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FABRICATION OF INTENSE NEUTRON SOURCES FOR MEDICAL APPLICATIONS

by

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ABSTRACT

Simulated sources containing ^{252}Cf equivalents of 0.1 to 1.0 milligrams were prepared. Samarium was used as the simulant in a modified chemical plating technique similar to that used to prepare palladium-californium oxide cermet for industrial applications.[1] The length of the platinum-10% iridium doubly encapsulated source with its protective sheath is 0.545 in. (14.1 mm). Outside diameter of the source, including its sheath, is 0.109 in. (2.8 mm). Existing "Brachytrons" can accommodate this source form. This capsule system will withstand internal gas pressures from helium due to alpha decay and fission gases from a 1 milligram ^{252}Cf source after ten years if the source is subjected to a maximum temperature of 800°C, the theoretical temperature of an accidental fire. Under these conditions the safety factor is 3. The capsule system is being tested with tracer amounts of ^{252}Cf to ensure that it will withstand adverse service conditions as well as tests specified for Special Form Materials.

The information contained in this article was developed during the course of work under Contract No. AT(07-2)-1 with the U. S. Energy Research and Development Administration.

INTRODUCTION

The first ^{252}Cf sources for radiotherapy research were prepared at the Savannah River Laboratory about ten years ago.[2,3] Initially, these sources resembled the classical radium needles familiar in clinical radiotherapy. Eventually, afterloading cells and applicator tubes were supplied to medical evaluators, and all medical sources were improved by the use of californium-palladium cermet wire sheathed in precious metal alloy as source material.[4,5] The most recent medical source form produced in quantity for therapy research was described in a previous presentation at this meeting.[6] The purpose of my presentation is to describe our progress in the preparation of physically small, intense ^{252}Cf sources for remote afterloading. Remote afterloading is used by many hospitals in the U.S. and abroad to avoid radiation exposure to personnel caring for patients.[7] At least five designs of remote afterloaders are available — the "Curietron" in France, the "Cervitron-II" in Switzerland, the "Cathetron" in England, the "Hicesitron" in U.S., and the "Brachytron" in Canada. The afterloading sources described in this paper were designed specificially for the "Brachytron," manufactured by AECL, Ottawa.

SLIDE 1 (REMOTE AFTERLOADER)

The first remote afterloader in the United States to use cycling sources was installed for clinical use at Memorial Hospital, New York in 1967. Traditionally, the remote afterloader has physically small, intense ^{60}Co sources attached to the ends of long cables which pass through the wall of the shielded cell or treatment room. With the patient in the treatment room and the shielded-room door closed, the sources are advanced to the treatment position in previously installed applicators by means of the cables and a mechanism in the control system outside the treatment-room wall. Patient control is maintained by means of an intercom and closed-circuit television system. Source position control is achieved by a system of count rate meters, lights, and buttons and visually through a shielding window. Source-cycling control is maintained visually and manually or automatically through a system of pre-set stops and friction rollers, which is controlled by a scaled oscillating device. Our goal is to adapt ^{252}Cf sources containing between 0.1 and 1 milligram ^{252}Cf to use in remote afterloaders in the United States and Japan.

SLIDE 2 (^{60}Co SOURCES AND CATHETERS)

Hardware for the "Brachytron" was designed for use with ^{60}Co sources. Straight or curved catheters are available. The "stiff length" of the source is dictated by the minimum curvature radius through which the source must travel during cycling. This slide shows that catheters exist which will accept sources whose "stiff length" is less than 0.600 in. (15.24 mm). Catheters exist which will accept ^{60}Co sources containing up to 10 ^{60}Co pellets. The "stiff length" of such a source is 0.960 in. (24.38 mm). Our goal has been to prepare a 1 milligram ^{252}Cf source with the shortest possible "stiff length" and the greatest possible integrity.

SLIDE 3 (^{252}Cf "BRACHYTRON" SOURCE)

We have prepared simulated ^{252}Cf "Brachytron" sources containing up to 1 milligram ^{252}Cf equivalents using samarium as the ^{252}Cf stand-in. This design consists of a double encapsulation in platinum-10% iridium alloy which is protected by a sheath crimped to the 1/16-inch diameter of a 304L stainless steel, 7 × 7-strand cable. This connection withstood the required 100 pounds of tensile force. "Stiff length" of this assembly is ∿14 mm; diameter is 2.8 mm. The assembled source will pass through existing catheters. Inner and outer capsules have been fabricated from Pt-10% Ir tubing and by machining swaged Pt-10% Ir rod. The encapsulated source was welded with a Linde PWM-4 Plasma Needle Arc Welder fitted with a PT-10 Plasma Needle Arc Torch. Preparation of the ^{252}Cf source pellet and integrity tests of the encapsulation will be described in subsequent slides.

SLIDE 4 (^{252}Cf JAPANESE REMOTE AFTERLOADING SOURCE)

Neutron sources for ^{252}Cf research in Japan can be slightly larger in physical size and must contain approximately 100 to 200 micrograms ^{252}Cf. The source carrier that is provided to us will accept a doubly encapsulated source 11 mm long by 4.7 mm in diameter. The double encapsulation is made of platinum-10% iridium alloy and is plasma arc-welded. We have prepared ten of these sources for physical testing, which is in progress.

SLIDE 5 ($^{252}Cf_2O_3$-Pd POWDER BY CHEMICAL PLATING)

A chemical plating technique has been used for about 3 years to prepare palladium-californium oxide cermet powder from which industrial ^{252}Cf neutron sources are made. Since "Brachytron" sources are much smaller and contain more ^{252}Cf than other sources for medical or industrial applications, we modified our chemical plating technique used to prepare industrial cermet pellets and wire to produce smaller batches of powder containing more ^{252}Cf in greater yield.

A synthetic feed solution containing samarium nitrate plus typical oxalate-forming impurities found in ^{252}Cf feed stocks is added to the small glass reaction vessel. The volume of solution is chosen so as to provide up to 1 milligram ^{252}Cf equivalents. Approximately 40 milligrams oxalic acid in solution is added. The mixture is sparged with air through a hypodermic needle immersed in the solution. After californium oxalate has precipitated, 1 ml of 85% hydrazine hydrate is added, and the mixture is again sparged with air. Air sparging promotes mixing and the formation of finely divided oxalate particles when the hydrazine hydrate is added. This dispersion promotes uniform coating, or plating, of the oxalate particles during subsequent reduction of palladium ion to metal and precipitation of the metal around the oxalate particles. Finally, 45 milligrams of palladium are introduced as a solution of palladium tetramine dinitrate while the oxalate particles are kept suspended by air sparging. When the reduction of palladium ion to palladium metal by the hydrazine hydrate is complete, the oxalate particles have been coated with palladium metal. The metal-coated precipitate is permitted to settle under an argon atmosphere. The supernate is aspirated to a recovery vessel.

The reaction vessel is lowered into the furnace and valved to the argon gas line, and the precipitate is dried under argon at 100°C. The dried powder is calcined under a 96% helium — 4% hydrogen gas mixture for approximately 30 minutes. The calcined powder is then cooled from approximately 200°C to room temperature in an argon atmosphere.

SLIDE 6 ($^{252}Cf_2O_3$-Pd CERMET PELLET FABRICATION)

The glass reaction vessel containing the powder is designed to be mated and sealed to a polished metal funnel so the entire charge of metal powder can be transferred through the funnel to a compaction die. A vibrator is used if necessary to aid in transferring the small powder batch.

The compaction die has an inside diameter of only 0.050 in. (1.27 mm), so the ram is 0.0495 in. (1.21 mm) x 2 in. (50.8 mm) long and must be supported throughout its travel in the die. Ram support bushings are assembled on top of the die after the powder charge has been transferred. As the powder is pressed into a pellet, ram support bushings are removed sequentially until the pellet is formed. This "green" pellet is pressed at approximately 12,500 psi. The pellet is next transferred to a combustion tube and sintered. Sintering takes place under 96% helium — 4% hydrogen up to 1000°C and under argon at 1050°C for two hours.

The sintered pellet is coined in a special die to a 1.17-mm diameter. Coining is performed at approximately 15,600 psi. Average length of 10 coined pellets was 0.070 in. (1.8 mm).

SLIDE 7 (CAPSULE INTERNAL GAS PRESSURES)

Maximum permissible internal gas pressures at 25°C and 800°C in platinum-10% iridium capsules of the "Brachytron" design were calculated using the latest published data available for tensile strengths at elevated temperatures. Internal gas pressures from helium due to alpha decay and from the fission gases was calculated using currently accepted nuclear data for half-life and spontaneous fission decay of ^{252}Cf. These data are summarized in the slide.

At the theoretical fire temperature of 800°C, the primary capsule can accommodate a 1-milligram source with a safety factor of approximately 1.5. The secondary capsule, under the same conditions, will provide an additional safety factor of approximately 3.

SLIDE 8 (CAPSULE SAFETY TESTS)

Using tracer amounts of ^{252}Cf, the capsule system will be subjected to tests to simulate expected adverse service conditions. Four tests (impact, percussion, heating, and immersion) are specified in *ERDA Manual Chapter 0529-05*, "Safety Standards for the Packaging of Radioactive and Fissile Materials, Annex 4, Tests for Special Form Materials." Four other tests (sterilization, pinching, crushing, and abrasion), though not specified in the ERDA Manual, will be conducted to demonstrate the safety of the capsule system under conditions which might be expected during service. These tests will also satisfy requirements of the Nuclear Regulatory Commission's Regulatory Guides 6.1 ("Leak Testing Radioactive Brachytherapy Sources") and 6.2 ("Integrity and Test Specifications for Selected Brachytherapy Sources") both dated February 1974.

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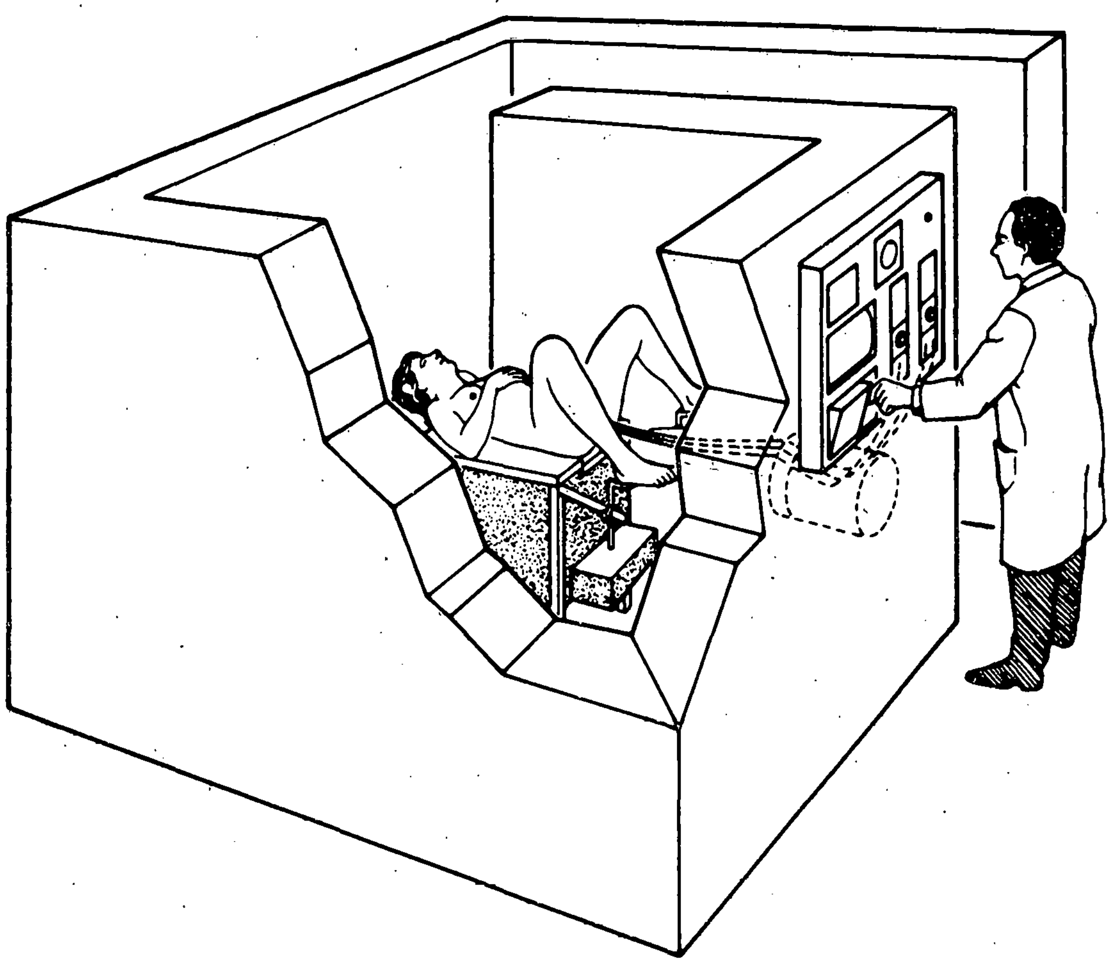
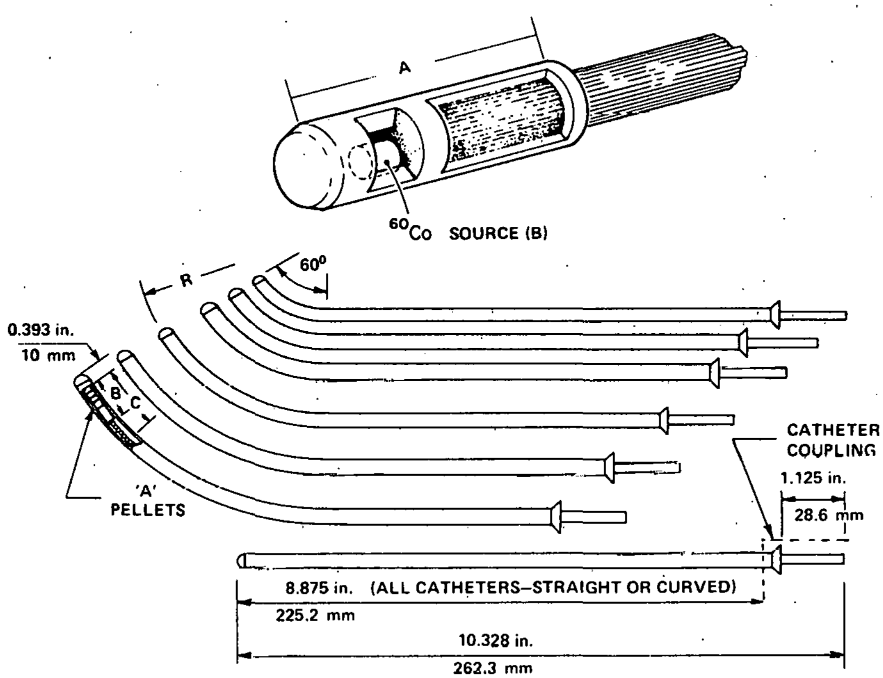
7. Basil S. Hilaris, M.D. "A Manual for Brachytherapy," pp 75-81. Memorial Hospital, New York, Department of Radiation Therapy, Second Edition (1970).

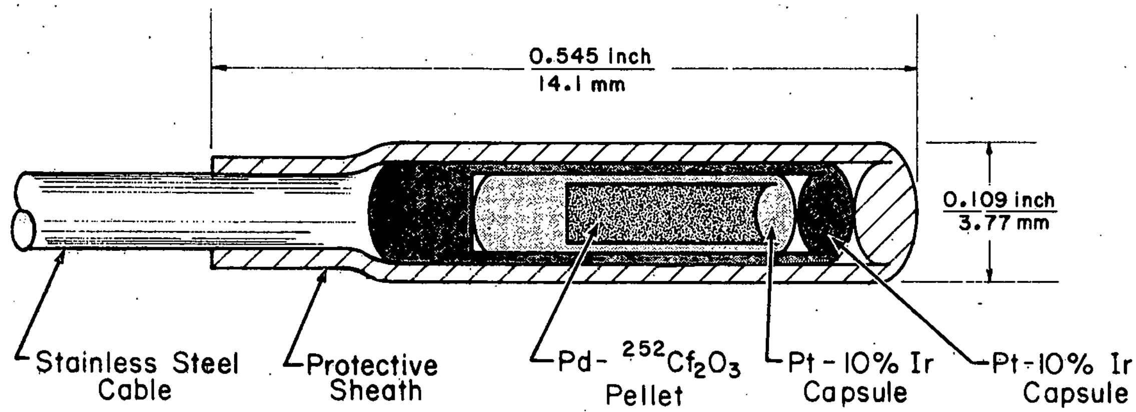


SLIDE 1 Remote Afterloader

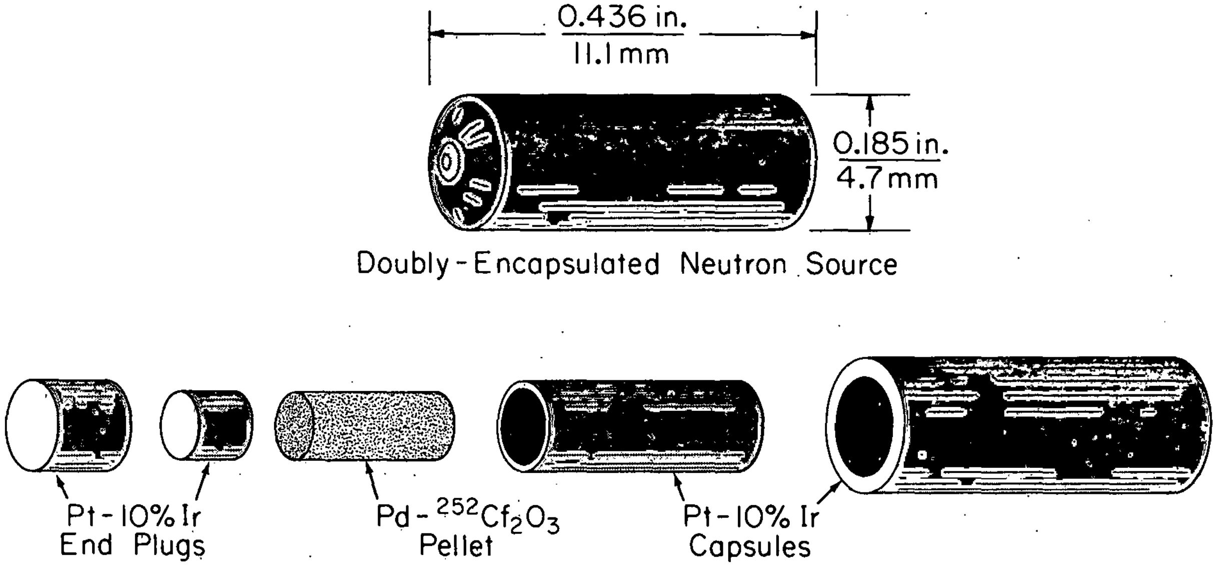
A No. of Pellets	B Source Length		C "Stiff" Length		R = Minimum Curvature Radius	
	inch	mm	inch	mm	inch	mm
Two	0.350	8.89	0.600	15.24	2.00	50.8
Ten	0.710	18.03	0.960	24.38	5.00	127.0



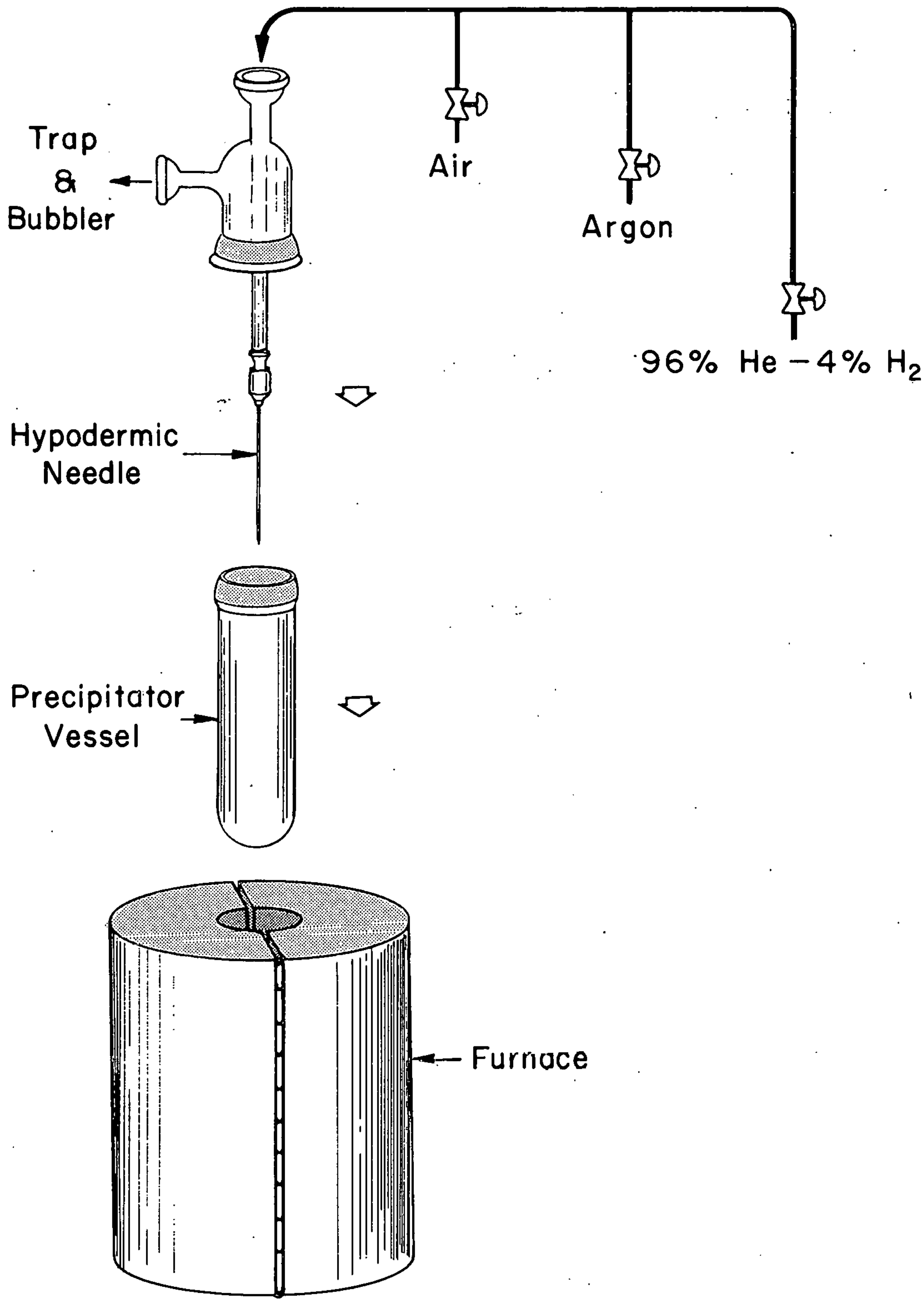
SLIDE 2 ^{60}Co Sources and Catheters



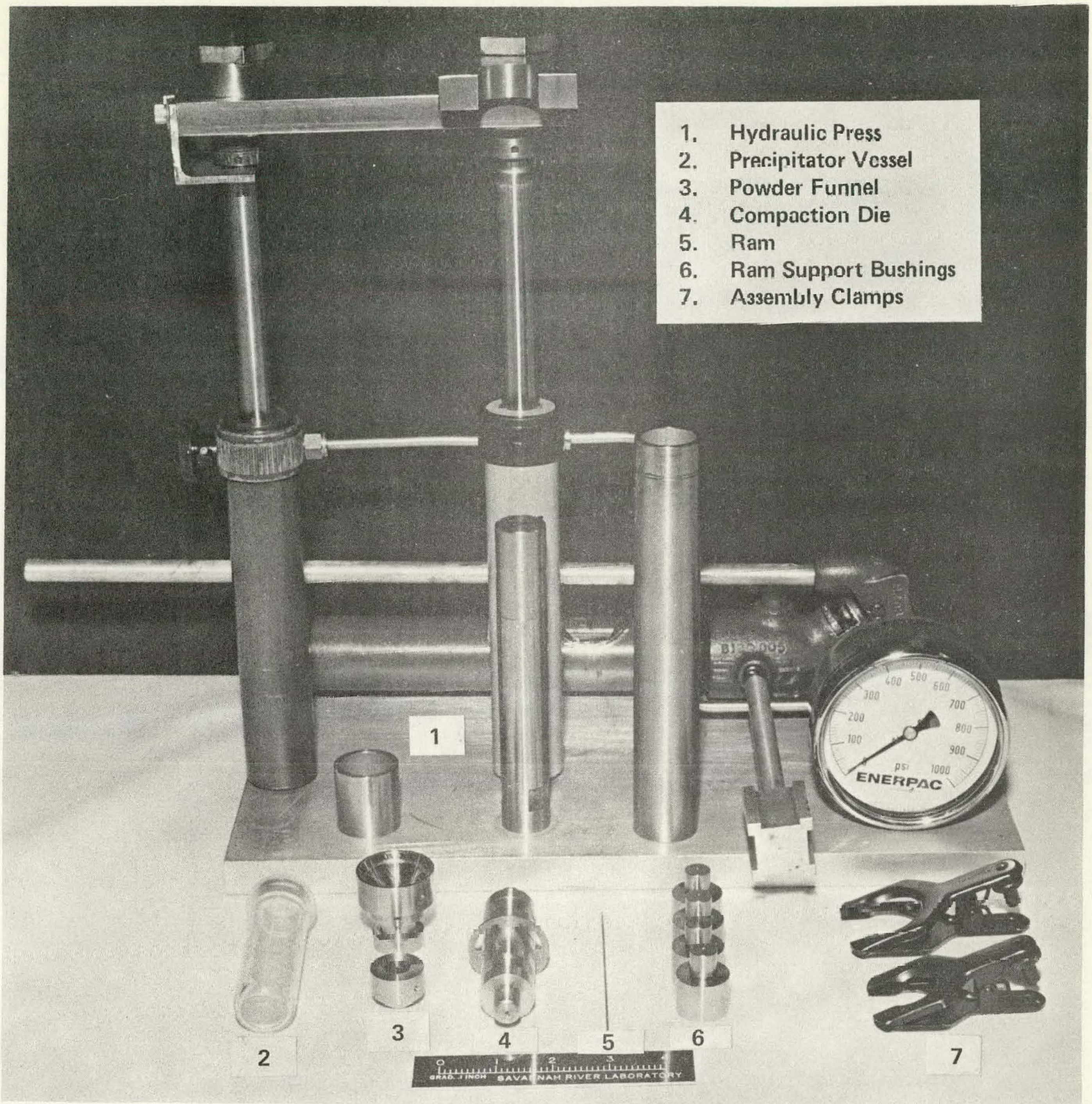
SLIDE 3 ^{252}Cf "Brachytron" Source



SLIDE 4 Japanese ^{252}Cf Afterloading Source



SLIDE 5 $^{252}Cf_2O_3$-Pd Powder by Chemical Plating



SLIDE 6 $^{252}Cf_2O_3$-Pd Cermet Pellet Fabrication

Capsule Description	Pressure (psi) at 25°C: Permissible	After 10 yr Decay: 0.2 (milligrams)	1.0	2.0	Pressure (psi) at 800°C[a]: Permissible	After 10 yr Decay: 0.2 (milligrams)	1.0	2.0
Primary[b]	8,500	155	715	1,430	4,500	605	2,780	5,560
Secondary[b]	10,445	110	500	1,000	5,610	420	1,940	3,875

a. Theoretical temperature during an accidental fire.

b. Platinum - 10% Iridium alloy.

SLIDE 7 Capsule Internal Gas Pressures

Impact - free drop	Sterilization
Percussion	Pinching
Heating	Crushing
Immersion	Abrasion

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a. Specified and described in *AEC Manual Chapter 0529-05*, "Safety Standards for the Packaging of Radioactive and Fissile Materials, Annex 4, Tests for Special Form Materials."

SLIDE 8 Capsule Safety Tests