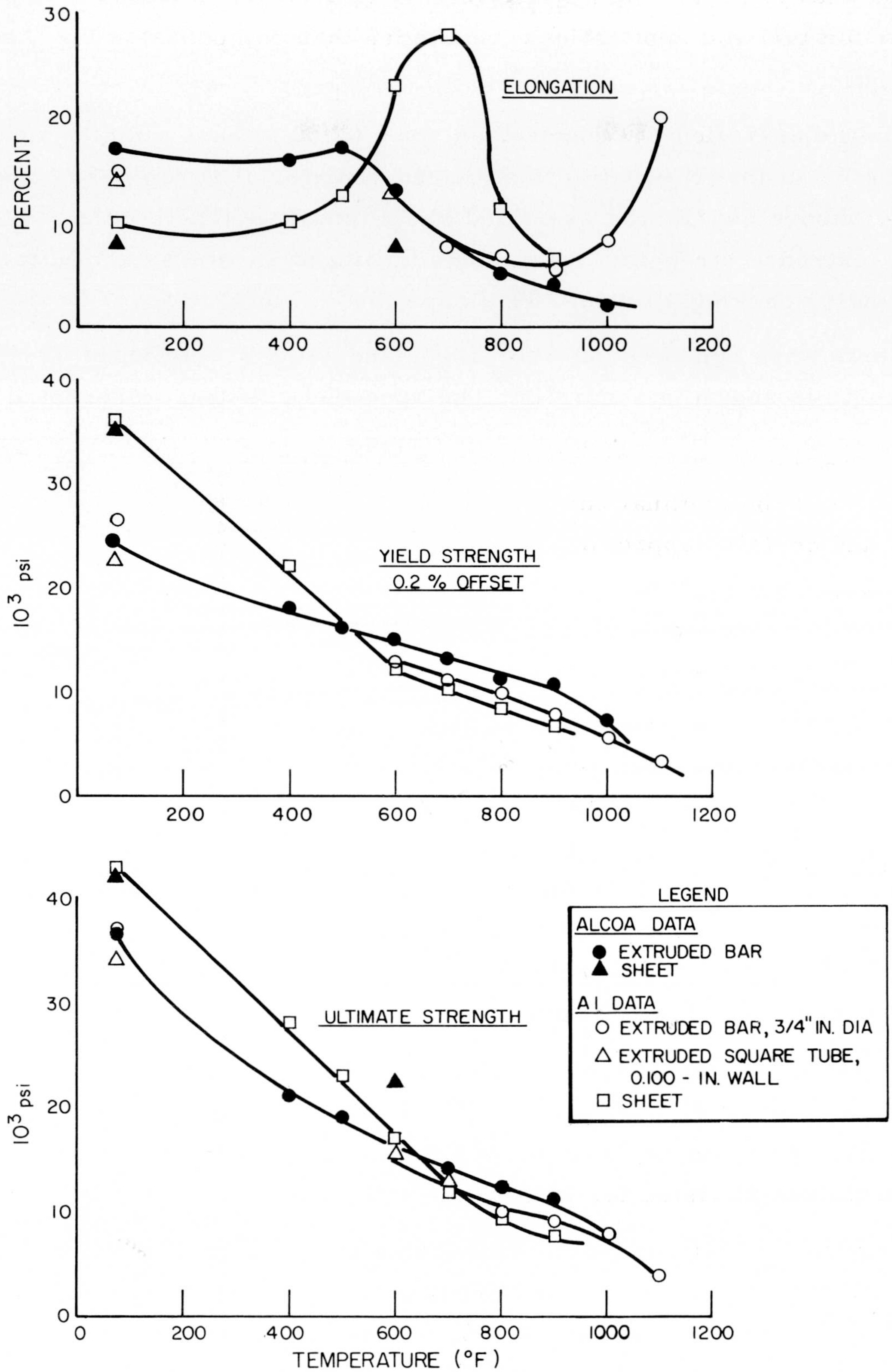


Figure 1. Tensile Properties of APM Alloy M257

temperatures above 900°F. This improvement in ductility is believed to be advantageous for fuel rod applications, and more than compensates for the lower tensile strength.

M257, in the as-rolled sheet condition, was found to have slightly higher tensile strength and lower ductility than extruded material at temperatures up to about 600°F. Above 600°F, the as-rolled sheet tensile strength falls slightly below the as-extruded strength. Also, the ductility of sheet was shown to increase markedly, peaking at about 28% elongation at 700°F.

Creep tests were conducted at 900, 1000, and 1100°F. A plot of creep-rupture strength is shown in Figure 2. The specimens tested represented four

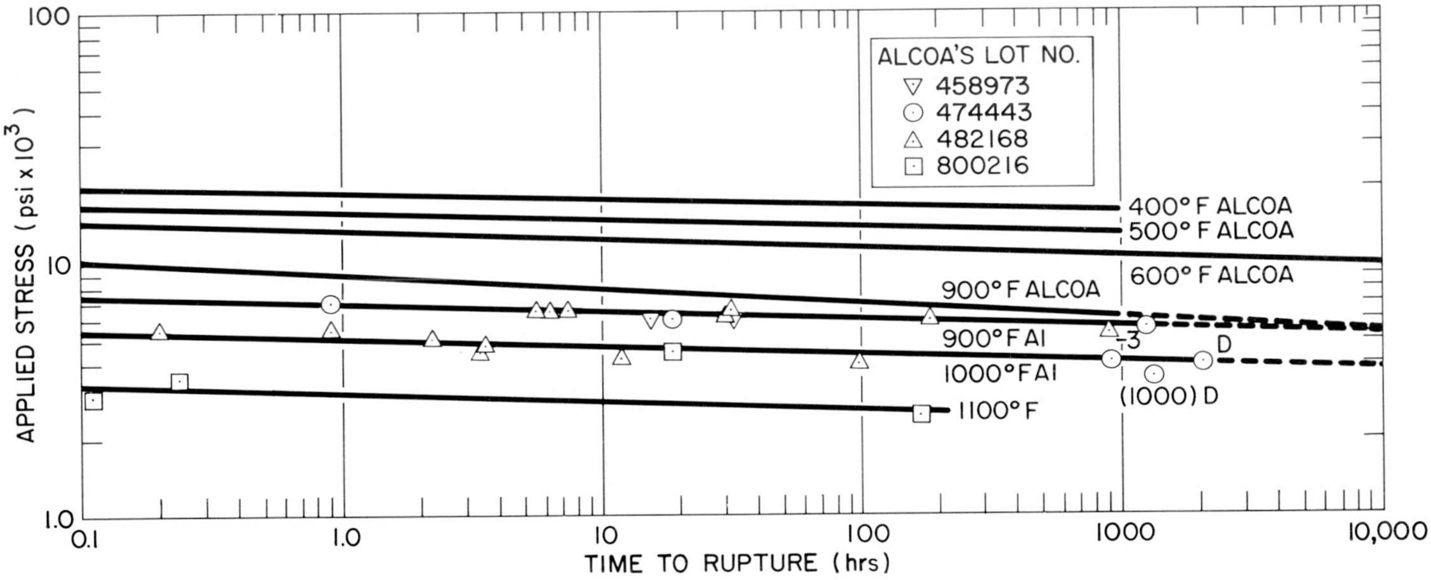


Figure 2. Creep Rupture Strength of M257 (As-Extruded)

different lots of extruded stock, and the data indicate that there is no significant lot-to-lot variation in creep-rupture strength. Previously published data are also plotted for reference. As found for M257's tensile properties, the data developed by Atomics International show a small but significant decrease in the creep-rupture strength listed for M257.

The creep characteristics of Al-Al_2O_3 alloys are unique, in that most of the deformation occurs in the primary creep stage, with only negligible secondary creep until rupture. APM alloy M486, a pre-alloyed powder, rather than a mixture of Al_2O_3 in Al, creeps in much the same manner as conventional ductile metals, with the expected primary, secondary, and tertiary stages of creep. However, the creep strength of M486 deteriorates rapidly at temperatures above 800°F.

Tensile and creep tests have shown that elevated temperature strength properties of M257 are lowered somewhat by cold working, as induced by cold rolling and swaging. However, the effect of cold working is small, as shown by the tensile data of Figure 1 and the creep-rupture data of Figure 3. All of the

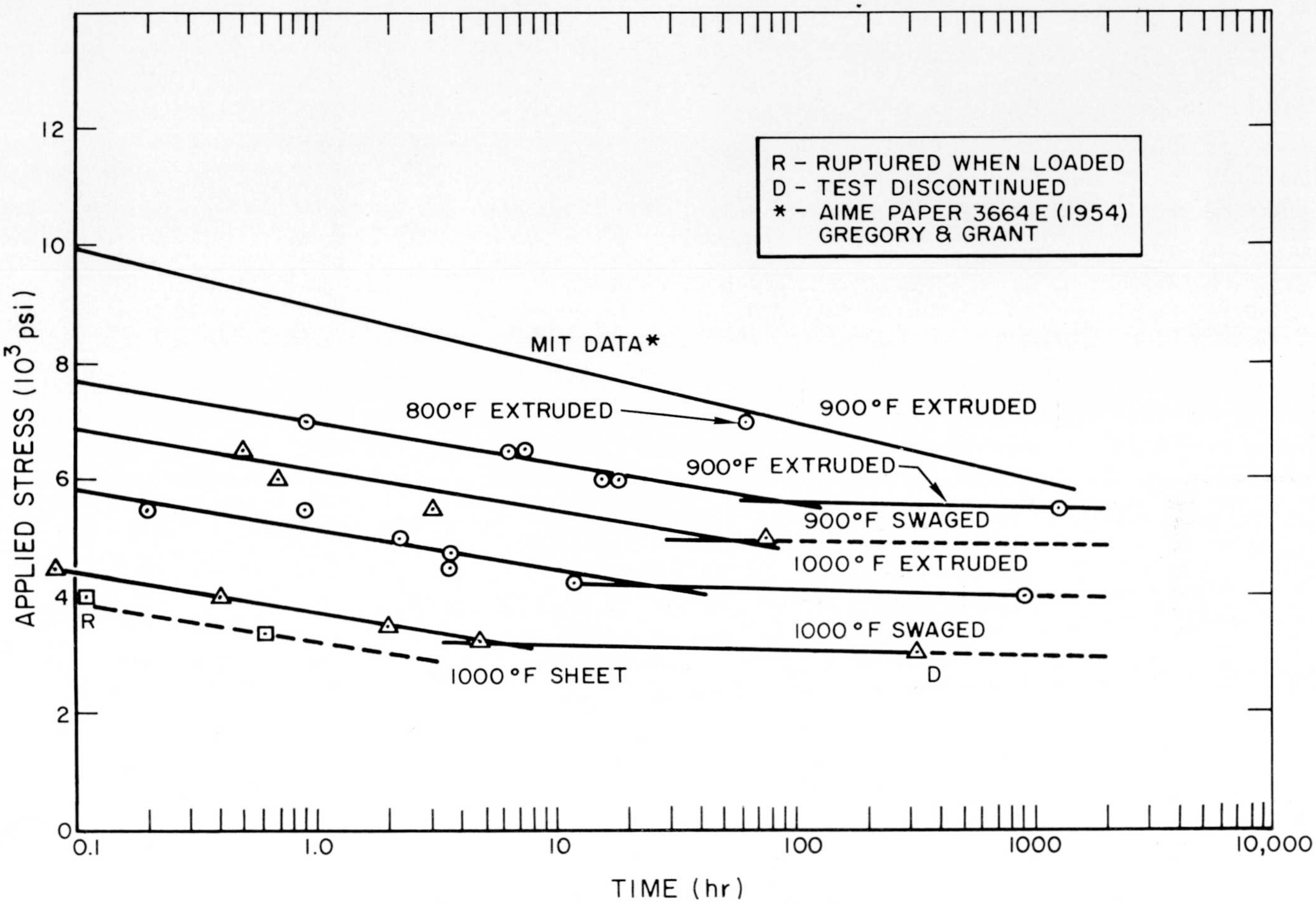
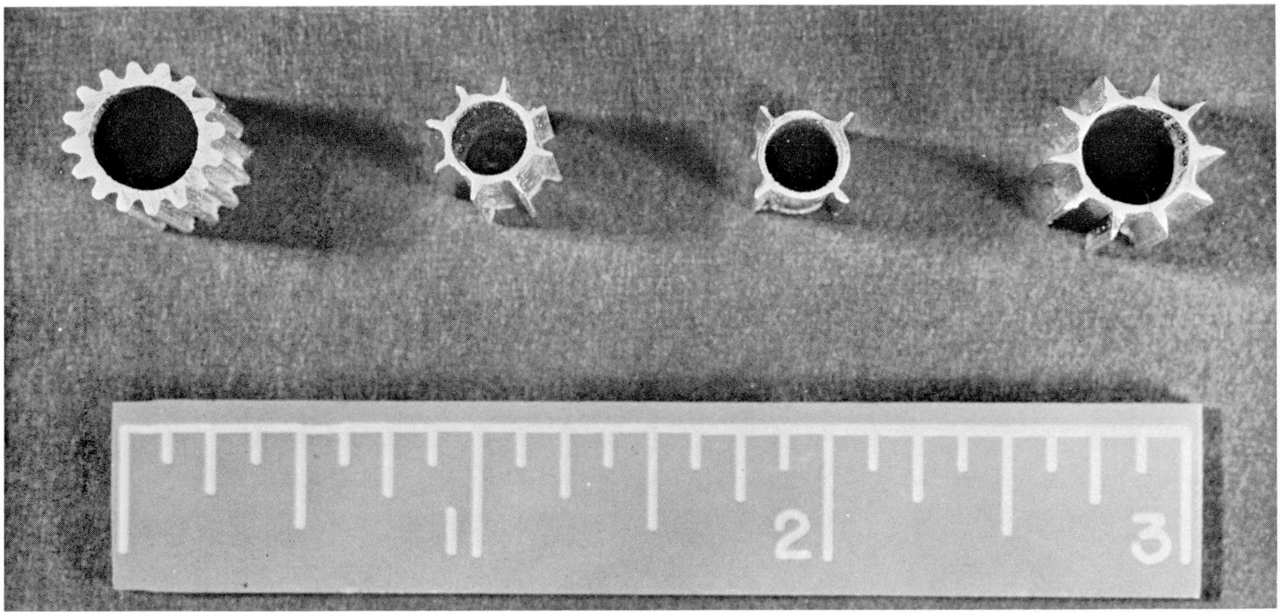


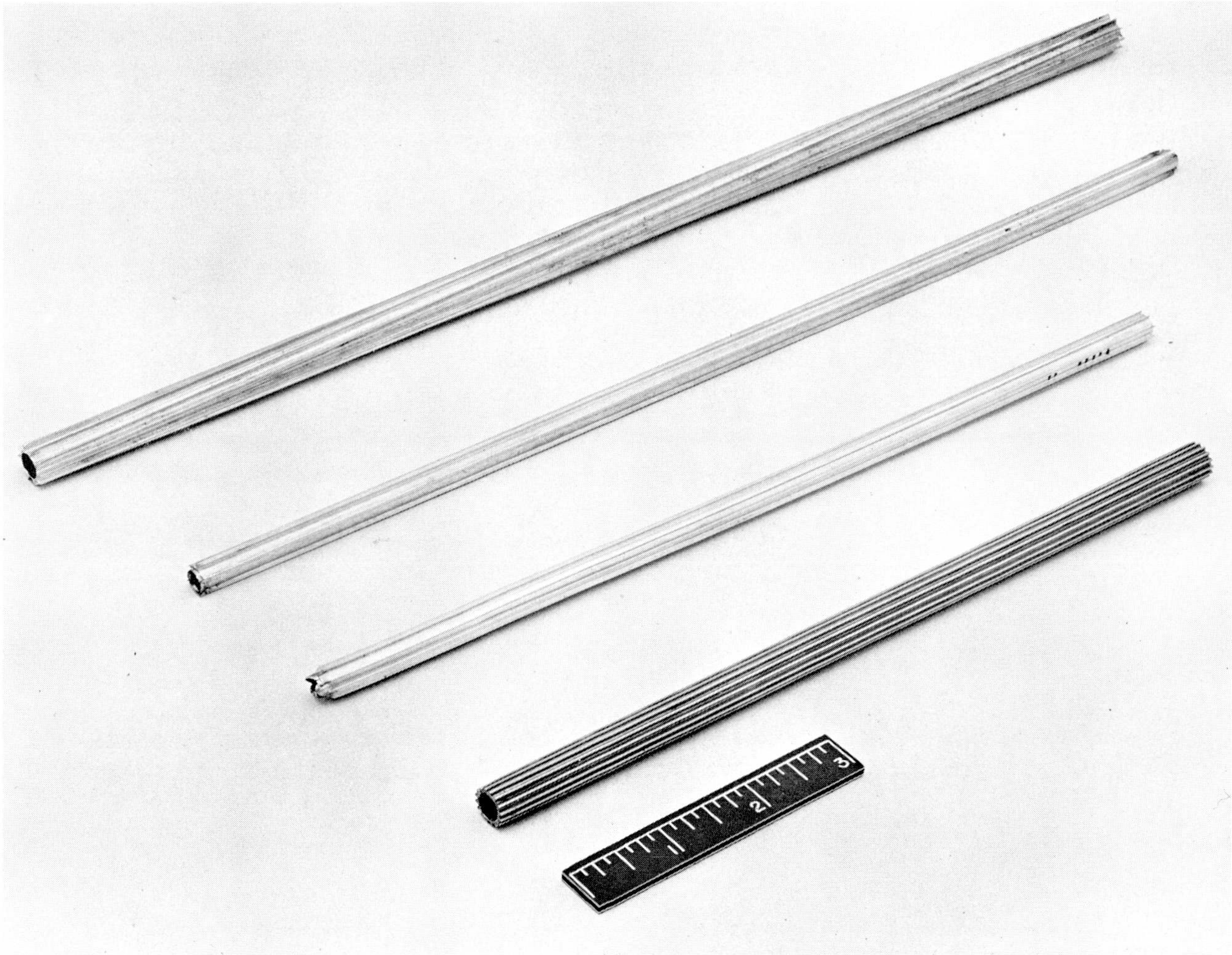
Figure 3. Creep Rupture Data for M257

APM alloys are used in the as-fabricated condition, since they do not respond to aging, annealing, or solution heat treatments.

The effects of irradiation on tensile and impact properties of M257 and M486 have been investigated, and differences between irradiated and unirradiated properties were found to be insignificant.[4]



a. End View



b. Side View

Figure 4. Tubular Shapes Extruded from M257

III. FABRICATION OF APM

APM alloys can be fabricated to many of the common mill shapes by conventional forging, rolling, or extrusion processes. In general, the fabrication characteristics of M257 were found to be very similar to those of Type 7075 aluminum alloy. One of the principle shapes used by Atomics International is a longitudinally finned tube, which serves as a fuel tube for containing UO_2 pellets. The external fins provide extended surfaces which increase capability of the tube to transfer heat to the organic coolant. Finned tubes of various configurations and sizes (See Figure 4), have been produced by forward extrusion techniques. The most successful technique has been hot impact extrusion at 800 to 900°F. By this process, finned tubing, of excellent quality and dimensional control, has been produced from M257. An 8-fin design configuration, with dimensions and tolerances, is shown in Figure 5. Although tubes of this configuration are presently

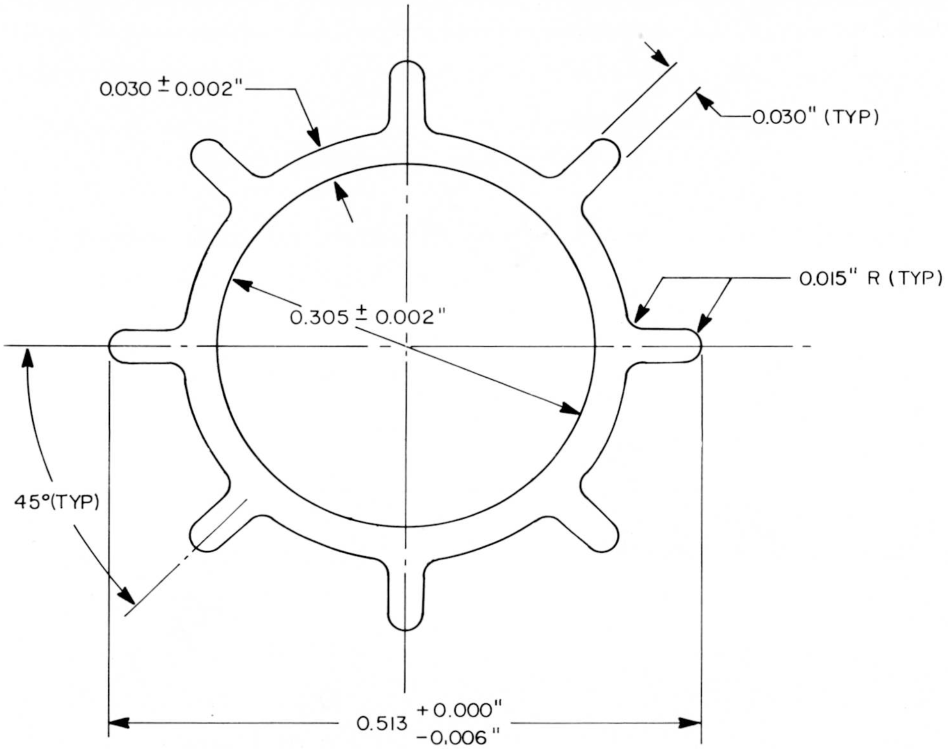


Figure 5. Eight-Fin M257 Fuel Tube

extruded in lengths of about 5 ft, to attain the closest dimensional control possible, lengths of up to 14 ft have been produced by impact extrusion.

Based upon a pilot run of 1000 ft of tubing, dimensional tolerances consistently obtainable in these lengths are as follows:

OD (taken on opposite fins)	0.505 ± 0.003 in.
ID	0.305 ± 0.002 in.
Fin Height	0.075 ± 0.002 in.
Fin Base Thickness	0.030 ± 0.003 in.
Wall Thickness	0.030 ± 0.0025 in.

Ten- and twelve-fin variations of the same size (0.30 in. ID by 0.030 in. wall) tubing have also been successfully extruded, with good indications of comparable dimensional control. Extrusion experience has shown that acceptable quality tubing can be expected, in wall thicknesses as low as 0.015 in.

Various tests, both destructive and nondestructive, have been used to establish reasonable confidence in extruded M257 finned tubes. Since cold forming operations after extrusion (especially severe tube drawing) have shown evidence of lowering the mechanical properties of tubing, only as-extruded tubing is recommended for reactor fuel cladding.

Finned tubing is inspected for cracks, seams, slivers, blisters, burns, die marks, or other injurious imperfections which are known to affect the physical properties of the tubing. Surfaces must be clear to fluorescent penetrant inspection.

The M257 tubing has conformed to the following composition and impurity levels (all values in weight percent):

Aluminum	Remainder
Aluminum Oxide	5.0-7.0
Iron	0.70 max.
Silicon	0.30 max.
Boron	0.001 max.
Cadmium	0.003 max.
Other Impurities, Total	0.015 max.
Individual	0.005 max.

Minimum tensile properties of billet or tubing are as follows:

	Room Temperature	900°F*
Ultimate tensile strength (psi)	32,000	8,000
Yield strength, 0.2% offset (psi)	19,000	6,500
Elongation in 2 inches (%)	10 - 18	4

Each length of tubing must be leaktight, when tested by a mass spectrometer leak detector which will detect a leak rate of 1×10^{-9} std. cc/sec of helium. Testing is performed at room temperature and 900°F, at a pressure differential of 1 atm for a period of at least 5 min.

Borescope inspection of surfaces, and isostatic internal pressure tests (3000 psig for 8-fin, 0.030-in. wall by 0.305-in. ID tubes), complete the incoming inspection of as-extruded tubing. Tubes which pass as-extruded inspections are stretch-straightened and spiralled in one operation, end cropped, and chemically cleaned. The results are shown in Figure 6.

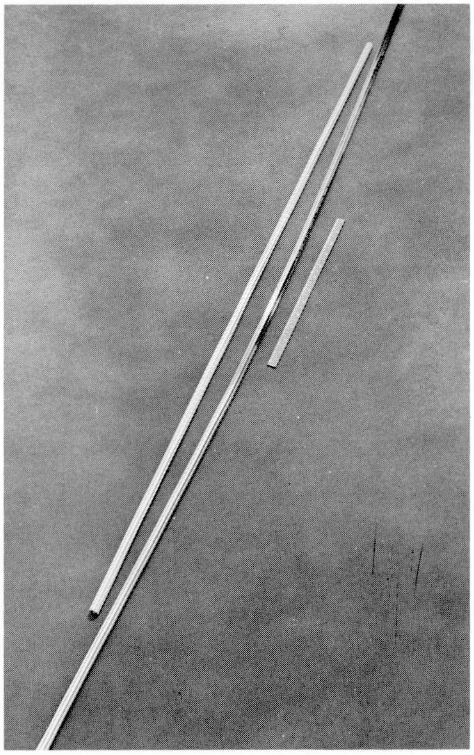
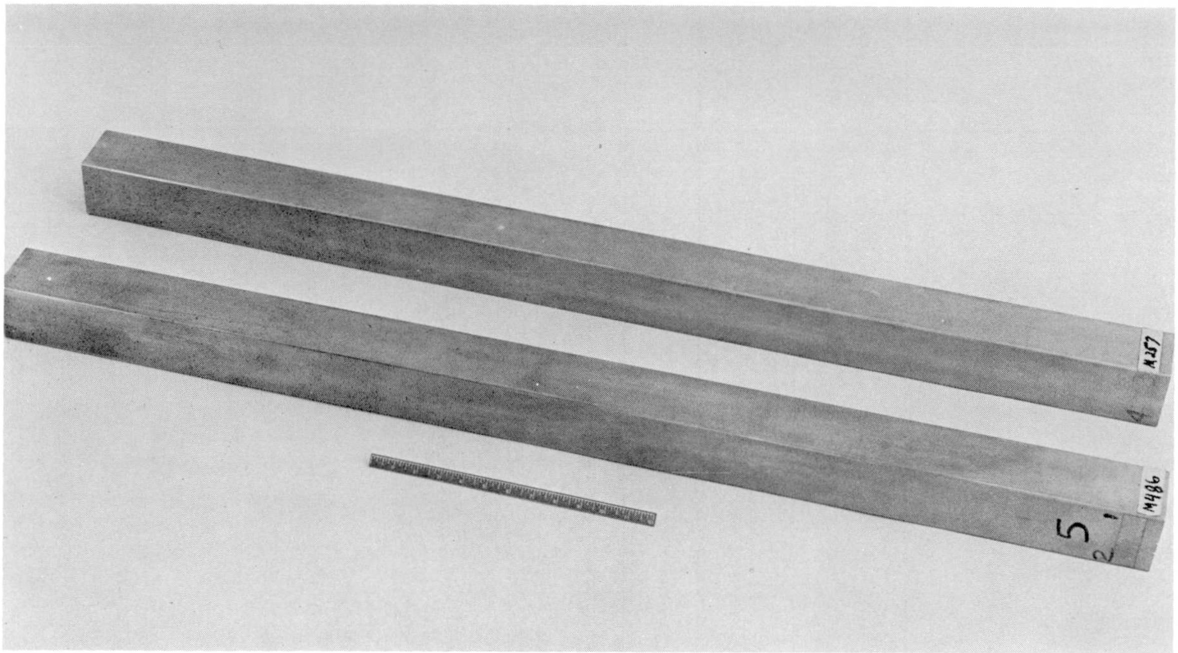
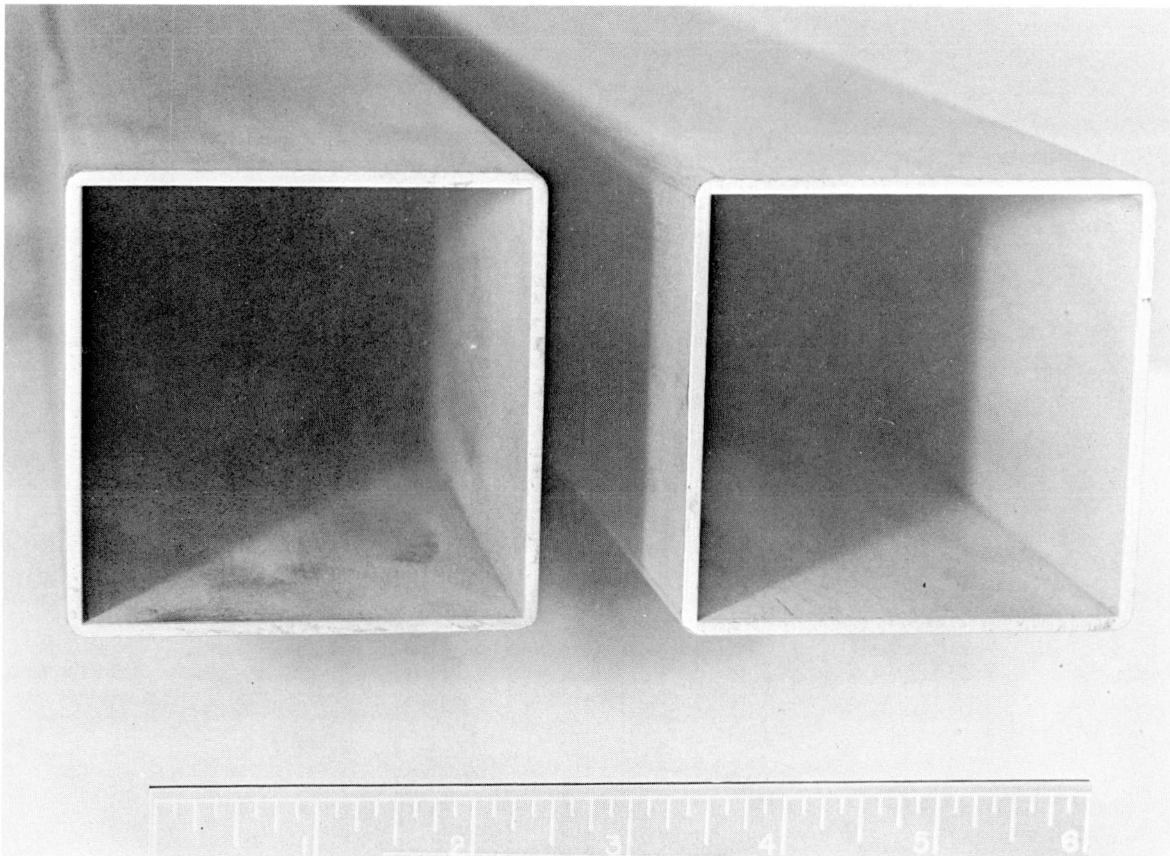


Figure 6. M257 Tubes, As-Extruded and After Straightening Spiralling, and Cleaning

*After heating for one hour at temperature.



a. Longitudinal



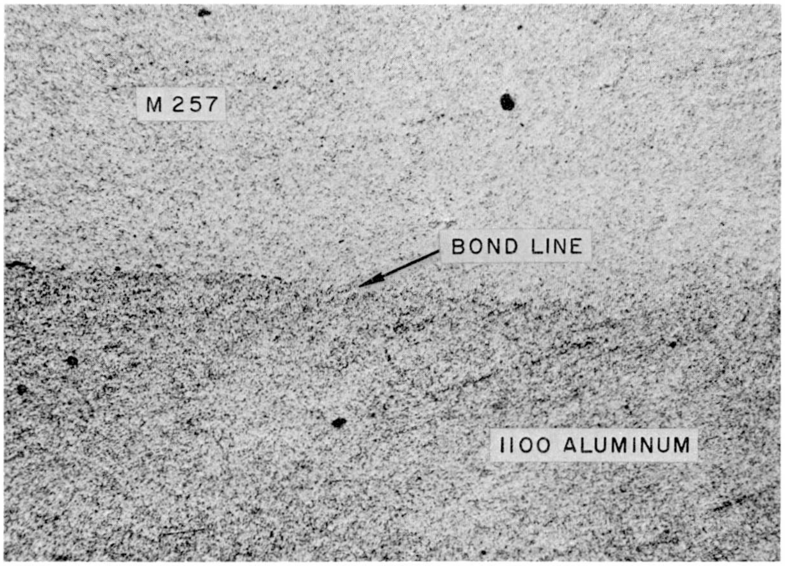
b. Cross Section

Figure 7. Extruded Square Boxes of M257 and M486

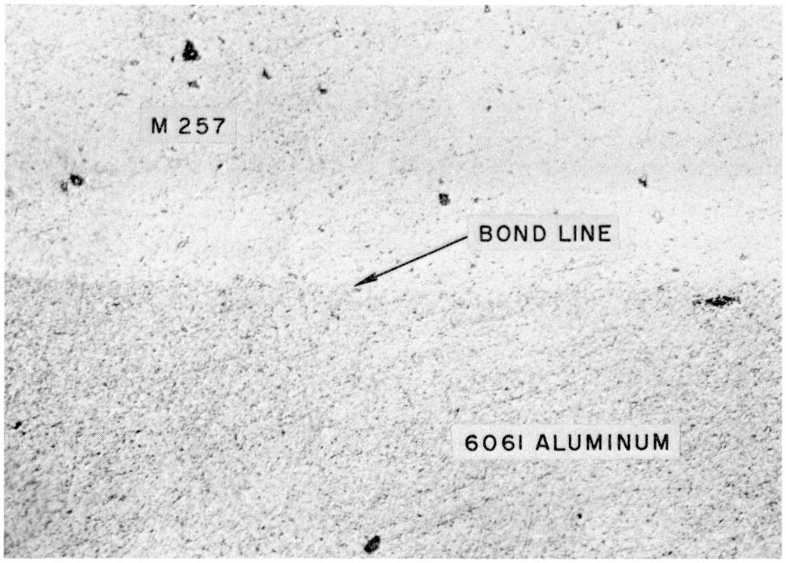
If tubing is to be used for in-pile experiments, a more thorough inspection is made before fuel is loaded. This inspection is similar to previous inspections, except that a complete dimensional profile of each tube is made, with all dimensions being recorded. A fin twist of 45.0 ± 5°/ft is required, and inspection is made at this time to assure conformance. Complete dimensions are taken at 4-in. intervals along the entire length of the fuel tube. Such data is useful in ascertaining fuel tube dimensional changes, which may occur during irradiation and burnup tests. Air gauge probes are used for tube ID measurements, while specially constructed inspection fixtures are employed to measure tube exterior profiles.

Twisting the tubing, to produce the helical fin configuration, is performed during the stretch-straightening operation. This operation consists of clamping the ends of the tube in the rotatable grips of a modified tensile machine and exerting a load which slightly exceeds the yield strength of the metal. While this load is maintained, the end grips are rotated equally. Due to springback, the rotation required is twice the 45°/ft twist desired. The stretching load is released before the twisting forces are relaxed. The described procedure yields tubing with overall straightness of ±0.005 in. TIR in 5-ft lengths. A slight and uniform reduction in tube dimensions (0.0005 in. max.) also occurs. Thermal cycling of processed tubing to 1000°F has been shown to produce no relaxation in fin twist.

APM alloys are also being developed for structural applications in organic reactors. One of these applications is as a pressure tube, or fuel box, which contains the fuel tubes and directs the coolant flow. In recent development efforts, square tubes, 2.950 ± 0.005 in. outside dimensions, with 0.100 ± 0.005 in. walls, have been successfully extruded in 48 in. lengths from two APM alloys, M257 and M486. Typical tubes are shown in Figures 7a and 7b. Tests have shown that these square tubes possess the necessary strength and dimensional stability for this application.



a. M257 - 1100 Al



b. M257 - 6061 Al

Figure 8. Microstructure of Eutectic Diffusion Bonds

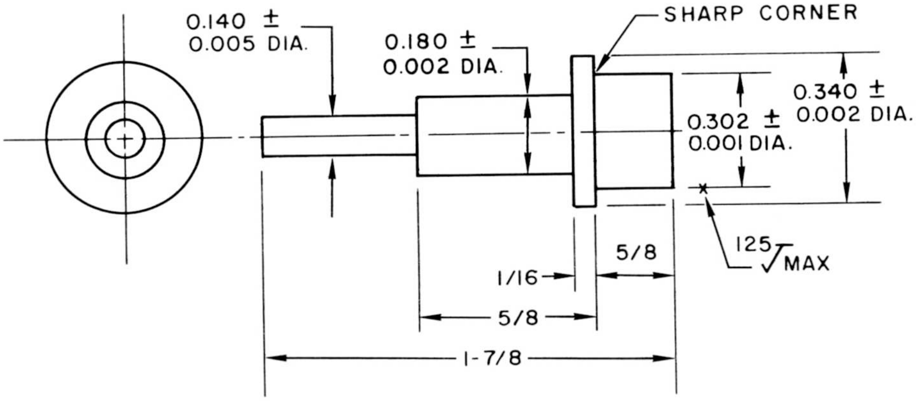
IV. TUBE END CLOSURES BY EUTECTIC DIFFUSION BONDING

APM products do not lend themselves to joining by conventional techniques, such as inert gas arc welding. Any fusion by melting usually segregates the oxide particles from the matrix aluminum. A technique for obtaining helium leaktight end closures on M257 tubes was developed, using a eutectic diffusion bonding process.

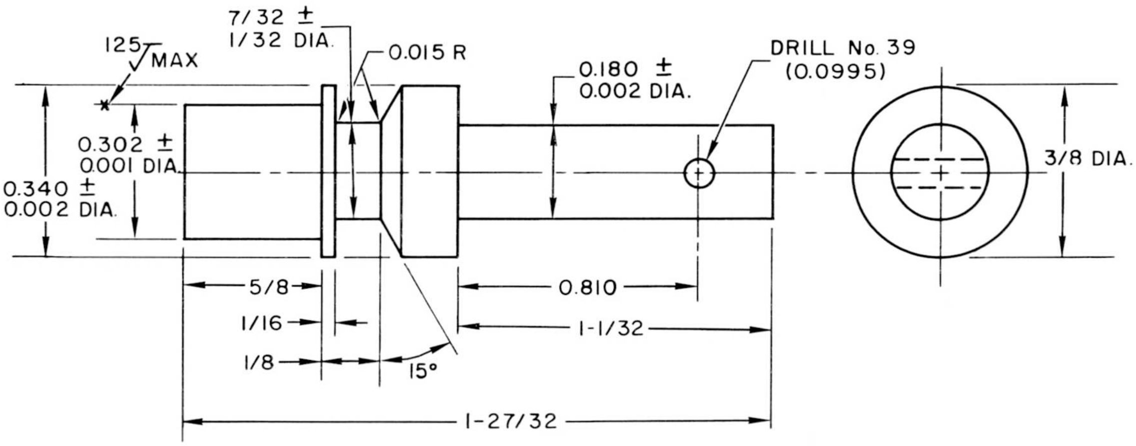
Metals, including Ag, Cu, Sn, Zn, and Al - 12 wt % Si, which all form a eutectic with aluminum at least 100°F below the melting point, were investigated. Preliminary trials were made, using both vacuum deposited and electroplated materials where appropriate. Metallographic examination revealed bonds were attained using Ag, Cu, and Sn. However, Ag always exhibited more total diffusion, and yielded bonds free of contamination. Bonding has been achieved, utilizing M257, and Types 1100 and 6061 Al alloys, for end plug material. The 1100 and 6061 Al alloys permit the use of much lower bonding pressure; and, for this reason only, they have been selected in preference to the M257. Microstructures of typical bonds are shown in Figure 8.

In preparation for the bonding operation, straightened and twisted tubing is end cropped, machined to a final length, and the fins removed from each end, for a distance of 1/4 to 1/2 in. Tubing and end plugs are then vapor degreased in trichlorethylene and alkali cleaned, using a bath of Oakite 160 in water, followed by a nitric acid pickle. It has been determined that 0.3 mil per surface of metal is removed during the cleaning cycle; which is, in turn, partially compensated for by the silver plate.

Machined dimensions of the top and bottom end plugs used for test elements to be irradiated in the OMRE are shown in Figure 9. A 0.04-in. diameter hole, extending through the top end plug, was formerly used to evacuate the tube and backfill with helium, subsequent to the bonding of both end plugs. This method was required when bonding was done in air. However, recent results indicate the desirability of inert atmosphere bonding within a glove box. End plugs are cleaned immediately prior to the 0.1-mil thick silver electroplate. The surface of the plug and tubing are then brought into intimate contact, using a pressure of 5000 psi exerted by a pneumatically loaded collet die (see Figure 10) that applies uniform radial pressure. Induction heating is used to heat the restraining die, as well as the components to be bonded. Sufficient heat is applied to reach the



a. Top



b. Bottom

Figure 9. Design of End Plugs

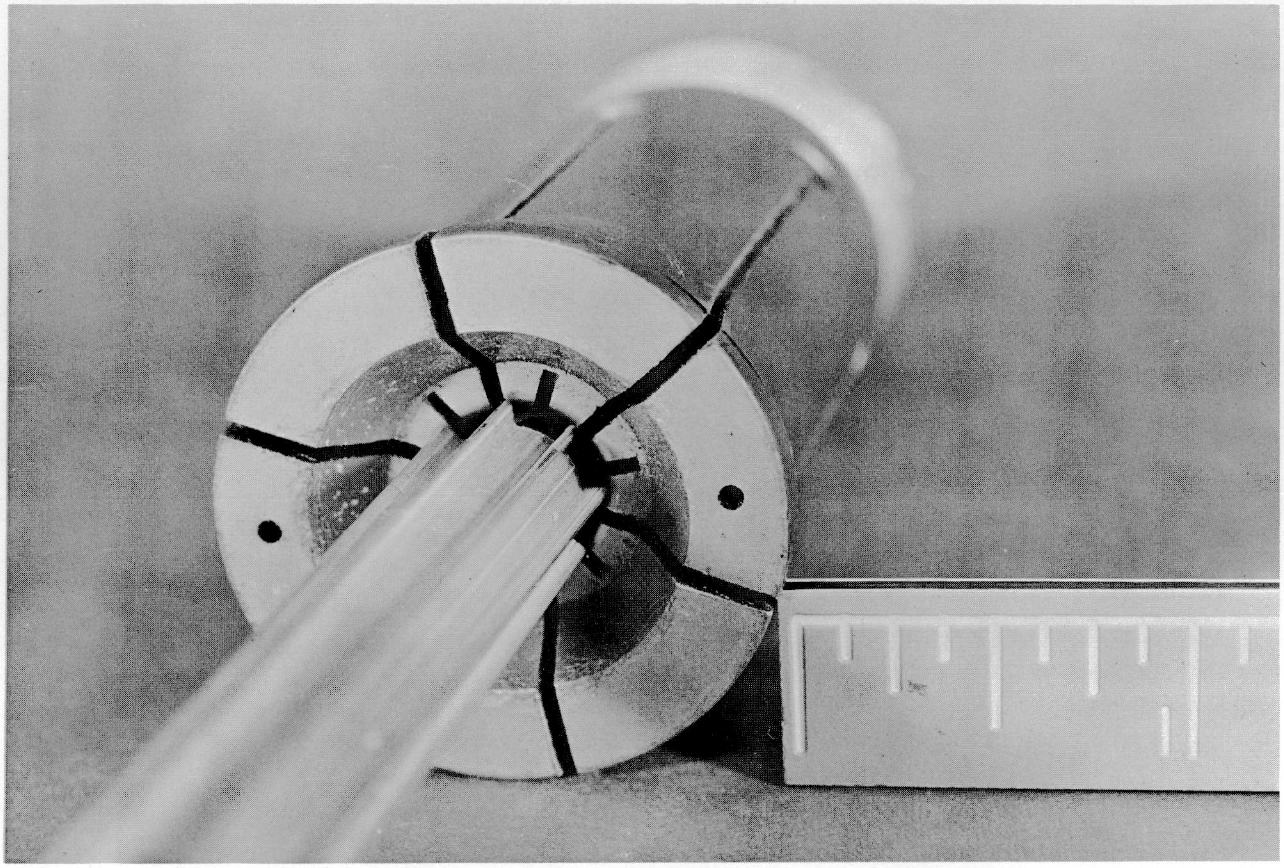


Figure 10. Six-Segment Collet Die for Bonding

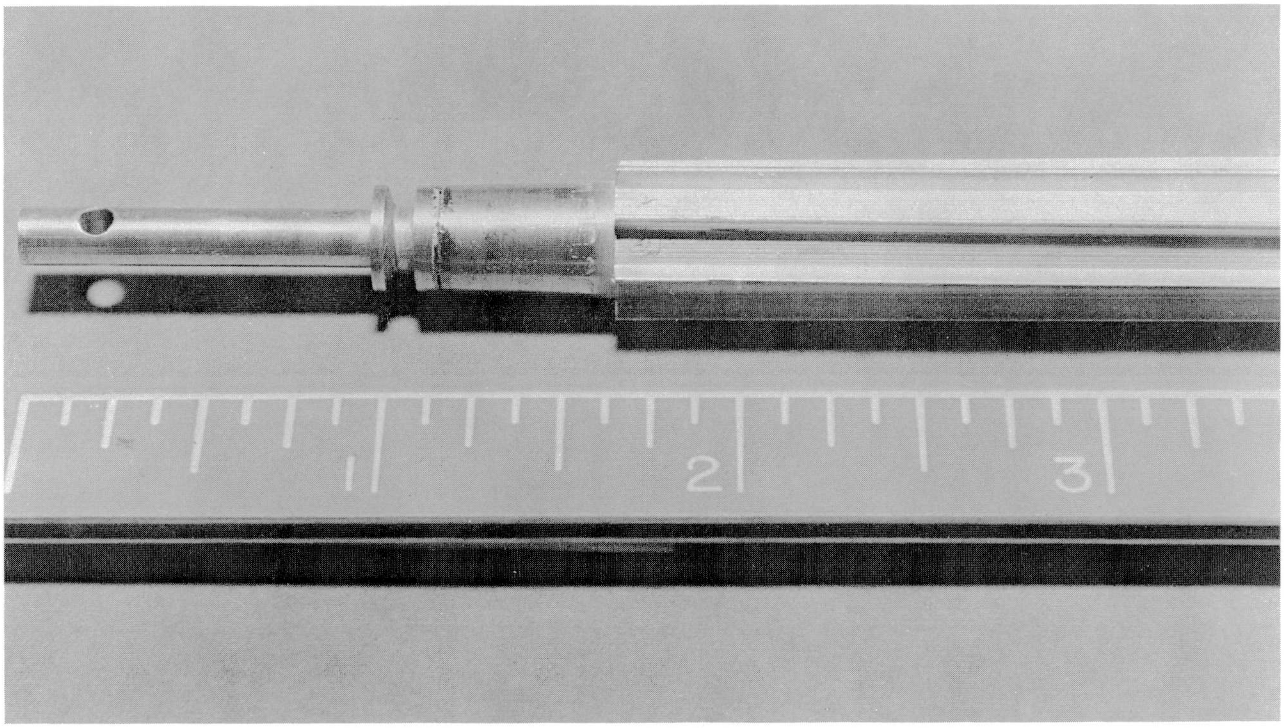
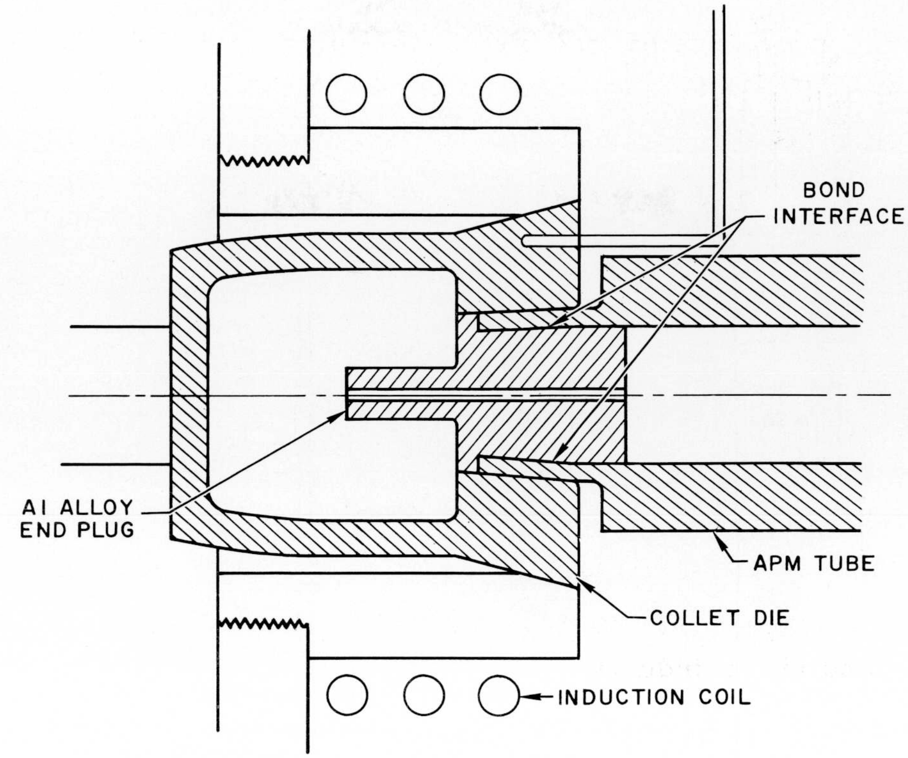


Figure 11. Typical Fuel Tube End Closure

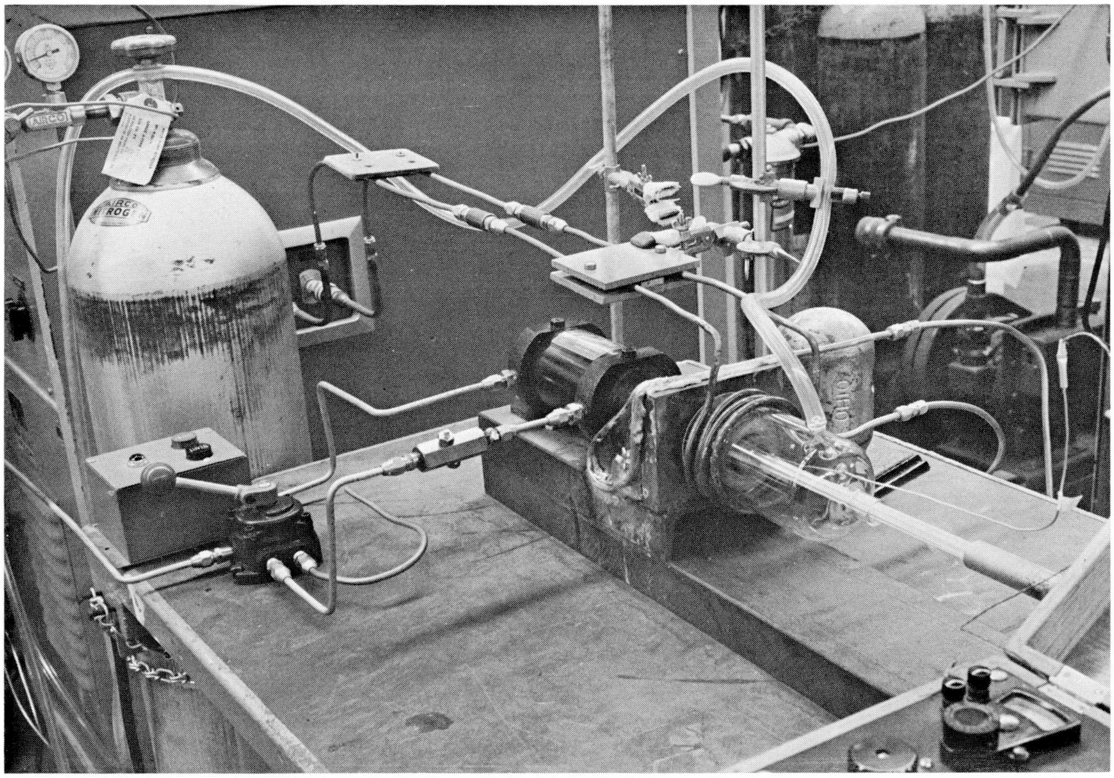
eutectic melting temperature of silver and aluminum (1050°F). Localized eutectic melting occurs at the interfaces, to establish a bond between the end plug and tube. Continued heating causes metal of eutectic composition to diffuse into the end plug and tube matrices. This process has been reduced to practice for making M257 tube end closures of the type shown in Figure 11. End closures obtained in this manner, using 30-mil wall tubing, have remained helium leak-tight after: (a) thermal cycling between 70 and 900°F for 30 min. intervals, and (b) internal pressure tests of 1500 psig at 900°F and 3000 psig at 70°F. In addition, the end closures have always proved stronger than the tubing itself, when burst tested at 750°F. A schematic diagram and photograph of the bonding equipment arrangement are shown in Figures 12a and 12b respectively. The apparatus consists of: (a) the six-segment collet-type die, (b) a matching tapered die sleeve, (c) a pneumatic cylinder loading device, (d) temperature sensing thermocouples, and (e) an induction coil for heating.

The microstructure of a well established bond is that of complete grain continuity extending across the interface, with no observable silver or intermediate diffusion zones remaining.

Metallographic examinations of bonds indicate reproducibility, irrespective of whether bonding is accomplished in air or inert atmosphere. However, indications are that inert atmosphere bonding provides improvement in bond strength and reliability, due to less danger of oxide formation at the interfaces. Thus, the arrangement shown in Figure 12b has since been installed inside a glove box.

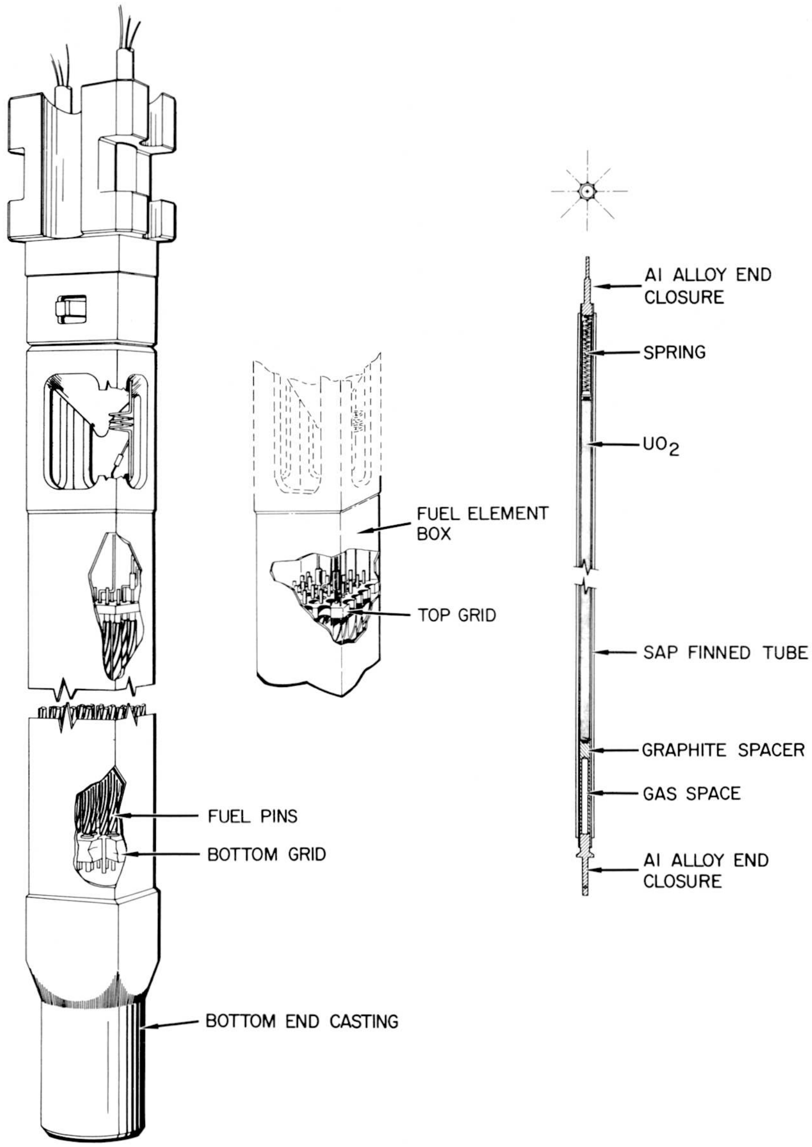


a. Schematic

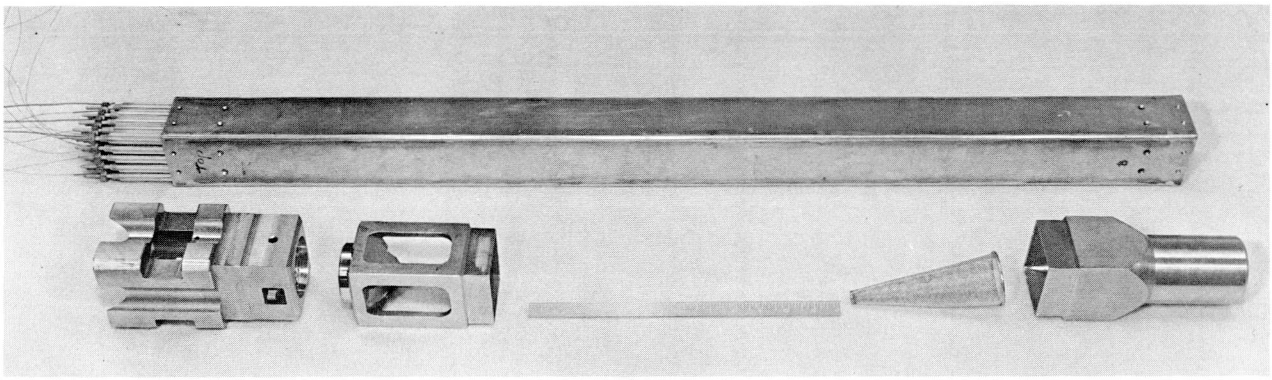


b. Photograph

Figure 12. Bonding Apparatus



a. Design



b. As-Fabricated

Figure 13. APM-UO_2 Irradiation Test Element for OMRE

V. FUEL ELEMENT FABRICATION

Based upon a prototype fuel element design for the 50 Mwe improved cycle OMCR, test elements, 3 ft long, have been fabricated for irradiation in the OMRE. A test element, shown in Figure 13a, consists of a 25-rod tube bundle of UO_2 pellets within the M257 alloy finned tube cladding. A 25-rod bundle was selected because of its ability to be inserted into existing fuel element positions in the OMRE. The 25 fuel tubes are spaced in a 5 by 5 array, by means of stainless steel end spacers. After bundling and the attachment of thermocouples, the assembly is inserted into a stainless steel box, and stainless steel fixtures are welded to the ends. An exploded view of the complete element can be seen in Figure 13b.

Sintered and ground UO_2 pellets, 94% theoretical density, are being used as the fuel material. An annulus of 2.5 to 3.0 mils has been provided between the UO_2 pellets and the APM tube wall. Pellets were air gauge inspected, individually, prior to loading.

After bonding the bottom end closure, the required length ($\simeq$ 32 in.) of weighed fuel pellets are manually loaded into the fuel tube, together with end spacers and springs. Stainless steel tube end spacers are used to provide space for the accumulation of fission gases. Dished-out molybdenum spacers, adjacent to each end pellet, assure that center melting of UO_2 pellets does not cause melting of adjacent metal components. An Inconel spring is used to hold the fuel pellet loading stationary, during storage and handling. This practice permits the expansion of pellets and prevents their separation during handling.

Sealed rods, containing fuel and 1 atm helium, which pass thermal cycling and hot helium leak tests are ready for final assembly into the multi-rod fuel element. Helium filling is accomplished, either by bonding in a helium filled glove-box or by evacuating and back filling with helium through the hole drilled in the top end plug; fusion welding is then employed to seal the hole.

In each prototype test element, thermocouples are attached to fuel rods, and are also located in the coolant channels, in order to monitor both cladding surface and coolant temperatures. In some cases, flow and fission gas pressure measuring devices have been incorporated.

Fuel rods are positioned in end support plates; the identity and respective position of each rod are recorded. The rods are then locked into position with bottom end plug locking pins of Type 304 stainless steel. The fuel bundle is inserted into the 304 stainless steel fuel box, and the sides of the fuel spacer support plates are aligned to pre-drilled holes in the fuel box. Support plates are then plug-welded to the box. Thermocouple leads and other conduits are drawn through the head extension diverter assembly, and coolant channel thermocouples are also attached. The fuel element end pieces, made of machined 304 stainless steel, are then aligned and welded to the fuel section in a specially constructed rotating alignment fixture. Thermocouple splices and connections are made, with the leadout thermocouple wires being housed in a 304 stainless steel process tube welded to the head extension.

Use has been made of the APM tube fins to attach 20-mil OD sheathed thermocouples (the Chromel-Alumel wires are 3 mils in diameter). This is accomplished by splitting the fin with a 10-mil thick milling cutter, laying the wire in the formed groove, and then folding over the fins to securely hold the wire in place. This method has been found to be superior, in all respects, to other attachment techniques, such as tack welding or brazing of bare wires directly to the tube surface. In addition, this method produces no deleterious metallurgical effects on the M257 cladding.

Because of the high electrical resistance of 3-mil diameter Chromel-Alumel thermocouple wires, it has been found necessary to make a splice of this wire, for leadout purposes, with 10-mil wire housed in 63-mil stainless steel sheathing. Splicing by silver brazing is made, so as to yield a completely leaktight junction.

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