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FABRICATION OF PROMETHIUM SESQUIOXIDE HEAT SOURCES BY HIGH ENERGY RATE FORMING

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FABRICATION OF PROMETHIUM SESQUIOXIDE HEAT SOURCES BY HIGH ENERGY RATE FORMING

By

N. C. Davis and D. W. Brite

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Chemistry and Metallurgy Division

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ABSTRACT

High Energy Rate Forming (HERF) has been used as a fabrication method for the densification of encapsulated promethium sesquioxide. The development and characteristics of the HERF process are presented. Simulated radioisotope materials have been used to develop capability for controlled dimensional integrity, and promethium sesquioxide has been densified in various metal claddings for compatibility testing.

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INTRODUCTION

The Division of Isotope Development (DID) of the Atomic Energy Commission (AEC) sponsored the Pacific Northwest Laboratory (PNL) in a program to develop methods for producing promethium sesquioxide heat sources. Fabrication methods that have been evaluated include:

- Cold pressing and sintering[(1)]
- Hot pressing[(2)]
- Slip casting[(3)]
- High energy rate forming (HERF).

As a part of the source development program, High Energy Rate Forming (HERF) has been developed as a process to densify encapsulated sources of promethium sesquioxide. With a single impact, HERF densifies the core and maintains the integrity of the encapsulation to yield a power source with intimate fuel-to-cladding contact and a close approach to theoretical density.

Although other fabrication processes are preferred in applications requiring extremely accurate dimensional uniformity, various experimental HERF techniques with special tooling for dimensional control have proven successful for specific applications. In the cases where relatively thick-walled cladding is a part of the design, minimum dimensional control tooling is needed. The HERF process has served as an alternate to pressing and sintering in the major promethium application programs at PNL. It has been used extensively in the densification and encapsulation of compatibility capsules. The intimate metal-fuel contact and high density are desirable features in these compatibility studies.

SUMMARY

High Energy Rate Forming has been developed as a fabrication method for the simultaneous densification and encapsulation of promethium sesquioxide. The process may be considered as a simple closed die forging operation to densify materials, or as an extremely complex process with many stress factors affecting the final shape and integrity of a heat source. The isotope is subjected to high impact pressure and temperature in a closed die arrangement. The radioisotope fuel compound, completely contained in a primary or compatibility cladding, is encased in a secondary mold serving as an energy transfer medium. This mold, a sacrificial billet component, also serves as ductile safety containment during impaction. The mold is readily removed by machining or chemical dissolution after HERF. The HERF process has been used in the fabrication of promethium compatibility capsules both in single and multi-capsule impactions. The high density, and the intimate interface contact between the fuel core and the cladding are desirable for optimum compatibility conditions.

Although the HERF process produces maximum fuel power density, it does not have the dimensional control capabilities of more conventional fabrication processes. To maintain the best possible dimensional control of a capsule and uniformity of the cladding, some of the HERF parameters to be considered are:

- Fuel core initial density
- Cladding thickness
- Cladding ductility at impaction temperature
- Capsule shape
- Tooling arrangement
- Impaction pressure and temperature.

These major parameters and their associated characteristics must be evaluated for any heat source encapsulation design.

For many conventional right cylinder shapes requiring relatively ductile thick-walled cladding, the HERF process will yield a high integrity, dense, and intimately bonded heat source capsule with minimal developmental effort. Minor surface machining of the HERF processed shape may be necessary, depending on the final application. In the cases of thin-walled or more complex shapes, the extent of required developmental effort must be compared to the need for end product advantages. HERF in some cases may be the most attractive fabrication method.

HIGH ENERGY RATE FORMING (HERF)

High energy rate forming as performed by a pneumatic-mechanical machine uses the energy stored in a compressed gas to accelerate a ram to high velocity. The kinetic energy in the moving ram results in deformation when a workpiece is impacted.

The high energy rate forming machine (Figure 1) used in this development work has a rated capacity of 145,000 ft-lb of energy with a 12-in. stroke and 2000 psi fire pressure. By using increments of this available fire pressure, dependent on the item to be impacted and the selection of tooling, effective pressures in excess of 300,000 psi have been developed.

The mold or impaction container with the fuel core evacuated, sealed, and heated is placed in the HERF machine's impaction die. Activation of a trigger valve allows compressed gases to drive a one-ton punch assembly into the closed die containing the heated billet (Figure 2). Rapid deceleration of the one-ton punch results in the extremely high-rate forming pressures. The very short initial pressure pulse, in the range of 0.6 to 10 msec, depends on the billet density and composition. Machining and/or chemical dissolution of the container,

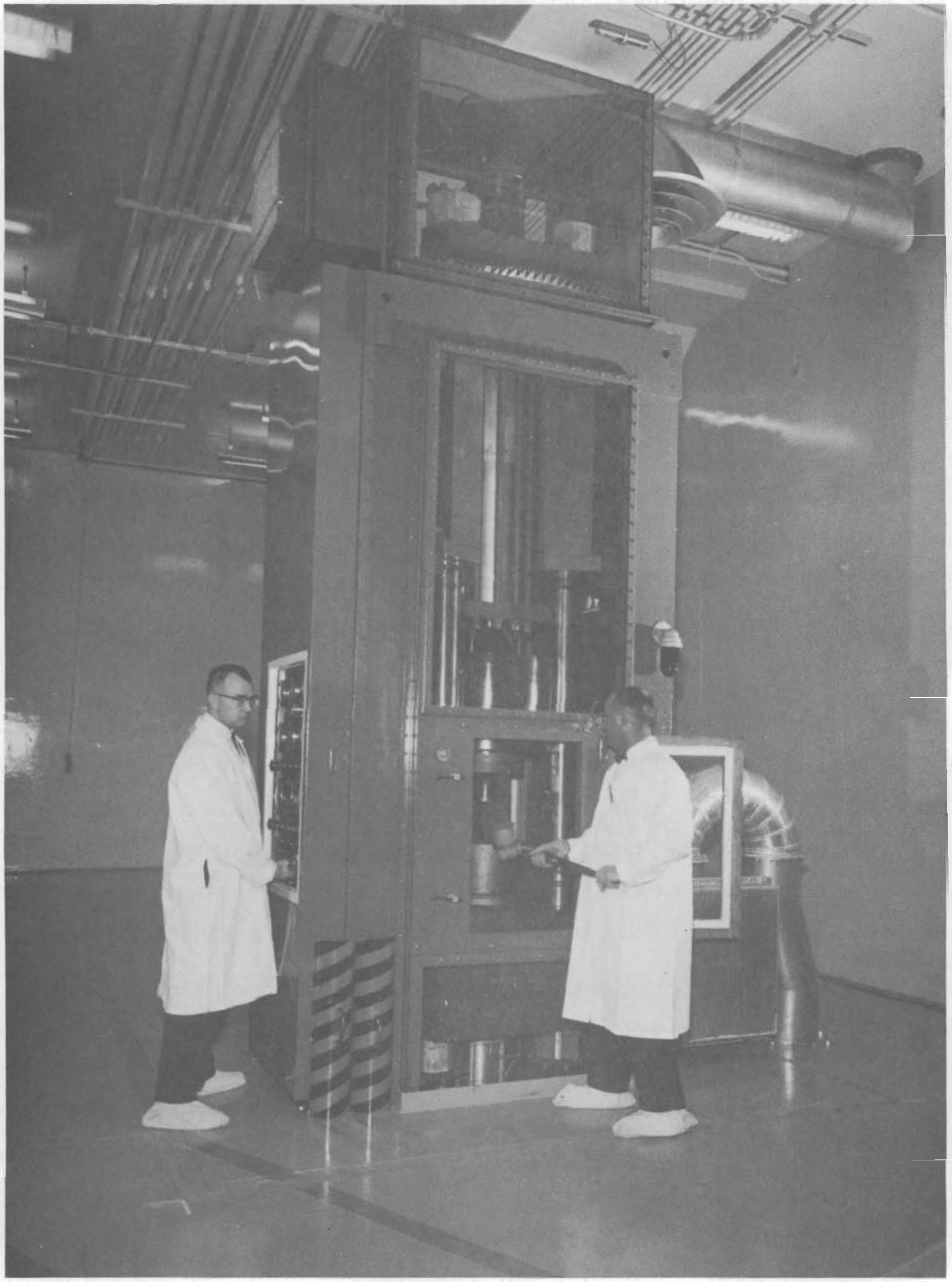

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FIGURE 1. High Energy Rate Forming Machine (General Dynamics Model 1220-B-Dynapak)

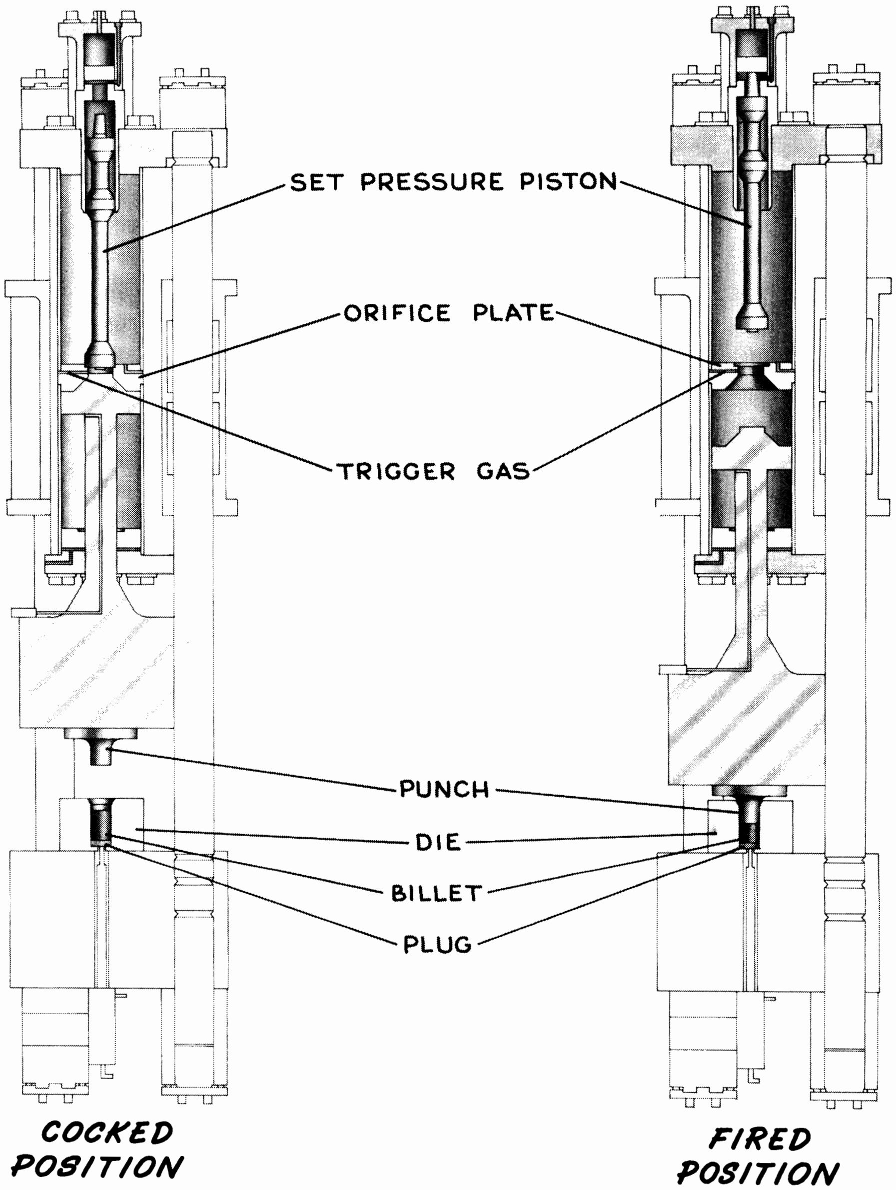

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FIGURE 2. HERF (General Dynamics) Machine Schematic View

molds, and other sacrificial shape forming components is usually required to recover the highly dense compact heat source from the impacted billet.

The HERF process, depending on the specification or end product requirements, can be extremely complex. A heat-source configuration may vary from a simple right cylinder, singly clad capsule to complex shapes consisting of several components constructed of several different materials. If the final shape and integrity of the heat source is to be predicted, the various stress factors arising in this process must be considered.

Stress factors (Figure 3) and other significant phenomena involved in the HERF process (also known as pneumatic impaction) are listed in the order of their appearance:

1. Chemical reaction, solid solution formation, alloying, diffusion bonding, gaseous decomposition, sintering, and other phenomena occurring during preheating of the impaction billet prior to pneumatic impaction.
2. Temperature gradients developed in billet due to nonuniform heating or to uneven cooling in die before impaction.
3. Axial compressive stress as punch impacts against billet.
4. Densification of low density portions of billet and thickening of dense components as billet length decreases during impaction. Frequently, this thickening of initially thick-walled containers produces primarily radial, rather than axial compressive stress on the low density central portion of the billet.
5. Elastic expansion of die as pressure builds up to maximum pressure of approximately 300,000 psi.

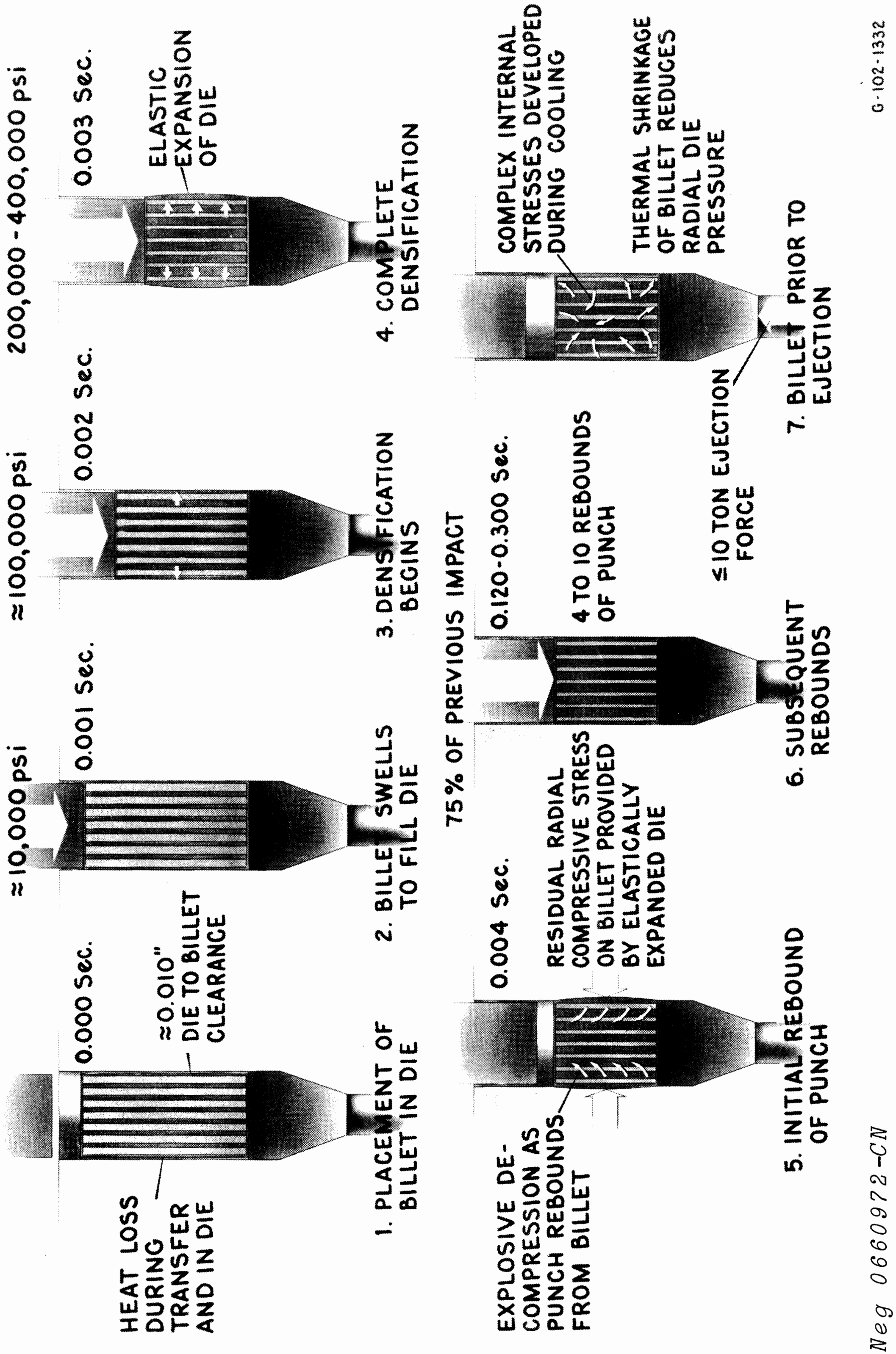

FIGURE 3. *Stresses During HERF*

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6. Explosive decompression as punch rebounds from billet abruptly reverses axial stress in billet from compression to tension due to springback as pressure falls from 300,000 psi to zero in less than 1 msec.
7. Radial compressive stress is applied to the billet by the elastically expanded die, while no axial pressure is simultaneously applied to the billet by the punch.
8. Reapplication of axial compressive stress and phenomena (Steps 4 through 6 in this list) as punch recoils and impacts against the billet a total of from four to 10 times at intervals of approximately 30 msec. Each impact results in a die pressure approximately 75% as great as that developed during the preceding cycle.
9. Rapid quenching of the billet as heat flows into the die develops a temperature differential from the edge to the center of the billet, which causes thermal stresses in the billet.
10. Gradual decrease in radial compressive stress applied by elastically expanded die as billet shrinks on cooling.
11. Elimination of compressive die stress as the billet is ejected from the die.
12. Complex residual stresses developed during cooling as different components in billet shrink different amounts due to differences in thermal expansion coefficients.
13. Disruption of stress balance in billet as impaction container is removed by machining or chemical dissolution.

Frequently, it is advantageous to include an annealing step following impaction to minimize the consequences described in Step 13. When the impacted materials are relatively ductile, some of the preceding considerations may be ignored. However, in the case of the radioisotope promethia ($^{147}Pm_2O_3$), these factors as well as the contamination potential and self-heating effects must be considered in the billet design to optimize safety, dimensional control, and quality assurance of the final heat source.

The degree of densification and dimensional uniformity of the final product dictates the extent of tooling techniques and requisite control of impaction parameters. A typical HERF process for the simultaneous densification and encapsulation of a promethium-147 heat source could include:

1.	Select Primary Cladding	Compatibility considerations. Radiation shielding characteristics. Deformation properties.
2.	Load Pm_2O_3 Fuel	Step press into container. Press and load with pellets. Sintering to improve core density may be included.
3.	Closure	Weld (high integrity), and location critical to withstand high energy-rate deformation.
4.	Assembly Impaction Billet	Compatible with capsule cladding. Safety containment consideration.
5.	Pre-Heat	Time and temperature compatible with materials.
6.	Impaction	Major factors affecting pressure include: fire pressure, temperature, ductility, and initial density.
7.	Finishing	This includes the ejection from the die, cooling, and final machining to desired finish, size or shape.

These seven major steps could be considered as a superficial outline of the complex HERF process.

DEVELOPMENT OF HERF PROCESS

Technology developed during the densification of ceramic nuclear fuels led to the first concept of the HERF process for encapsulating radioisotope heat sources. This early work fell into two categories:

- Densification of oxide particles for use in fuel element fabrication.
- Simultaneous densification and encapsulation of ceramic fuel materials.

Literally tons of ceramic materials (uranium dioxide, thorium dioxide, and mixed uranium-plutonium dioxide) have been densified for fabrication into nuclear reactor fuels.[4] Dimensional control of the impacted containers was of little concern, while high density and fuel containment was of great importance. The nuclear fuel materials were loaded in thin-walled stainless steel containers. These containers were welded, heated while outgassing the contents, and impacted at pressures up to 300,000 psi. The oxide fuel was recovered in the form of highly dense particles for use in subsequent fuel element fabrications.

During the extensive work in fuel material densification by HERF, it was noted that the side walls of ductile heavy wall containers would thicken on impaction and maintain a high degree of uniformity. Containment of the nuclear fuels during impaction was critical and it soon became apparent that an impacted container could provide the required integrity of containment for the life of the fuel.

A thorium dioxide fuel element (Figure 4) was simultaneously densified and encapsulated in a single impaction. A

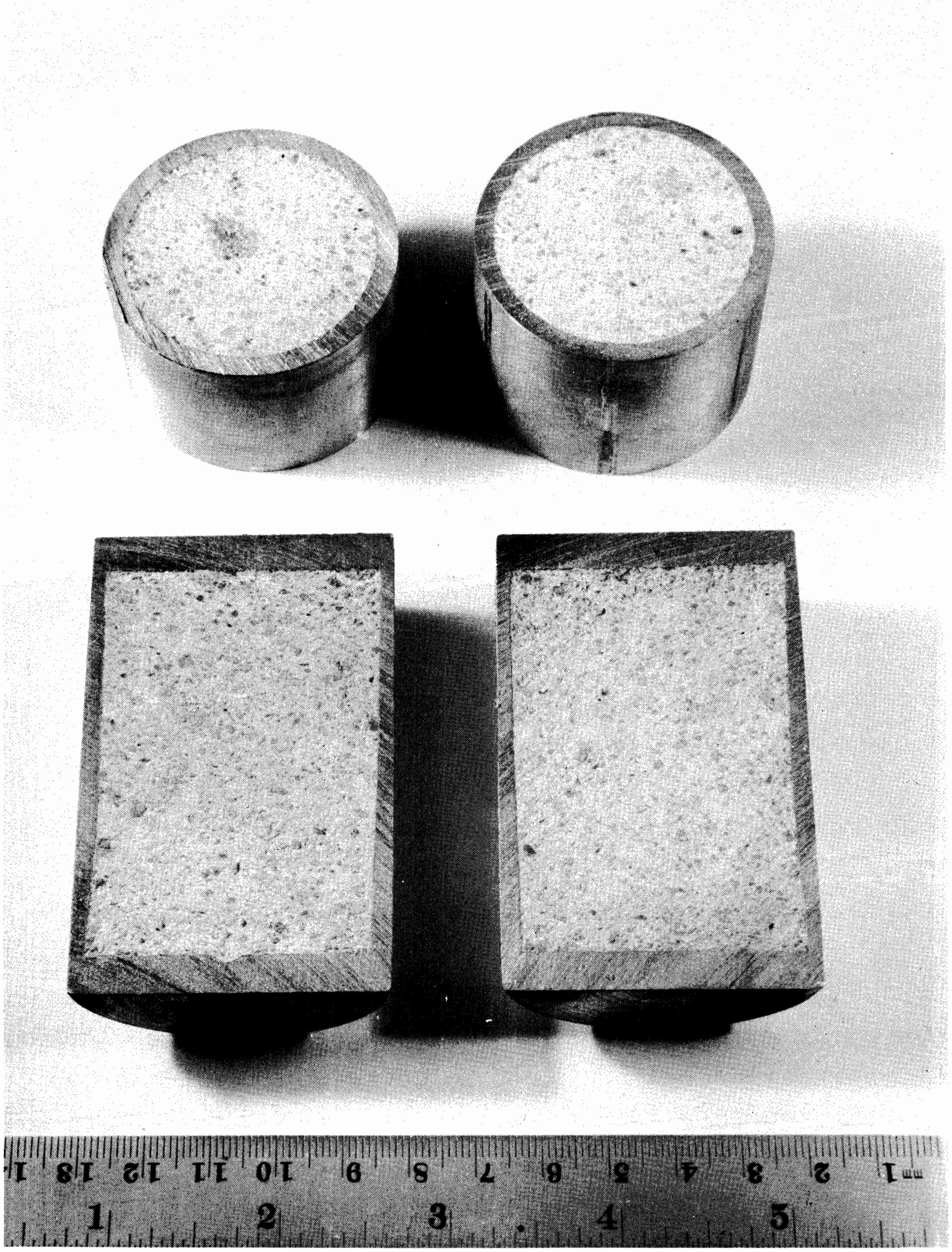

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FIGURE 4. Simultaneous Densification and Encapsulation of Thorium Dioxide

heavy-walled aluminum impaction mold was used as the cladding for the thorium dioxide powder. An aluminum powder end cap was pre-pressed and placed in position to complete the encasement.

This low temperature, high pressure impaction produced the first, extremely compact fuel element (98% TD) in a single HERF impaction. This fabrication technique was also successful using lithium aluminate fuel cores (Figure 5). The heavy-walled ductile aluminum container deformed uniformily and required only a minor machining operation to clean up the finished fuel element.

Some of the problems concerning dimensional control of the fuel core were first noted in an early experiment to encapsulate and densify uranium dioxide in a fuel form with an internal annulus (Figure 6). The internal annulus was maintained during HERF by use of an expendable mandrel dissolved by acid in a finishing operation. In this experiment, the thinning of the cladding at the transition point between the side walls and the end cap was believed to be caused by the radial pressure developed during impaction. The fully dense end cap, by failing to compress as much as the porous fuel core, allowed the fuel to migrate to the intersection between the side wall and the end cap. On closer examination, this thinning of the clad at stress points was noted in previous impactions. Although there were definite container design problems to be resolved, these early experiments with nuclear fuels indicated that a HERF fabrication method might be quite applicable for an extremely compact isotopic heat source.

SIMULATED ISOTOPE HEAT SOURCES

Simulated heat sources using nonradioactive material as stand-ins for the core have been produced with uniform walls and cores approximating theoretical density (99% TD).[5] The experience gained from the impaction of nuclear materials in

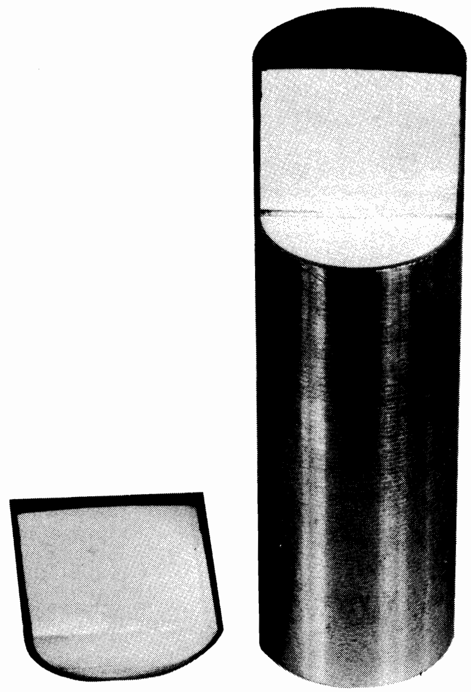

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FIGURE 5. Lithium Aluminate HERF Fuel

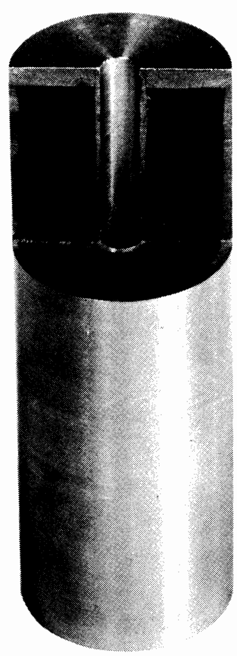

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FIGURE 6. Uranium Dioxide Encapsulation

thick-walled containers indicated the requirement for a special radius at the side wall-end cap transition point. During impaction, the internal shape of the container returned to nearly a right angle configuration. This modification added structural strength to the container at its otherwise weakest section and allowed uniform thickening of the capsule walls during impaction. With the capsule's wall remaining relatively thick (minimum approximately 0.125 in.) and the compaction chiefly in the longitudinal rather than the radial direction, reproducible dimensional control could be obtained. The other major factors affecting the final capsule's dimensional integrity include:

- The initial green core density.
- Selection of cladding material.
- Impact pressure and temperature.

In many cases, materials difficult to obtain or fabricate are required for compatibility or radiation shielding. These types of materials may require the use of a thin-walled primary cladding. Feasibility has been established with the HERF process to use thin-walled primary cladding by adopting one or more of the following techniques:

- Semi-isostatic molds.
- Compensating dimensional control dies.
- Double or secondary impaction.
- Multi-cladding impactions.

SEMI-ISOSTATIC IMPACTIONS

Some development work has been done with semi-isostatic impaction molds. By using an extremely plastic metal as the impaction mold, a nearly isostatic condition exists on impaction. In a low temperature experiment, lead was used as the highly plastic energy transfer medium (Figure 7). In a temperature range before a metal melts, an extremely plastic

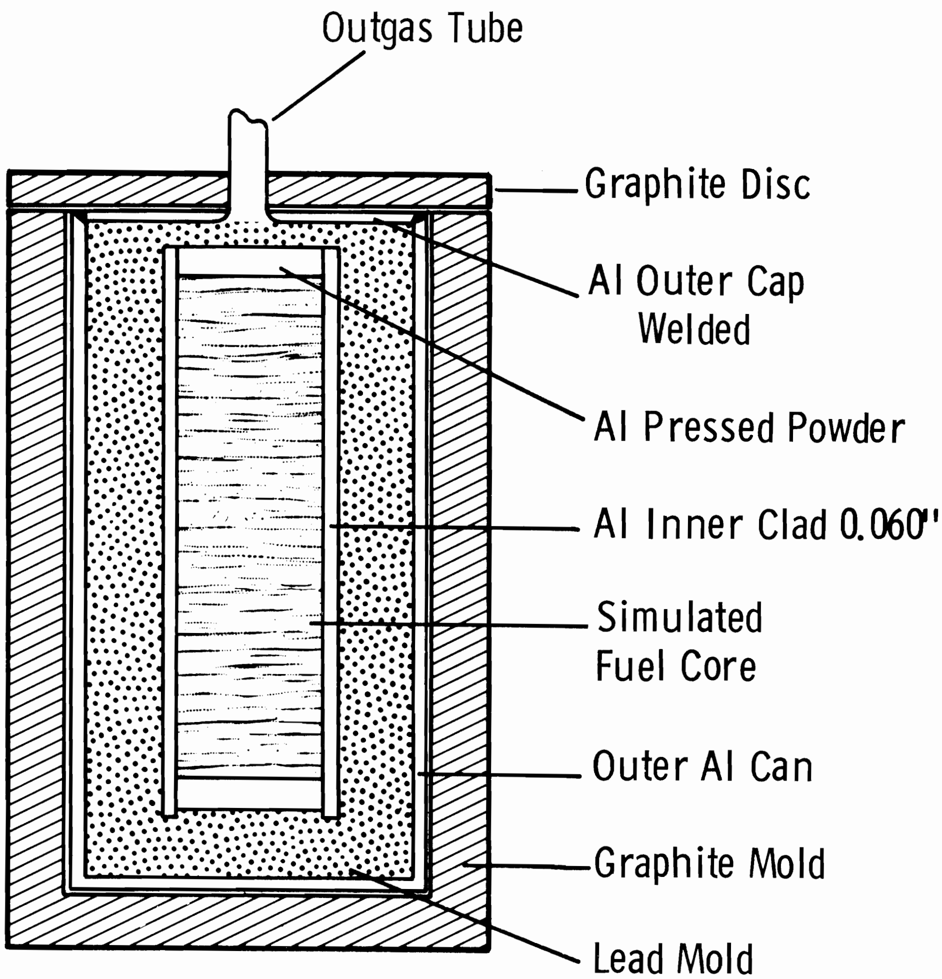


FIGURE 7. HERF Semi-Isostatic Impaction Assembly

condition exists. The metal in this condition behaves almost as fluid under HERF and a uniform radial and length reduction will occur on impaction. This reduction, which densifies the fuel core material, can be controlled to some extent by such variables as initial density, temperature, and impaction pressure. The energy transfer medium can be a substance other than a metal. Glass, for example, has been used successfully in experiments at temperatures up to approximately 1000 °C. Since the pressure is semi-isostatic, near-symmetrical reductions will occur. The fueled capsule prior to impaction requires a uniform cross section density to prevent undesirable hourglassing effects on the side walls and concave dishing of the end caps.

COMPENSATING DIMENSIONAL CONTROL IMPACTIONS

The compensating dimensional control die arrangement (Figure 8) is made with a tapered hourglass relief so that the reduced section is adjacent to the central portion of the fuel core in the mold assembly. This design compensates for the tendency of the fuel core to deform to an hourglass configuration. The billet void space and location must be precise to

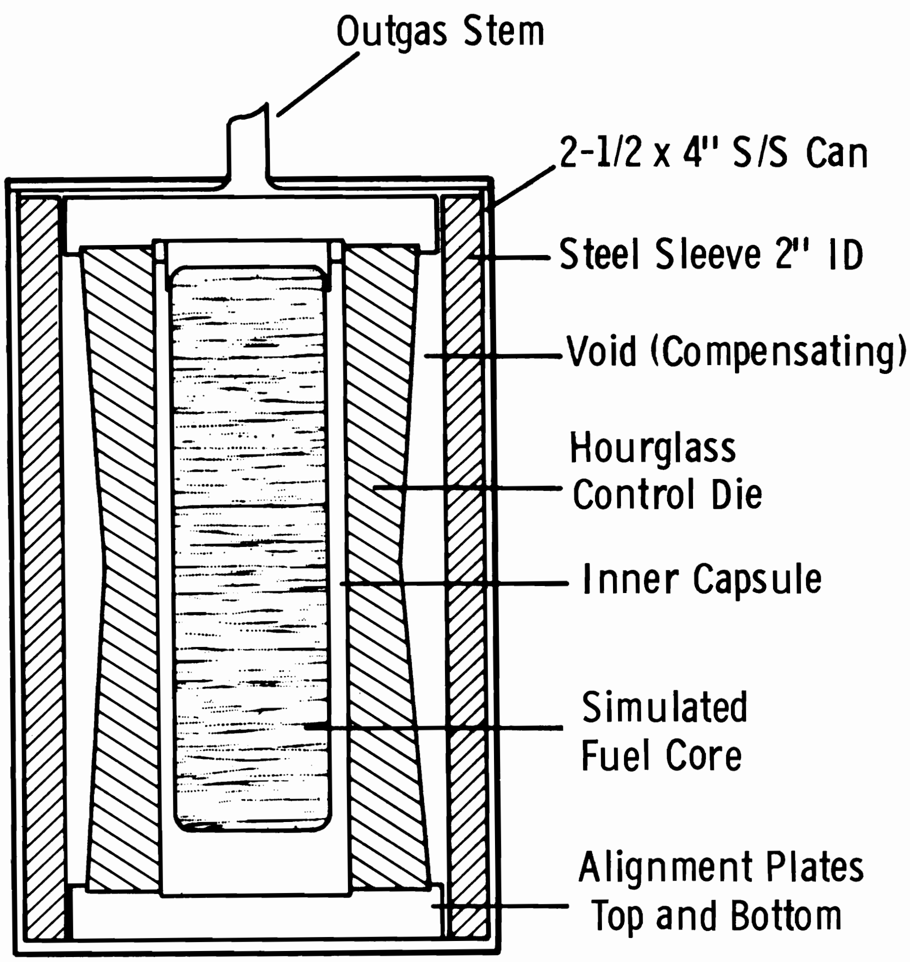


FIGURE 8. HERF Impaction Assembly - Hour Glass Control Tooling

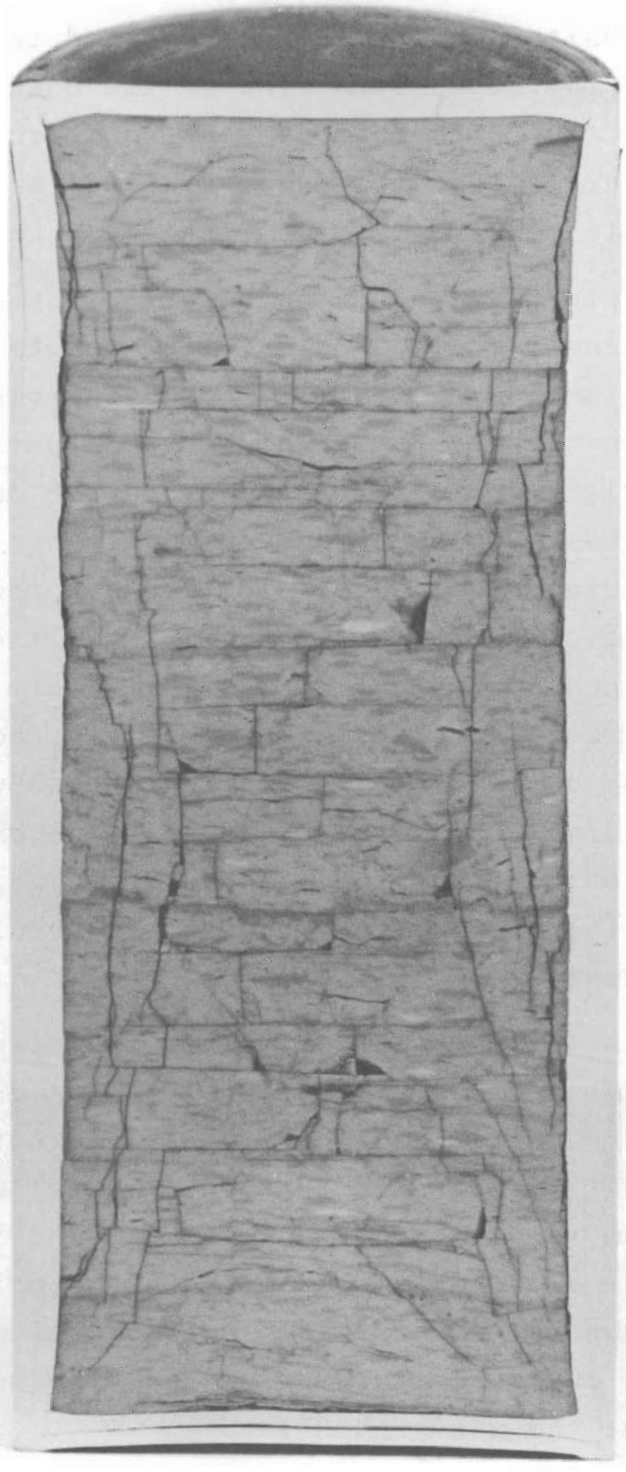
prevent an opposite (barreling) effect, and the void relief is calculated to provide uniform flow of the mold metal during the impaction reduction. The end caps are dome shaped to prevent migration of the fuel at the side wall-end cap transition area, and to eliminate concave dishing of the end caps.

The HERF process, using the compensating dimensional control die technique, was a candidate for the final fabrication of the promethium-147 fueled multi-clad radioisojet application.[6] Extensive developmental work was performed using stand-in materials for both tne core and the multi-cladding. The technique was developed to the extent that minimum cladding shielding could be predicted and reproduced. With diameter variations of less than 0.010 in., the HERF capsule could be machined to a final precise diameter. Although an irregular fuel core-shielding cladding interface was inherent to the process, nondestructive examination showed specified shielding cladding thickness. A destructive examination (Figure 9) of one of the stand-in capsules indicated areas where modification to compensating tooling could have improved the final product.

In the case of the micro thruster radioisojet application, the HERF process was abandoned when problems developed with seam welded components. Both the inner and outer cladding materials were fabricated from a powder metallurgy product, rolled and seam welded to form capsules. The integrity of these longitudinal seams could not be assured in the HERF process. Fortunately, the assembly of sintered $^{147}Pm_2O_3$ pellets in the designed cladding provide the required power density without the use of HERF.

DOUBLE HERF IMPACTIONS

A capsule was fabricated using the hour glass compensating tooling concept. The densified capsule was inserted into an



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<u>*FIGURE 9*</u>. *Longitudinal Section of Samarium Oxide ($^{147}Pm_2O_3$ Stand-in) Heat Source*

impaction die arrangement, (Figure 10), providing a secondary special shaped containment. The second impaction of the assembly affected core density very little, since it was nearly theoretical after the first impaction. Due to the highly dense fuel core, very little deformation occurred and the secondary cladding was mechanically bonded to the primary cladding during the second impaction. The mild steel mold material was dissolved by acid and the final dome-ended (Figure 11) shape required minimum machining.

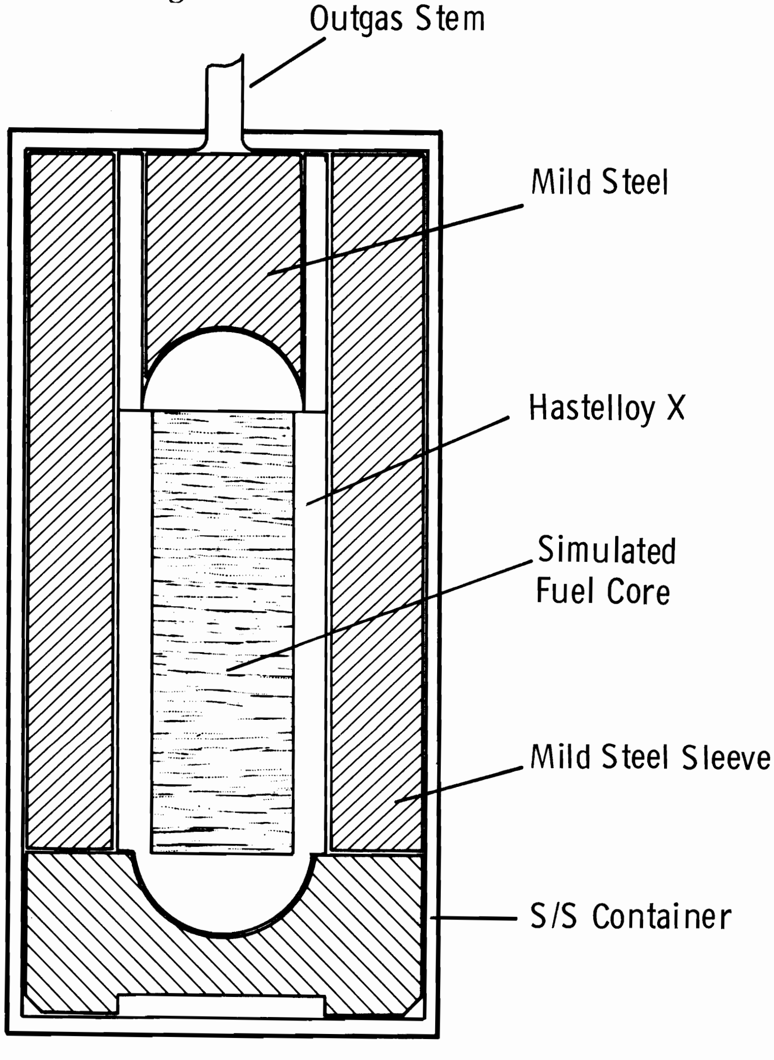

FIGURE 10. *HERF Impaction Assembly - Second Impaction Special Shape*

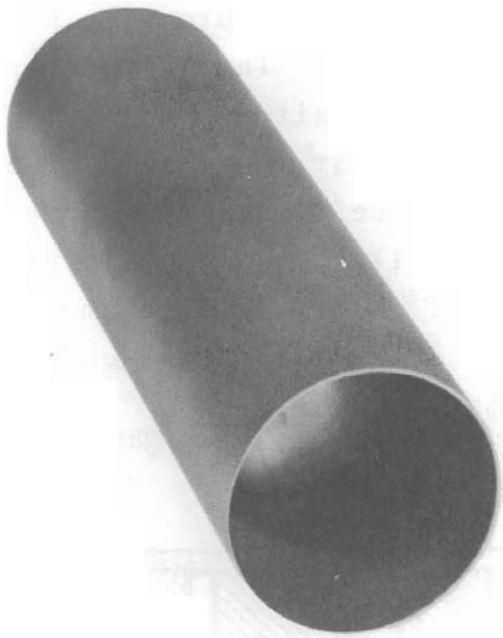

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FIGURE 11. Dome-Ended Heat Source

MULTI-CLAD HERF IMPACTIONS

Multi-clad simulated isotope heat sources have been fabricated by the HERF process by using several tooling techniques. The most important factors include:

- A high fuel core density.
- Claddings having similar deformation characteristics.

Use of a previously densified and clad fuel core, or an assembly of pressed and sintered pellets, assures a high starting density. An initial assembly of this type results in minimum distortion during the second impaction and maximum bonding of the components.

If initial high core density cannot be provided and excessive deformation is expected in a multi-clad assembly, the different cladding materials must have similar deformation characteristics at impaction temperature. The only exception to this general rule would be in cases where an intermediate cladding is formed from powder (Figure 12). In this experiment, the refractory metal powder in the second cladding could be heated only to the limits of the stainless steel primary cladding. Hence, less than optimum density was obtained in this intermediate containment cladding. This approach of multi-cladding (Figure 13) using powder for the intermediate claddings would require additional development to obtain high quality assurance.

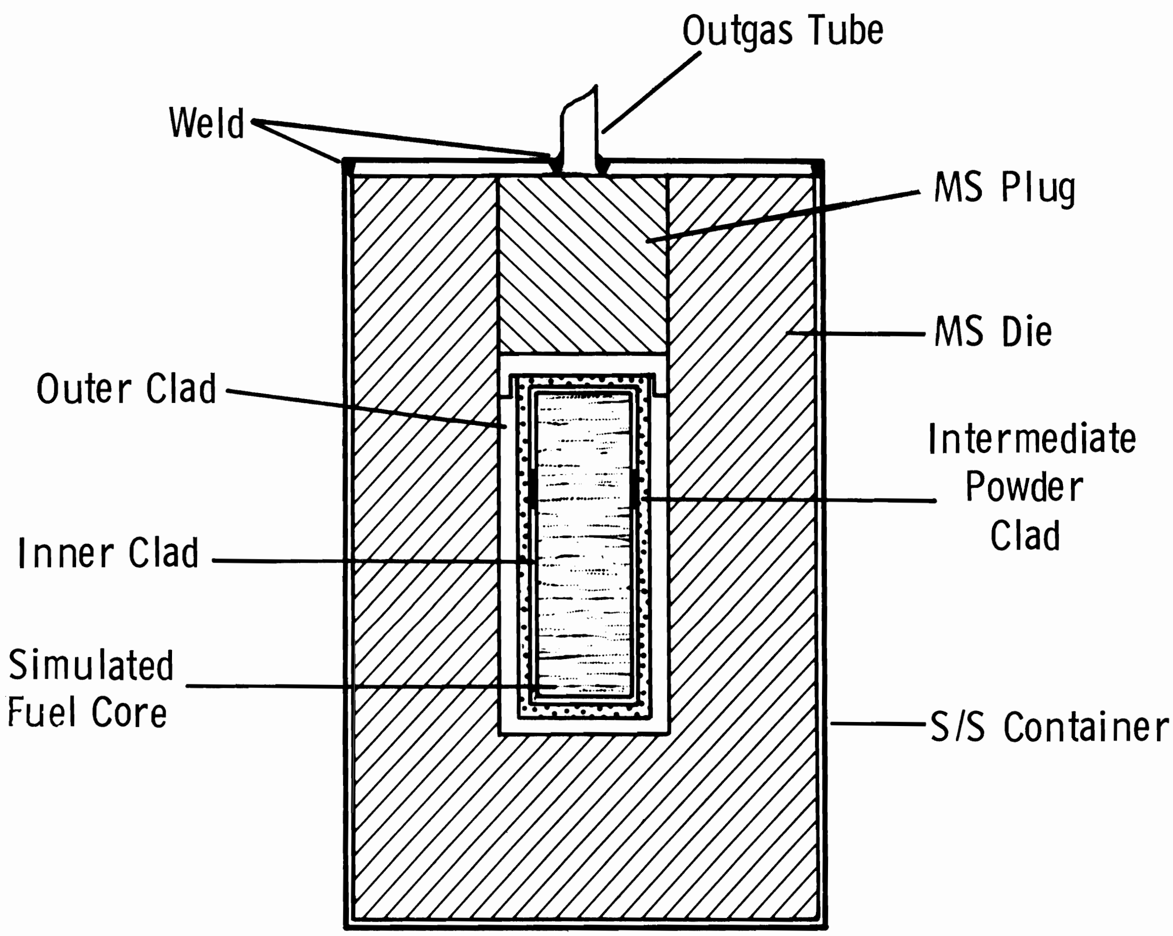
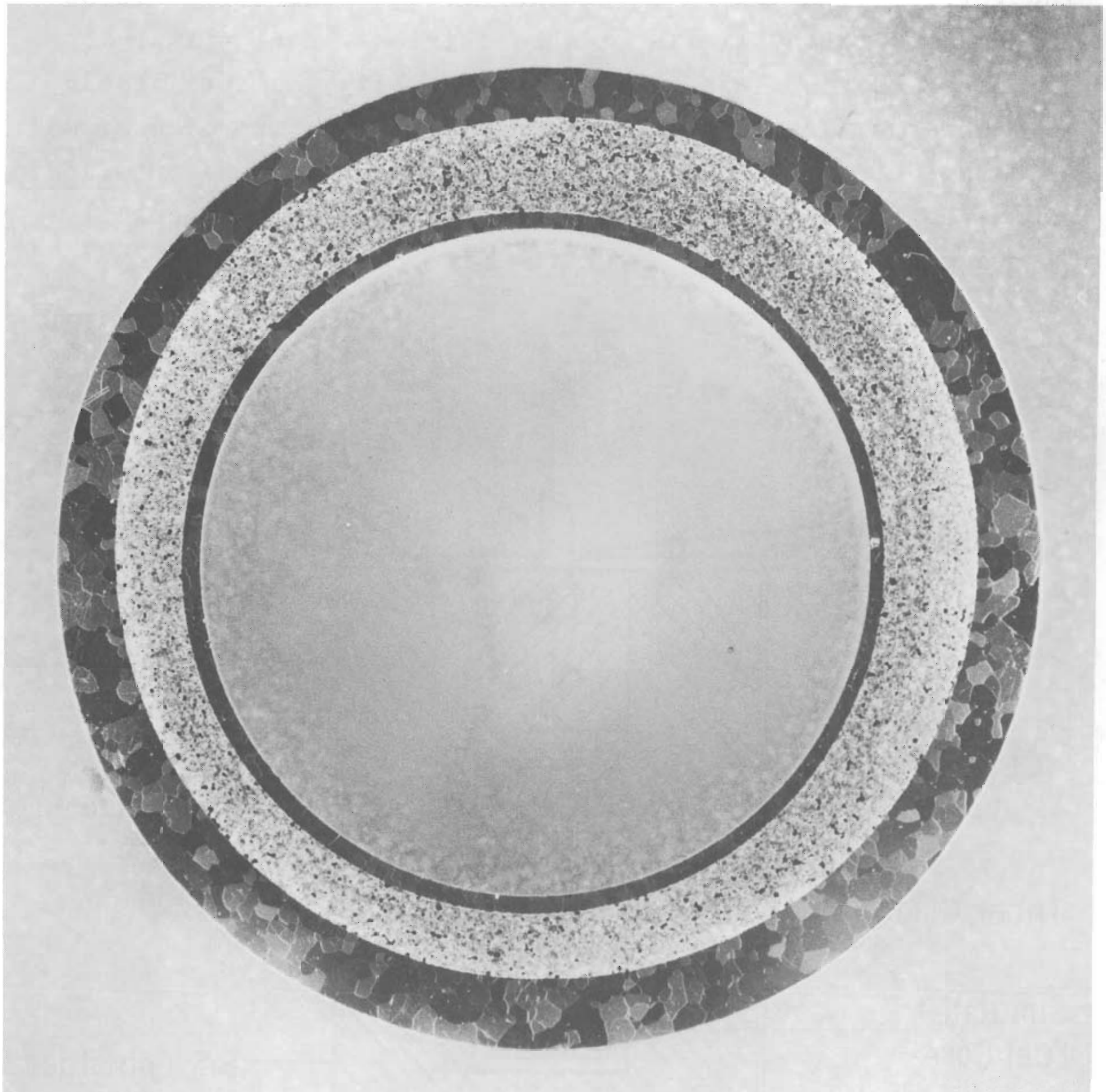


FIGURE 12. HERF Impaction Assembly Multi-Clad Encapsulation



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<u>*FIGURE 13.*</u> *Multi-Clad Simulated Isotope Heat Source with Intermediate Powder Clad*

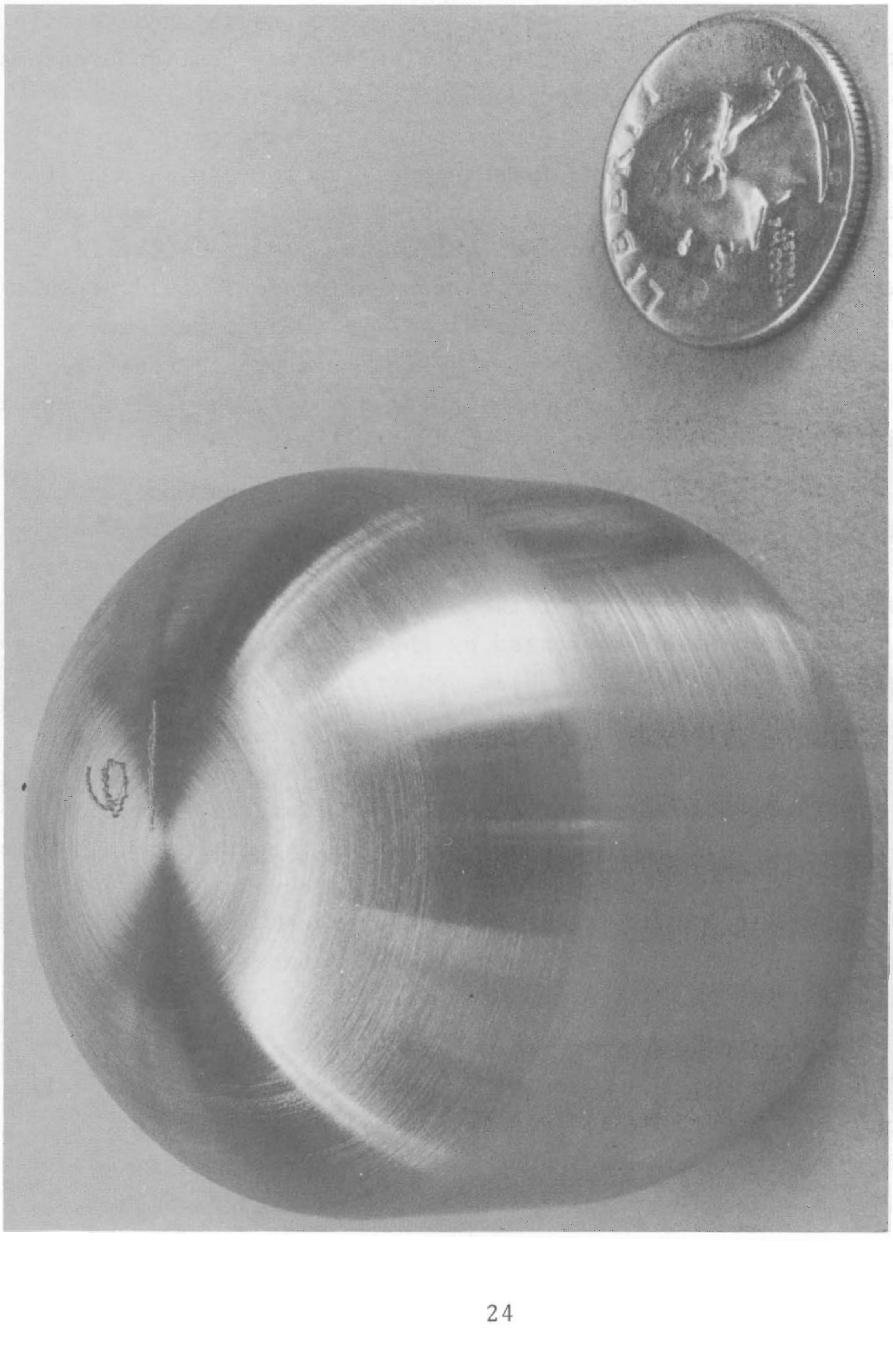
IMPACT TESTED HERF CAPSULES

The HERF fabrication method was evaluated as an alternate process in one of the major promethium-147 application programs at PNL.[7] In the Advanced Manned Strategic Aircraft (AMSA) program, a 60-W high strength, impact resistant capsule was designed, fabricated, and flight qualified for use as a heater in a navigation system. Preliminary process development and impact test results indicated that either a simple slip-fit assembly of sintered pellets or the HERF process would be suitable in this fabrication program. While the program time schedule dictated adoption of the simpler approach, capsules fabricated by the HERF process appeared to have improved impact resistance.

Five typical AMSA capsules were fabricated using samarium oxide as an inert stand-in for promethium oxide. A view of one of these capsules is shown in Figure 14. These dummy capsules were impact tested at the Sandia Rocket Test Site.[8] All of the capsules fabricated by the HERF process remained intact at velocities as high as 400 ft/sec against granite.

The HERF process could have been perfected with these heavy wall capsules to meet the dimensional requirements of the program. Due to the program time schedule, the tooling and components used in this evaluation of the HERF process were identical to those used in the slip-fit assembly. These tooling and component designs, although not ideal for the HERF process, were adequate for superficially comparing the processes.

The pressed and sintered samarium oxide fuel core was welded into a double-walled billet of 0.260 in. in total wall thickness. The assembled and welded billet was heated to 1100 °C and consolidated by HERF using pressures in excess of 200,000 psi. This consolidation process effected intimate



Neg 0670722-2-CN *FIGURE 14.* *HERF Fabricated AMSA Capsule* interfacial contact between the billet components (Figure 15), and increased the billet diameter about 0.060 in. Since this expected diameter growth was not compensated for in the components, losses ranging from 0.025 in. to 0.030 in. in the outer cladding thickness resulted during final machining to specified external dimensions. Due to the limited number of impact tests, final conclusions could not be made regarding wall thickness or the HERF process. The impact strength of the capsule was probably improved by the mechanical bonding of the cladding. Unfortunately, these points were not fully resolved, since the completely adequate power density of a slip-fit assembly for the AMSA capsule design obviated the need for further developing this application of the HERF process.

HIGH ENERGY RATE FORMING OF PROMETHIUM SESQUIOXIDE FUELED CAPSULES

The first $^{147}Pm_2O_3$ encapsulation at Pacific Northwest Laboratory was accomplished by the High Energy Rate Forming process. This process was subsequently used for numerous compatibility test capsules.

COMPATIBILITY CAPSULES

These first three capsules of $^{147}Pm_2O_3$ were processed in an impaction mold assembly (Figure 16), designed for handling safety and ease of recovery. The 7 g of oxide per capsule were pressed into the primary compatibility cladding, recanned in a secondary cladding, outgassed, and electron beam welded. These capsules were then placed in a heavy graphite sleeve and encased in Al_2O_3 filler material contained in a heavy steel sleeve and welded in a double-walled stainless steel impaction container. The impaction assembly was heated to 1200 °C while held under vacuum for 1.5 hr and was then impacted at 300,000 psi. Recovery of the three capsules was performed in a hooded lathe with relative ease without any contamination problems. The

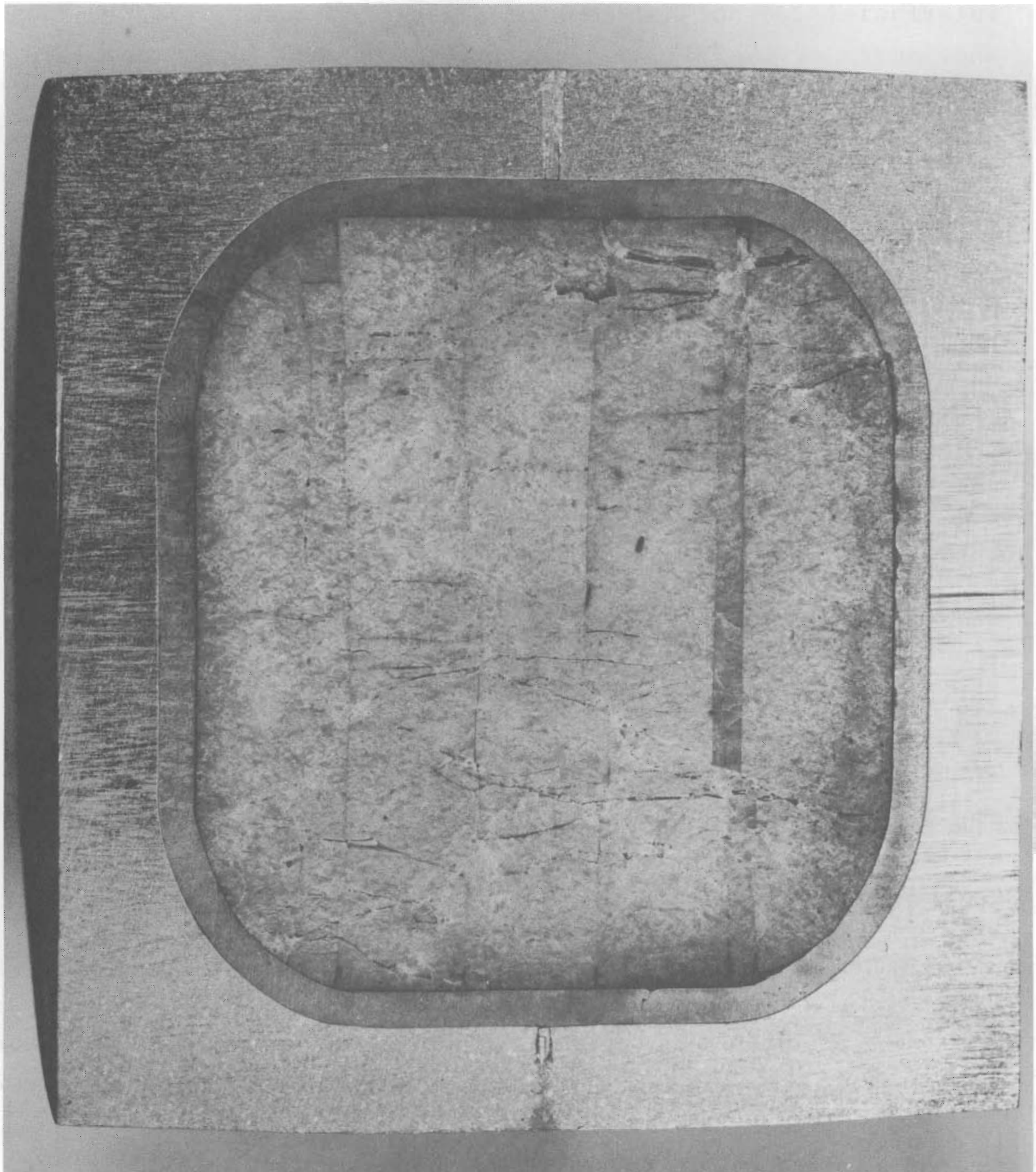


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FIGURE 15. Section View of HERF-AMSA Capsule Prior to Final Machining

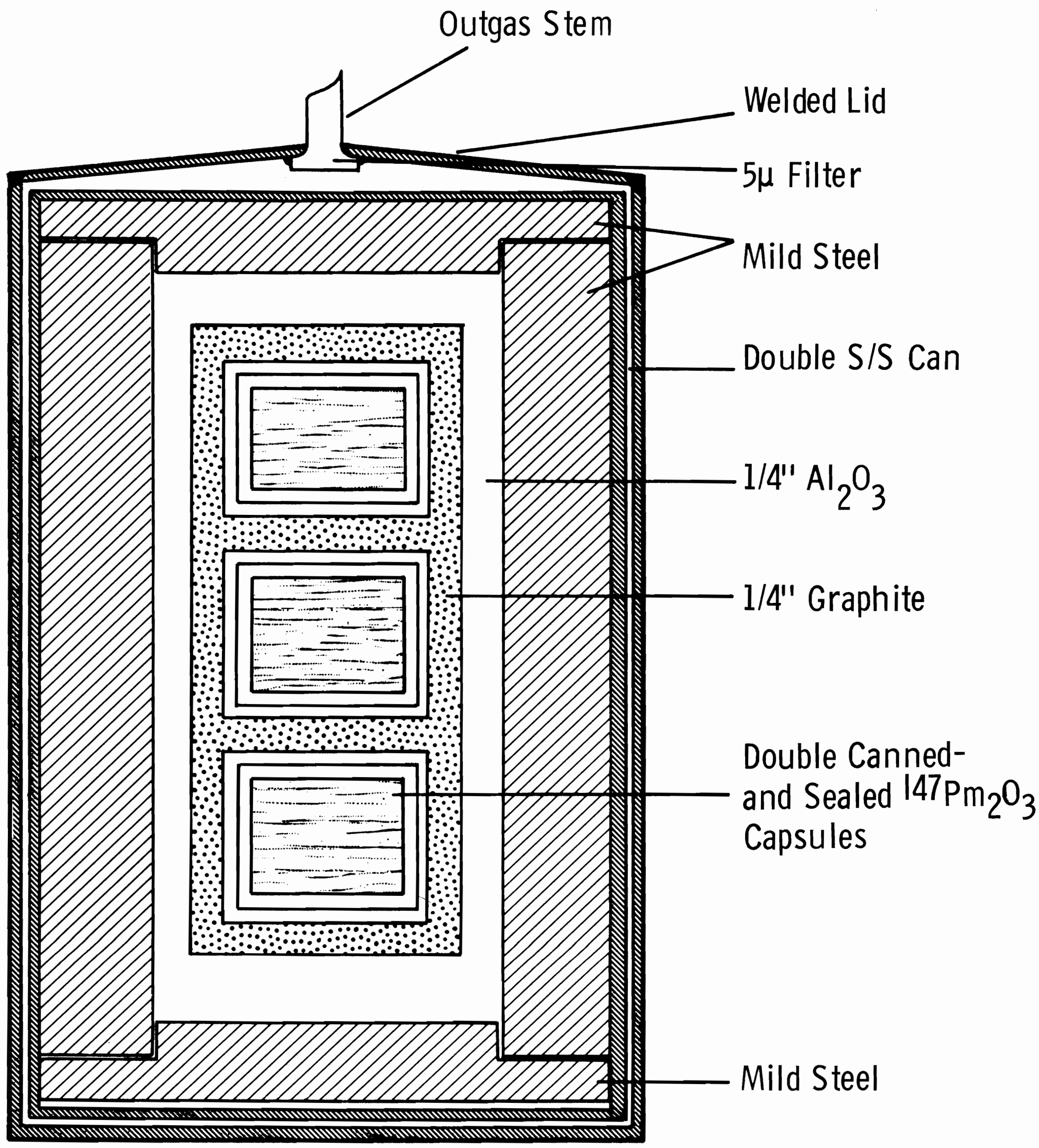

FIGURE 16. Impaction Assembly for $^{147}Pm_2O_3$ Compatibility Capsules

capsules (Figure 17) were grit blasted to complete the process. One of the capsules was later sectioned (Figure 18) and core density was determined to be near theoretical. Using this concept, dozens of samarium (stand-in for promethium) compatibility capsules have been processed and are being subjected to time versus temperature tests. Capsules fabricated by the HERF process have excellent interfacial contact between the core and metal cladding.

Another example of $^{147}Pm_2O_3$ compatibility test capsule fabrication by HERF[9] is illustrated in Figure 19. Five sealed capsules were encased in a 0.040 in. thick jacket of Type 304-L SS and placed in a massive mild steel mold. The assembly, representing approximately 60,000 Ci of promethium, was welded into a 1/16 in. wall Type 310 SS impaction container evacuated during heating through an outgas stem welded to the lid. A porous stainless steel filter was welded to the inside of the lid to protect the vacuum system from any contamination leakage of $^{147}Pm_2O_3$. The outgas stem was crimped off with a bolt cutter, sealing the can immediately before impaction. Strain gages attached to the HERF machine ram indicated a pressure of 200,000 psi on a recording oscillograph.

A power saw was used in an open-faced hood to remove the stainless steel outer container. The exposed mild steel mold was immersed in a nitric acid solution for complete dissolution of the expendable billet components. The five impacted capsules (Figure 20) were nondestructively tested by radiograph and helium leak detection. No material defects or questionable areas were found. Dimensional distortion produced by impaction is shown in Table 1. No special attempts were made to prevent distortions.

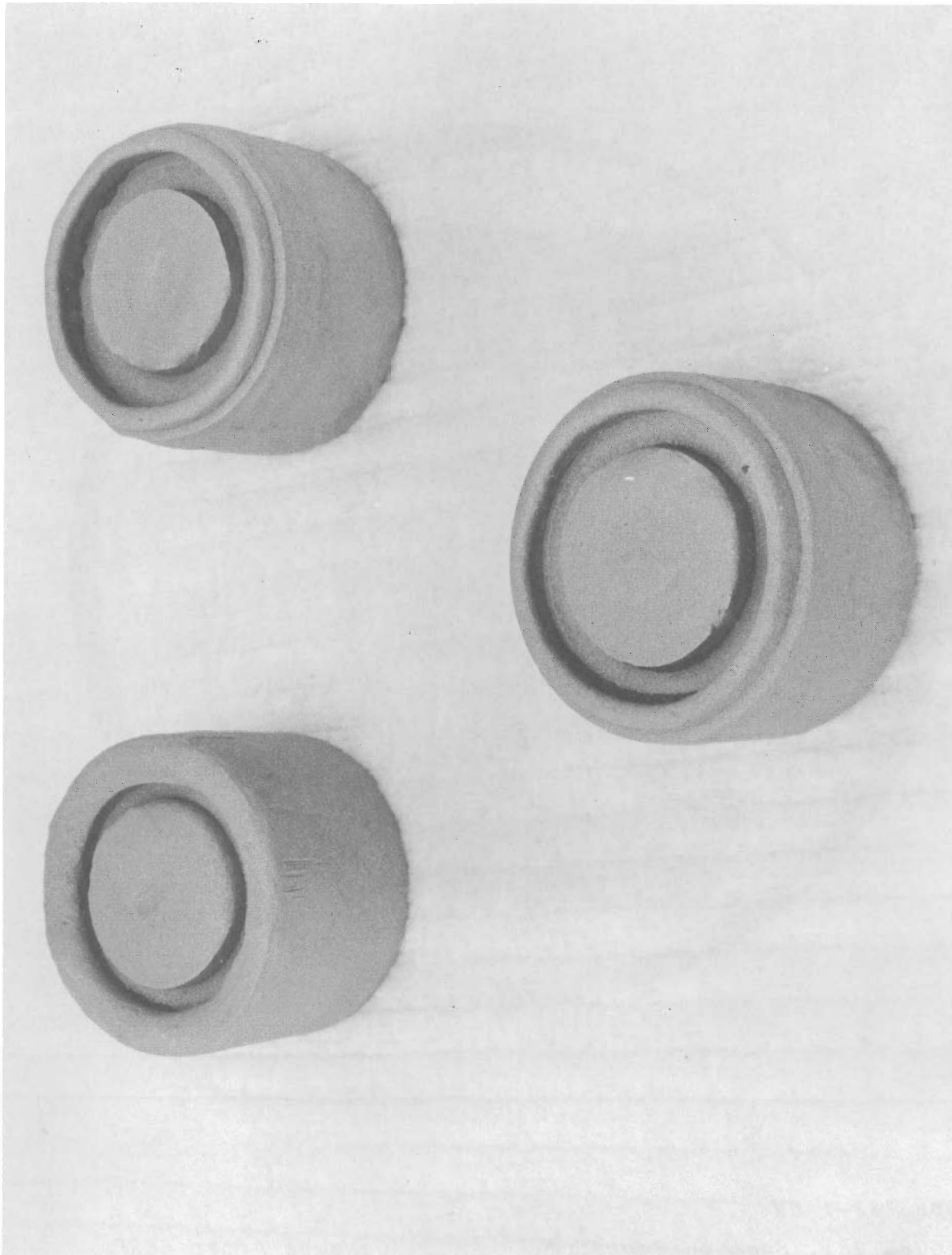

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FIGURE 17. Impacted $^{147}Pm_2O_3$ Compatibility Capsules

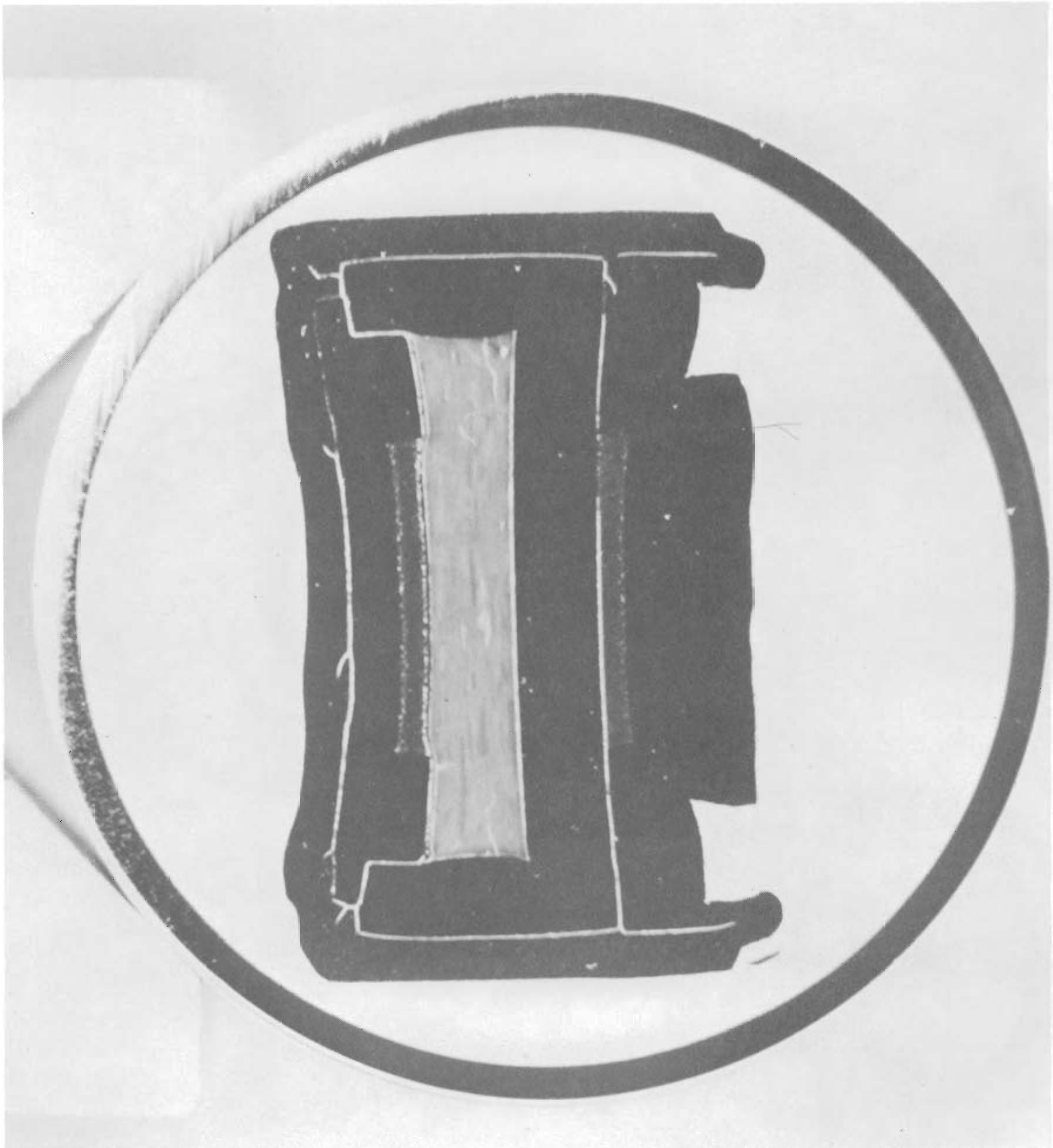

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FIGURE 18. Section Photo of $^{147}Pm_2O_3$ Capsule After HERF

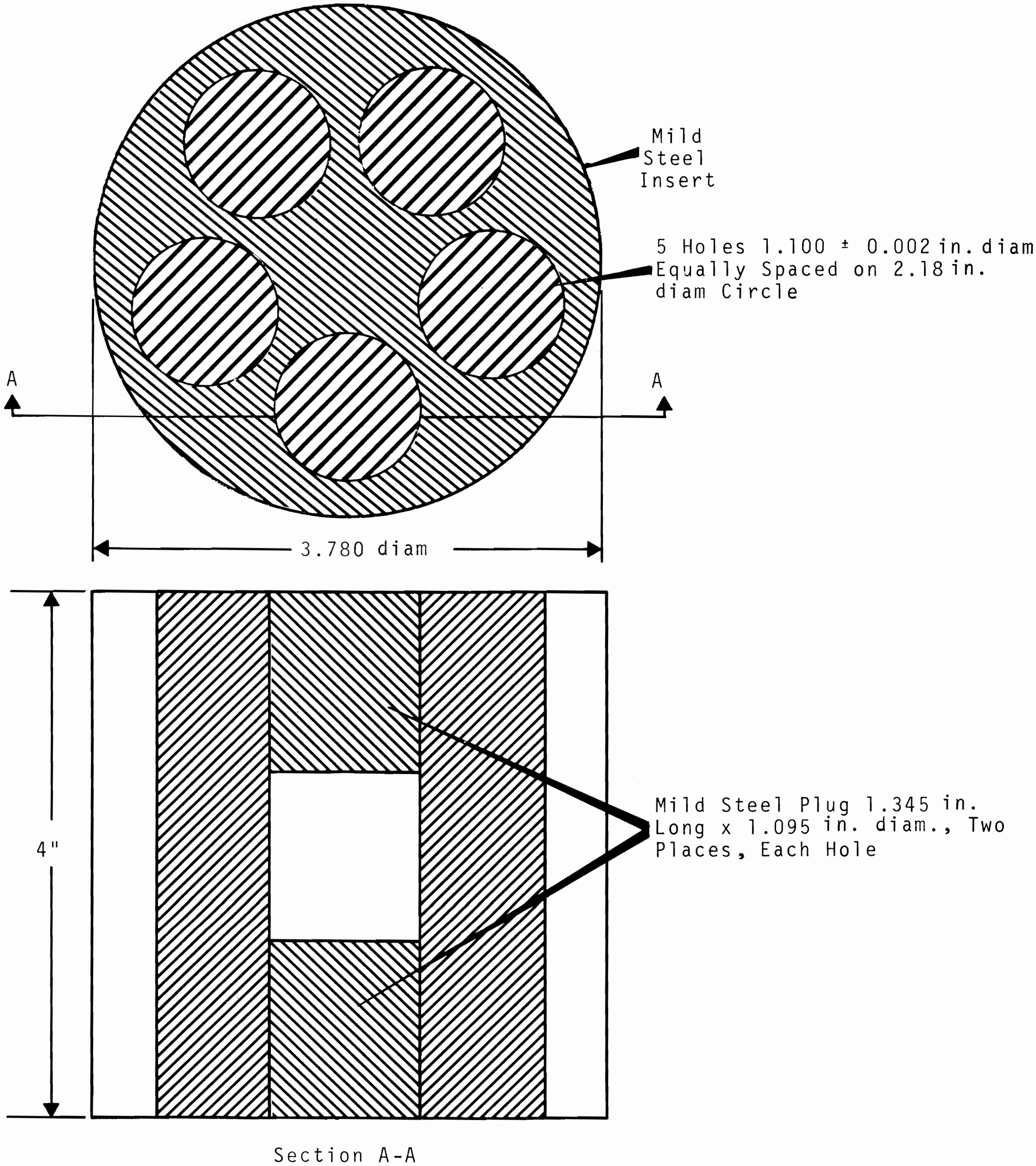

FIGURE 19. Impaction Assembly for $^{147}Pm_2O_3$ Compatibility Capsules Fabricated by HERF

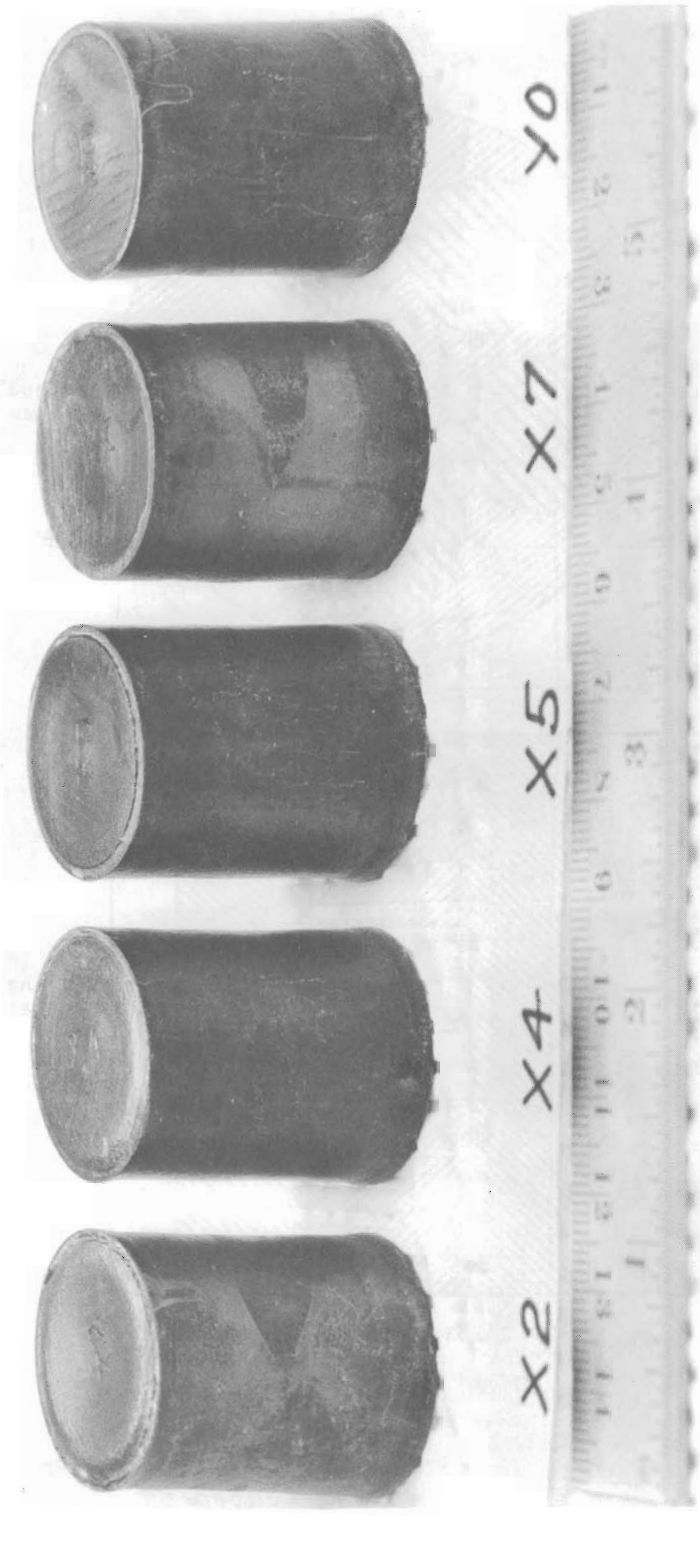

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FIGURE 20. Compatibility $^{147}Pm_2O_3$ Capsules Fabricated by HERF

TABLE 1. Dimensional Data of HERF Processed Capsules

Length, in.	Diameter, in.	
Average	Maximum	Minimum
1.218	1.140	1.100
1.195	1.142	1.100
1.193	1.133	1.090
1.162	1.130	1.110
1.212	1.135	1.110

In applications where compatibility capsules require densification and intimate cladding contact, the standard impact machine tooling is a "soft die" arrangement (Figure 21). This arrangement has been used to densify small single sources. Figure 22 indicates a similar arrangement for multi-source impactions. The fully clad and contained isotope is encased in a container filled with some expendable material such as Al_2O_3. The entire container is densified and the isotope capsule is recovered by breaking away the container and the expendable ceramic material. A subsequent grit-blasting of the capsule completes the process. A highly deformed extremely dense product can be expected. This process has proven adequate in applications where densification is important and dimensional control is of little concern.

SIMULATED COMPATIBILITY CAPSULES

Several modifications of a "hard die" arrangement have been used to produce dense simulated radioisotope heat sources with varying degrees of dimensional control.

Several cladding materials, including stainless steel, super alloys, and refractory metals have undergone HERF using the driver punch concept (Figure 23). This concept consists of the addition of a driver or punch to a massive mold. The

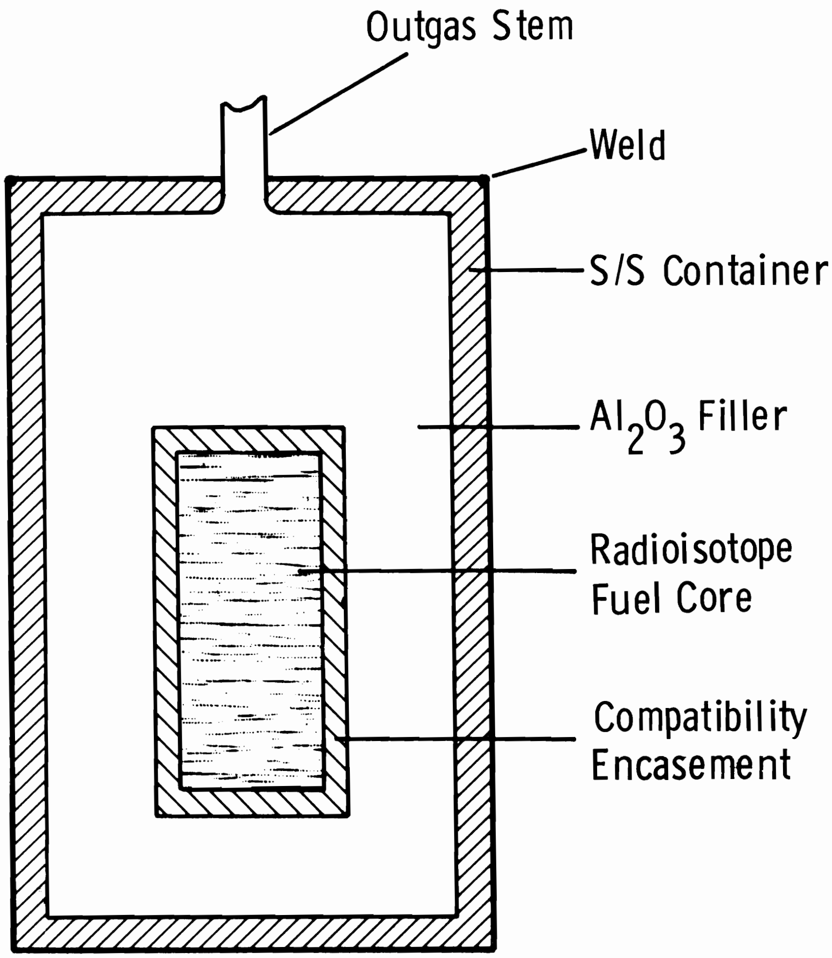

FIGURE 21. Single-Capsule (Soft Die) Impaction Assembly

desired effect is to provide an initial impact along the vertical axis of the capsule and to achieve densification by shortening the capsule. A series of compatibility capsules, using Sm_2O_3 as a stand-in core for $^{147}Pm_2O_3$, was impacted in applying this concept. These impactions were directed towards densifying the core to provide intimate metal-ceramic interfaces. Concurrently, technology was advanced to establish tooling techniques to maintain uniform and predictable dimensions.

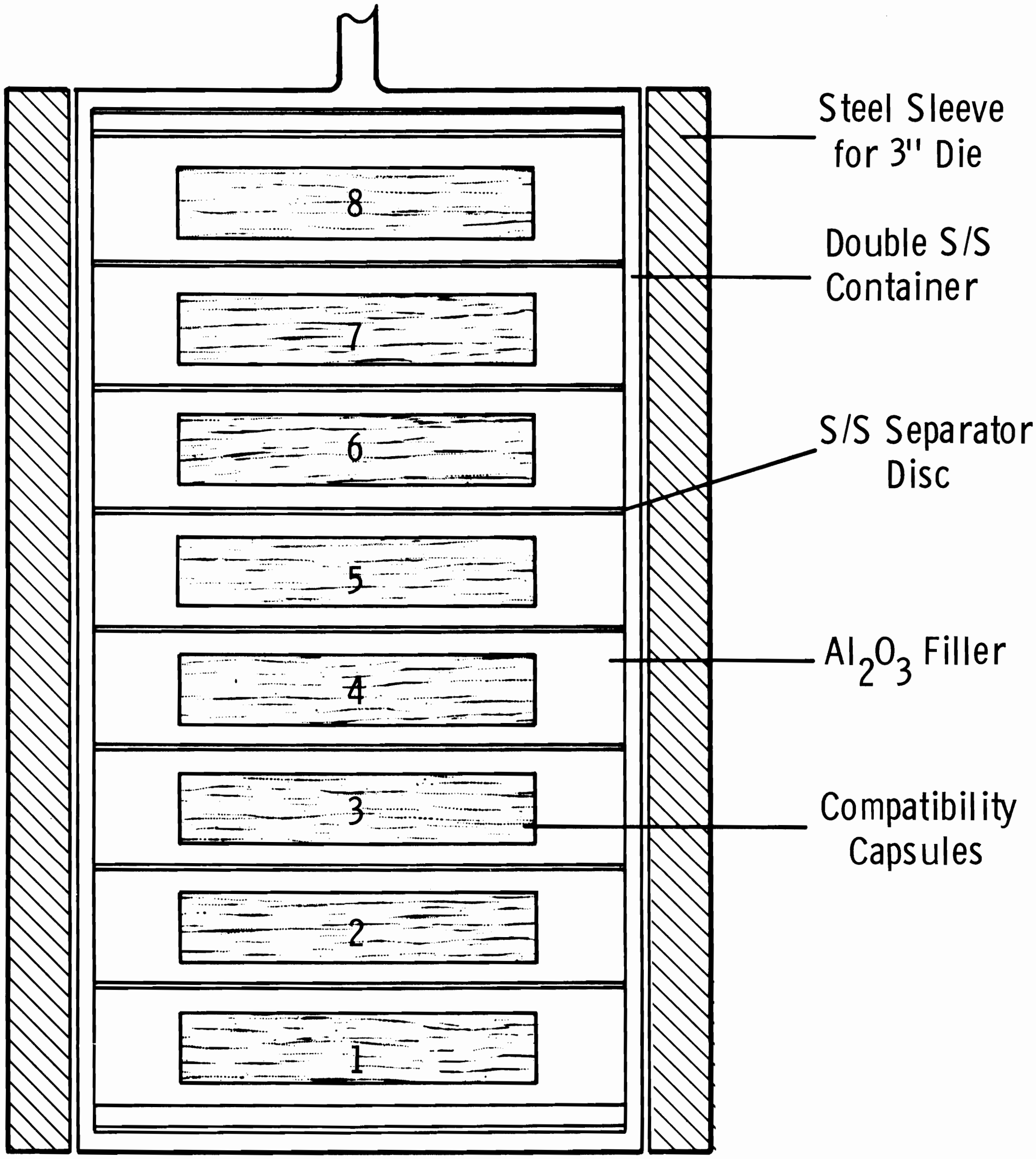

FIGURE 22. Multi-Capsule Impaction Assembly

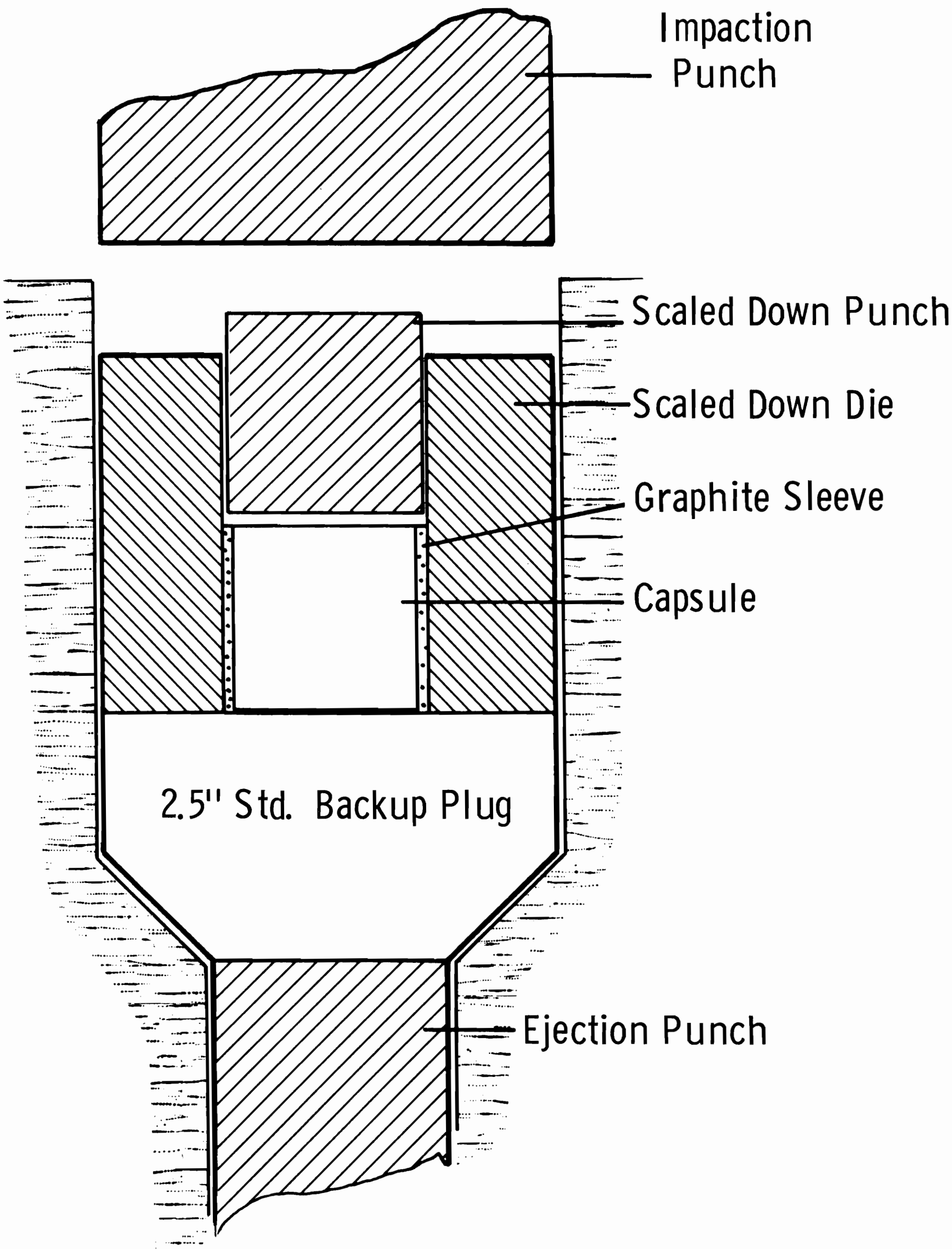

FIGURE 23. Impaction Assembly Using Driver Punch Concept

Dimensional data on this series of compatibility capsules are outlined in Table 2. The billets were heated for 30 min at 1150 °C and impacted in a modified 2.5 in. impaction die (Figure 23). The capsules were placed in a 1/16 in. wall cylinder of graphite, and this assembly was placed in a mild steel mold which, in effect, scaled down the 2.5 in. die to the capsule plus graphite diameter. A 1 in. diam driver punch completed the scale-down of the 2.5 in. impaction die, and the attainable pressures recorded as a function of area were increased accordingly. After impaction and ejection of the 2.5 in. billet, the graphite liner allowed the capsule to be ejected from the massive mold. The graphite sleeve was replaced and steel mold reused for subsequent impactions.

Many factors affect the final dimensions (e.g., initial density, clad material, and starting dimensional uniformity). As shown in Table 2 (Capsules A through H), these major factors were not controlled in the first series of impactions. Repeated reuse of the mild steel molds resulted in irregular dimensional control. The same technique minimizing the reuse of impaction molds produced stainless steel capsules with diameter variations of less than 0.010 in. These are shown as J, K, and L capsules (Table 2). The larger diameters of the J and K series are the result of a second use of the mild steel molds. It was apparent that further improvements could be made by controlling more closely the initial capsule dimensions and loaded density. For the optimum in dimensional control, the impaction molds and punches should be made of heat treated die steel formed to minimum allowable tolerances.

HERF OF HIGH TEMPERATURE $^{147}Pm_2O_3$ HEAT SOURCES

The technique described in the previous section was modified and subsequently used to encapsulate $^{147}Pm_2O_3$.[(10)] The required safety considerations in the HERF process, working

TABLE 2. Compatibility Capsule Data Sheet

Capsule No.	Can Material	Capsule Dimensions, in. (OD × Length)	Impaction Pressure, 1000 psi	Final Dimensions, in. Diameter Max.	Diameter Min.	Length
A-9	Refractory Metal	0.992 × 1.500	115	1.100	1.069	1.178
B-0	Refractory Metal	0.992 × 1.509	110	1.162	1.105	1.127
B-1	Refractory Metal	0.990 × 1.503	100	1.108	1.069	1.195
B-2	Refractory Metal	0.992 × 1.510	100	1.127	1.113	1.132
B-3	Refractory Metal	0.992 × 1.506	97	1.130	1.070	1.195
B-4	Refractory Metal	0.992 × 1.501	105	1.555	1.136	1.092
C-6	Stainless Steel	1.004 × 1.500	175	1.129	1.122	1.155
C-7	Stainless Steel	1.004 × 1.499	140	1.113	1.112	1.191
C-8	Stainless Steel	1.004 × 1.502	90	1.058	1.040	1.314
C-9	Stainless Steel	1.004 × 1.501	105	1.096	1.038	1.285
D-9	Refractory Metal	1.012 × 1.526	95	1.100	1.064	1.265
E-0	Refractory Metal	1.012 × 1.526	110	1.130	1.060	1.218
E-1	Refractory Metal	1.006 × 1.535	90	1.126	1.100	1.185
E-2	Refractory Metal	1.005 × 1.528	95	1.100	1.072	1.245
E-3	Refractory Metal	1.012 × 1.520	100	1.154	1.150	1.100
E-4	Refractory Metal	1.006 × 1.530	105	1.151	1.147	1.133
F-4	Super Alloy	1.012 × 1.502	105	1.132	1.056	1.187
F-5	Super Alloy	1.012 × 1.500	115	1.137	1.061	1.188
F-6	Super Alloy	1.0125 × 1.503	110	1.133	1.052	1.178
F-7	Super Alloy	1.0125 × 1.501	120	1.138	1.058	1.190
F-8	Super Alloy	1.012 × 1.503	105	1.134	1.065	1.202
F-9	Super Alloy	1.012 × 1.503	100	1.114	1.067	1.207
H-0	Super Alloy	1.005 × 1.501	130	1.118	1.050	1.218
H-1	Super Alloy	1.005 × 1.501	100	1.20	1.054	1.225
H-2	Super Alloy	1.006 × 1.506	100	1.130	1.057	1.189
H-3	Super Alloy	1.004 × 1.500	105	1.120	1.060	1.177
H-4	Super Alloy	1.005 × 1.497	110	1.135	1.057	1.172
J-6	Stainless Steel	1.005 × 1.257	115	1.100	1.090	1.029
J-7	Stainless Steel	1.005 × 1.254	107	1.098	1.070	1.047
J-8	Stainless Steel	1.005 × 1.254	110	1.094	1.091	1.040
J-9	Stainless Steel	1.005 × 1.256	112	1.108	1.100	1.042
K-0	Stainless Steel	1.005 × 1.256	110	1.094	1.087	1.055
K-1	Stainless Steel	1.005 × 1.257	106	1.110	1.100	1.037
L-1	Stainless Steel	1.006 × 1.253	110	1.078	1.073	1.083
L-2	Stainless Steel	1.006 × 1.255	109	1.093	1.085	1.082
L-3	Stainless Steel	1.006 × 1.254	115	1.087	1.081	1.098
L-4	Stainless Steel	1.006 × 1.255	112	1.083	1.077	1.112
L-5	Stainless Steel	1.006 × 1.256	100	1.080	1.074	1.096
L-6	Stainless Steel	1.006 × 1.263	95	1.087	1.083	1.084

with the highly radioactive isotope, does not always allow the use of optimum impaction parameters for maintaining dimensional control. For high temperature (1600 °C) impactions, a massive molybdenum impaction mold is used to completely encompass the capsule. The mold is homogeneous, and elimination of any unsymmetrical deformation can be expected. The use of a massive molybdenum mold has simplified investigation of impaction parameters and has indicated the direction for corrective tooling needed to improve final dimension uniformity. The first impacted capsules were highly symmetrical, as was expected. Hourglass-shaped deformation of the capsule side walls took place along with concave dishing of the top and bottom. The massive molybdenum mold provided good heat retention in the transfer of the assembly from the induction furnace to the HERF machine, and was also beneficial in controlling the cooling rate following impaction. The distortion of the capsule, including hourglass and concave dishing effects, was caused by the attempt to densify a porous fuel core in a fully-dense mold. The impaction mold was modified (Figure 24) with the addition of a graphite sleeve around the capsule and compensating dished punches at top and bottom to improve final geometry. Use of this optimized impaction mold concept produced heat source capsules approximately 3/4 in. diam × 3/4 in., with dimensional variations of less than 0.005 in. Slight loss in obtainable maximum core density was observed on detailed examination of the nearly perfect right cylindrical capsules. Although much experience was gained in capsule dimensional control of high temperature HERF, this particular program was directed toward maximum density development and did not allow additional detailed dimensional investigation.

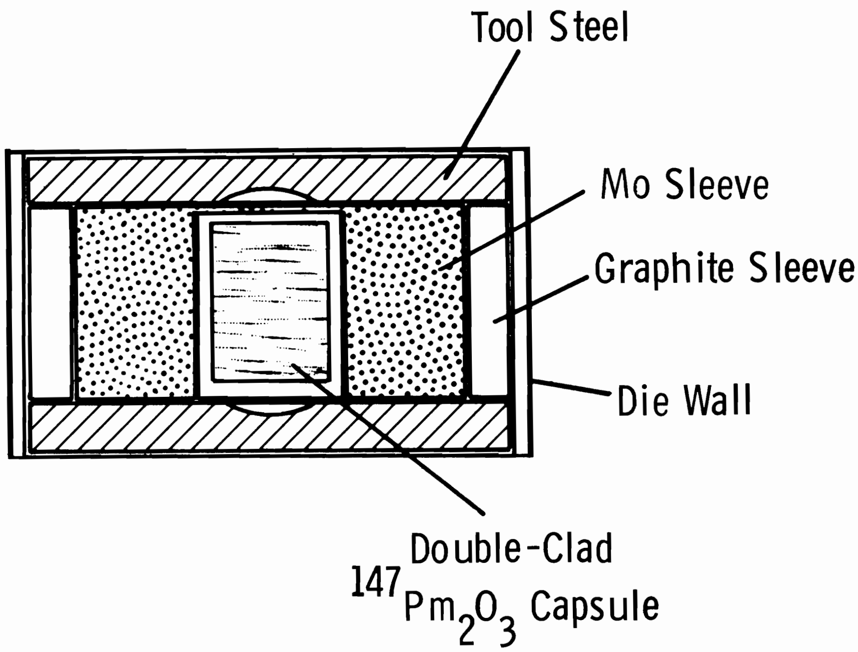

FIGURE 24. High Temperature Impaction Mold Assembly

CONCLUSIONS

In many of the potential terrestrial and aerospace applications of radioisotope heat sources, the apparent advantages of the High Energy Rate Forming process are:

- Maximum fuel power density.
- Compact size, low volume-to-weight ratio.
- Improved structural strength and impact resistance.
- Maximum thermal conductivity.

Additional development work could be beneficial in the areas of specific shapes and dimensional control of heat sources. The apparent improvement of impact strength of capsules fabricated by HERF should receive additional evaluation. An attractive extension of present fabrication technology would be a program to evaluate HERF capsules by physical impact testing. Evaluation of the effects of variations in cladding thickness and core density on impact resistance of capsules fabricated by the HERF process would be of particular interest.

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