Bill Remini
353-4265

Department of Energy
Washington, D.C. 20545

OCT 07 1983

Mr. Steven Baggett
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555

Dear Mr. Baggett:

This is in reference to your meeting with Thomas Anderson of my staff on October 6, 1983, regarding Nuclear Regulatory Commission (NRC) registry of cesium-137 capsules produced by the Department of Energy at the Hanford Reservation Waste Encapsulation and Storage Facility (WESF). We request that evaluation of the generic capsule for registry be undertaken by your office.

As discussed at the October 6 meeting, these capsules are of the same generic design, which is designated as the Model A WESF capsule (certification package enclosed). Also, two copies of the detailed information regarding registry of this capsule were provided at the meeting and this information should be used as the basis for the evaluation. Please contact Tom Anderson, 353-5560, if you have any questions or require further information regarding registry of the Model A WESF capsule.

Your expeditious response will be appreciated.

Sincerely,

John James Jicha Jr.

John J. Jicha, Jr., Director
R&D and Byproducts Division
Office of Defense Waste
and Byproducts Management

Enclosure

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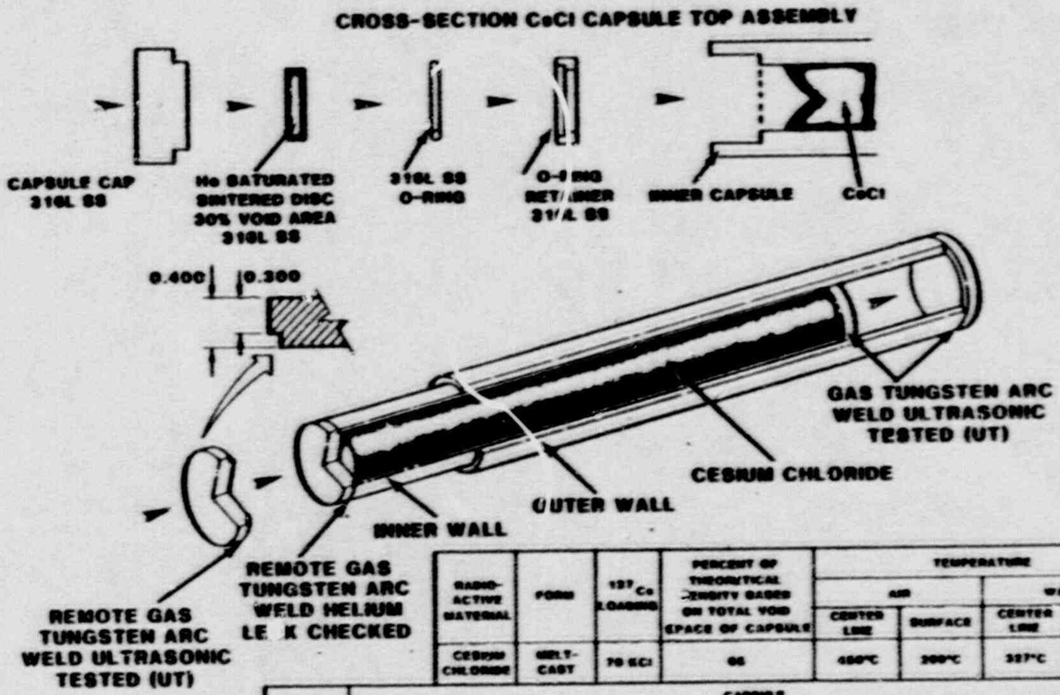

RADIO-ACTIVE MATERIAL	FORM	^{137}Cs LOADING	PERCENT OF THEORETICAL DENSITY BASED ON TOTAL VOID SPACE OF CAPSULE	TEMPERATURE			
				AIR		WATER	
				CENTER LINE	SURFACE	CENTER LINE	SURFACE
CESIUM CHLORIDE	MELT-CAST	70 KCi	66	450°C	200°C	327°C	50°C

RADIO-ACTIVE MATERIAL	CAPSULE									
	INNER					OUTER				
	MATERIAL	WALL THICKNESS	OUTSIDE DIAMETER	TOTAL LENGTH	TOTAL CAP THICKNESS	MATERIAL	WALL THICKNESS	OUTSIDE DIAMETER	TOTAL LENGTH	TOTAL CAP THICKNESS
CESIUM CHLORIDE	316L STAINLESS STEEL (UT)	0.136 (UT)	2.250	19.725	0.400	316L STAINLESS STEEL (UT)	0.109 (UT)	2.625	20.775	0.400

NOTE: ALL DIMENSIONS ARE IN INCHES

Detail of WESF ^{137}Cs Source Capsule.

NRC FORM 618
(6-83)
10 CFR 71

CERTIFICATE OF COMPLIANCE
FOR RADIOACTIVE MATERIALS PACKAGES

U.S. NUCLEAR REGULATORY COMMISSION

1. a. CERTIFICATE NUMBER	b. REVISION NUMBER	c. PACKAGE IDENTIFICATION NUMBER	d. PAGE NUMBER	e. TOTAL NUMBER PAGES
5939	10	USA/5939/B( )F	1	3

2. PREAMBLE

a. This certificate is issued to certify that the packaging and contents described in Item 5 below, meets the applicable safety standards set forth in Title 10, Code of Federal Regulations, Part 71, "Packaging of Radioactive Materials for Transport and Transportation of Radioactive Material Under Certain Conditions."

b. This certificate does not relieve the consignor from compliance with any requirement of the regulations of the U.S. Department of Transportation or other applicable regulatory agencies, including the government of any country through or into which the package will be transported.

3. THIS CERTIFICATE IS ISSUED ON THE BASIS OF A SAFETY ANALYSIS REPORT OF THE PACKAGE DESIGN OR APPLICATION

a. PREPARED BY (*Name and Address*):

General Electric Company
P.O. Box 460
Pleasanton, CA 94566

b. TITLE AND IDENTIFICATION OF REPORT OR APPLICATION:

General Electric Company application dated February 21, 1980, as supplemented.

c. DOCKET NUMBER 71-5939

4. CONDITIONS

This certificate is conditional upon fulfilling the requirements of 10 CFR Part 71, as applicable, and the conditions specified below.

5.

(a) Packaging

(1) Model No.: GE-1500

(2) Description

A steel encased lead shielded shipping cask. The cask is a double-walled steel circular cylinder, 31-inch-diameter by 48 inches high with a central cavity 7-inch diameter by 25 inches high. The diameter is reduced from 31 inches to 17-1/2 inches by cone construction at the top 7-1/2 inches of the cask. Approximately 11 inches of lead surround the central cavity. The cask is equipped with a cavity drain line and lifting device. Closure is accomplished by a gasketed and bolted steel lead-filled plug. A protective jacket consisting of an upright circular cylinder with open bottom and a protruding box section diametrically across the top and vertically down the sides attaches to a square pallet. Dimensions of the protective jacket are 60-7/8 inches high by 49-3/4 inches wide across the box section. The outer cylindrical diameter is 36-1/2 inches and the pallet is 59-1/2 inches square. The maximum weight of the packaging is approximately 15,000 pounds.

~~8309020134~~ 127 3pp.

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5. (a) Packaging (continued)

(3) Drawings

The packaging is constructed in accordance with the following General Electric Company Drawing Nos.: 129D4748, Rev. 3; 129D4749, Rev. 3; and 129D4750, Rev. 3.

Lifting and/or tie-down devices which are a structural part of the package must be in accordance with the above drawings.

(b) Contents

(1) Type and form of material

(i) Byproduct material and special nuclear material meeting the requirements of special form radioactive material and antimony pins encased in stainless steel; or

(ii) Byproduct material in the form of $^{90}\mathrm{SrF}_2$ or $^{137}\mathrm{CsCl}$.

(2) Maximum quantity of material per package

Not to exceed a decay heat generation of 3,120 watts and

(i) Item 5(b)(1)(i) above:

Plutonium in excess of twenty (20) curies per package must be in the form of metal, metal alloy or reactor fuel elements, and 500 grams U-235 equivalent mass. (U-235 equivalent mass equals U-235 mass plus 1.66 times Pu mass.)

(ii) Item 5(b)(1)(ii) above:

458,000 ci.

(c) Fissile Class III

Maximum number of packages per shipment 22

6. For the contents described in Item 5(b)(1)(ii) above:

$^{90}\mathrm{SrF}_2$ must be encapsulated in accordance with Vitro Drawing Nos. H-2-66759, Rev. 0; and H-2-66758, Rev. 0; or.

$^{137}\mathrm{CsCl}$ must be encapsulated in accordance with Vitro Drawing Nos. H-2-66760, Rev. 0; and H-2-66761, Rev. 0.

The $^{90}\mathrm{SrF}_2$ and $^{137}\mathrm{CsCl}$ capsules after fabrication must be leak tested using a method having sufficient sensitivity to detect a leak rate (air at standard

Page 3 - Certificate No. 5939 - Revision No. 10 - Docket No. 71-5939

temperature and pressure leaking to 10^{-2} atm) of 10^{-8} atm cc/sec. Any capsule with a detectable leak may not be delivered to a carrier for transport.

7. The package authorized by this certificate is hereby approved for use under the general license provisions of 10 CFR §71.12.

8. Expiration date: December 31, 1987.

REFERENCES

General Electric Company application dated February 21, 1980.

Supplement dated: September 8, 1982.

Oak Ridge National Laboratory letter dated April 3, 1980.

Supplement dated: May 7, 1980.

FOR THE U.S. NUCLEAR REGULATORY COMMISSION

Charles E. MacDonald

Charles E. MacDonald, Chief
Transportation Certification Branch
Division of Fuel Cycle and
Material Safety, NMSS

Date: SEP 06 1983

Tom Anderson, DOE
353-5560 X

REGISTRY OF RADIOACTIVE SEALED SOURCES AND DEVICES
SAFETY EVALUATION OF SEALED SOURCE

NO.: NR______-S-______ - _____ **DATE:** June 1, 1982 **Page** 1 of 7

SEALED SOURCE TYPE: WESF ^{137}Cs Capsule.

MODEL:

Serial Numbers C-1 through C1500 etched onto top end cap of outer capsule.

MANUFACTURER/DISTRIBUTOR:

Manufacturer:	Waste Encapsulation and Storage Facility (WESF), Rockwell Hanford, Richland, WA.
Distributor:	DOE, Oak Ridge Operations, Oak Ridge, TN.
Applicant:	Department of Energy, Albuquerque Operations, Albuquerque, NM.

ISOTOPE:	**MAXIMUM ACTIVITY:**
Cesium-137 (^{137}Cs)	80 kCi

LEAK TEST FREQUENCY:

Six (6) months

PRINCIPAL USE:

Gamma Ray Source for Gamma Irradiator, Category III

CUSTOM REVIEW: _____Yes __X__No

DESCRIPTION:

Approximately 6 kg of CsCl, containing a maximum of 80 kCi ^{137}Cs is melt-cast at 740° C into a 316L stainless steel (SS) capsule having one end cap welded in place by a tungsten/inert gas arc welding process (TIG). The top end cap is welded in place by TIG and, after a helium leak check to certify the weld, this inner capsule is cleaned and placed into an outer capsule made of 316L SS which has one end cap already welded in place. The top end cap is welded on by TIG and the weld is verified by ultrasonic inspection. The dimensions in inches are: inner capsule, 2.250 o.d. and 19.725 long; outer capsule, 2.625 o.d. and 20.775 long. Both capsules have wall thicknesses of 0.136 ± 0.012 inches. The ANSI classification is Group B-1 (medium toxicity), Category III (sealed source, wet storage).

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REGISTRY OF RADIOACTIVE SEALED SOURCES AND DEVICES
SAFETY EVALUATION OF SEALED SOURCE

NO.: NR______-S-______ - ______ DATE: June 1, 1982

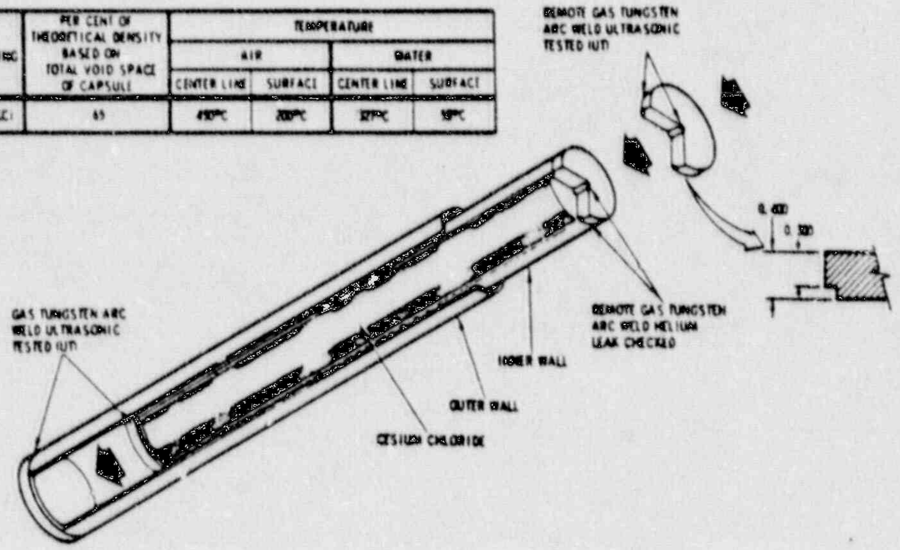
SEALED SOURCE TYPE: WESF ^{137}Cs Capsule.

LABELING:

Each inner capsule has an identification number etched on it. A unique serial number is etched on the top end cap of every outer capsule. This latter is used for identification of each WESF capsule. Individual capsules need not be marked, "RADIOACTIVE", because the capsules will be securely mounted in a source plaque contained within an irradiation facility. The facility will be labeled as a radiation zone and marked according to NRC requirements.

DIAGRAM:

	FORM	LOADING	PER CENT OF THEORETICAL DENSITY BASED ON TOTAL VOID SPACE OF CAPSULE	TEMPERATURE			
				AIR		WATER	
				CENTER LINE	SURFACE	CENTER LINE	SURFACE
CESIUM CHLORIDE	MELT-CAST	70 KCi	65	450°C	200°C	320°C	90°C



	CAPSULE									
	INNER					OUTER				
	MATERIAL	WALL THICKNESS	OUTSIDE DIAMETER	TOTAL LENGTH	TOTAL CAP THICKNESS	MATERIAL	WALL THICKNESS	OUTSIDE DIAMETER	TOTAL LENGTH	TOTAL CAP THICKNESS
CESIUM CHLORIDE	316L STAINLESS STEEL (UT)	0.095 (UT)	2.250	19.725	0.400	316L STAINLESS STEEL (UT)	0.136 (UT)	2.625	20.775	

NOTE: ALL DIMENSIONS ARE IN INCHES NOVEMBER 1977

REGISTRY OF RADIOACTIVE SEALED SOURCES AND DEVICES
SAFETY EVALUATION OF SEALED SOURCE

NO.: NR______-S-______ - ______ DATE: June 1, 1982

SEALED SOURCE TYPE: WESF ^{137}Cs Capsule.

CONDITIONS OF NORMAL USE:

The source capsules are to be used as gamma-ray sources in facilities designed to irradiate sewage sludge, food commodities, medical products or other products. The objective of sewage sludge irradiation is to reduce harmful pathogen populations to levels allowing unrestricted reuse of the sludge as a soil conditioner. Irradiation of food commodities would destroy pests of quarantine significance and could also eliminate parasitic organisms such as Trichinella spiralis in pork. Irradiation of medical products would be used to sterilize these items prior to use.

Any potential non-DOE users of cesium-137 sources in irradiation facilities would design, build, operate and maintain their facilities and would be responsible for obtaining a license for each specific facility from NRC or the appropriate state authority in agreement states.

Within each facility, the cesium source capsules would be arrayed and held securely in place in a source plaque. For instance, in the pilot sludge irradiator at Sandia National Laboratories (SNLA) Sandia Irradiator for Dried Sewage Solids (SIDSS), 15 of the WESF capsules are arranged horizontally in an open source plaque. Such a design permits heat dissipation by air convection assisted by forced air flow. The only physical contact made by the sources is with the source plaque. No contact takes place with the material being irradiated. Exterior surface temperatures of the source capsule have been estimated to be 200-220°C and centerline temperatures will be ca. 450°C. However, recent experiments conducted by SNLA on the SIDSS source plaque indicate much lower outer capsule surface temperatures. Contact thermocouple temperatures were measured across four of the WESF capsules in the SIDSS source plaque in air and were found to be from 30°C to 80°C depending upon location along the capsule length (30°C was measured near the capsule end where a cesium chloride shrinkage void is created during solidification of the material). A similar outer capsule surface temperature of 104°C was measured by Oak Ridge National Laboratory (ORNL) in a hot cell on the WESF capsule removed from SIDSS for destructive analysis.

In some irradiator designs using numerous capsules, ambient temperatures could be higher than those measured in SIDSS. Design of these facilities will include appropriate ventilation schemes to cool the capsules and will provide backup water storage options in case of air circulation system failure. The goal of these cooling mechanisms is to keep centerline cesium chloride temperatures below 450°C thus preventing any solid-solid phase transformation of the cesium chloride.

CONDITIONS OF NORMAL USE: (cont)

The expected useful lifetime of a capsule as a gamma source is about one half-life of cesium-137, i.e. 30 years.

Capsule integrity has been verified under the applicable abnormal physical conditions specified by ANSI N542 for Category III irradiators. This includes being subjected to temperatures of 800°C (1470°F) for up to 90 minutes, conditions similar to those experienced in a fire environment. Category III is the appropriate category for irradiators using cesium WESF capsules. This category of irradiator has a self-contained, wet-storage source. The cesium WESF capsules are self-contained in a double steel encapsulation and would be isolated in a controlled-access area within the irradiators. Some irradiator designs could utilize a water-pool for storage of the sources when not in use (as is common with current cobalt-60 medical product irradiators). Other designs such as SIDSS, would allow retraction of the source plaques into a dry storage area behind a shield. Even in these designs, though, initial loading and periodic recharge of the sources would take place under water.

Therefore, in most irradiator designs, it is probable that the sources will be intermittently exposed to a water environment. Category III, as a more stringent condition than dry source storage, is thus appropriate for these irradiator designs. DOE is proceeding with design of a TransPortable Cesium Irradiator (TPCI) which involves only dry storage of the cesium WESF capsules. This irradiator would be classified as a Category I - self-contained, dry source storage irradiator.

Diversion of a capsule from its intended use would not be of concern from a weapons standpoint because cesium-137 is not a fissionable material. From a radiological health viewpoint, theft of a capsule is a remote threat because of the very high radiation level from an unshielded capsule. To further ensure that the probability of theft was small, facilities would have to be designed to make access to the source area by unauthorized personnel very difficult.

REGISTRY OF RADIOACTIVE SEALED SOURCES AND DEVICES
SAFETY EVALUATION OF SEALED SOURCE

NO.: NR______-S-______ - ______ **DATE:** June 1, 1982 **Page** 5 of 7

SEALED SOURCE TYPE: WESF ^{137}Cs Capsule.

PROTOTYPE TESTING:

Studies during the past 10 years have demonstrated that the WESF capsule will maintain its integrity after being subjected to the adverse physical test conditions specified for Category III gamma sources in ANSI N542. Details are provided in the attached SAND document (Exhibit 1). Compatibility of CsCl with 316L stainless steel has been investigated to ensure that corrosion is not a problem. Studies are continuing using a capsule taken from SIDSS. This capsule was fabricated by DOE/Rockwell-Hanford in 1975 and was used in the SIDSS for 2 years from 1979 to 1981. It was removed in 1981, sent to Oak Ridge National Laboratory and destructively analyzed. Chemical analyses of the gaseous environments in the inner and outer capsules, detailed metallographic study of the capsule surfaces and mechanical tests of capsule parts were conducted. Results indicate no notable corrosion occurred which would jeopardize capsule integrity over a 30-year life and that the capsule basically was immune to the presence of the CsCl. Detailed results are given in SAND83-0928. Cesium WESF sources were certified as special form material for shipping by ERDA in 1973.

EXTERNAL RADIATION LEVELS:

This information is provided by the reference, K. M. Harmon, BNWL, letter to R. A. Libby, BNWL, "Source Efficiency Calculations for ^{137}Cs Irradiators," April 29 (1976). This is for an unshielded 70 kCi capsule (Exhibit 2).

Distance from Surface of Source (cm)	R/hr Measured	R/hr Calculated
0	--	7.7×10^5 *
5	--	6.1×10^5 *
20	3.02×10^5	3.0×10^5
30	1.69×10^5	1.6×10^5
50	0.55×10^5	0.68×10^5

*Extrapolated.

SEALED SOURCE TYPE: WESF ^{137}Cs Capsule.

LIMITATIONS AND/OR OTHER CONDITIONS OF USE:

The WESF ^{137}Cs gamma source capsule should not experience environments which:
1) will produce temperatures >800°C (>1470°F),
2) will produce external pressures >47 atm or internal pressures >7000 psi (480 atm),

If any of these environments are experienced, the capsules affected will be removed from service immediately and thoroughly inspected for any compromise of their integrity.

QUALITY ASSURANCE:

The quality control standards for maintaining source design specifications are the operational procedures and Quality Assurance Program implemented by Rockwell-Hanford. These pertain to the three major operations, viz. transfer of cesium solution to the conversion area, conversion of cesium carbonate to cesium chloride with subsequent drying/melting, and finally the casting/encapsulation. Each of these has a QC procedural form which must be checked and signed as the process continues. Procedures are outlined in various Rockwell-Hanford operation specifications as follows:

1. HWS-8835 - Procurement/acceptance criteria for 316L stainless steel tubing
2. H2-66760, H2-66761 - Fabrication criteria, procedures for capsules from 316L tubing
3. SDWM-OCD-003 - Capsule welding procedures, criteria
4. PSD-B-257-00053 (Rev. D-0) - Cesium chloride production process specifications (including purity of cesium chloride feed which requires molar ratio of combined amounts of Na, K, Rb to be less than .15. Weight percent impurities would thus range from <4 - 6% depending upon which atoms were present.
5. PSD-B-257-00054 (Rev. D-0) - Capsule handling, welding, leak checking, decontamination and calorimetry procedures.
6. PSD-B-257-00055 (Rev. D-0) - Capsule pool storage procedures.

REGISTRY OF RADIOACTIVE SEALED SOURCES AND DEVICES
SAFETY EVALUATION OF SEALED SOURCE

NO.: NR______-S-______ - ______ DATE: June 1, 1982

SEALED SOURCE TYPE: WESF ^{137}Cs Capsule.

SAFETY SUMMARY EVALUATION:

The accompanying SAND document (Exhibit 1) provides the details of the safety studies which have been performed on the WESF capsule. The results can be summarized by stating that these tests have shown that the WESF capsule meets or exceeds the requirements for a Category III gamma source as described by ANSI N542. There are numerous applicable references and they are given in Exhibit 1.

MANUFACTURER'S RECOMMENDATIONS:

The manufacturer makes no recommendations for leak testing, unpacking, or handling of these sources. These matters will be addressed in facility license applications. The question of disposal has not been addressed as of this time. However, arrangements for ultimate disposal of WESF Cs capsules will be made with DOE or whatever successor agency has responsibility for the remaining WESF capsules at the time disposal is sought.

Pertinent radiological safety and operating instructions for the sources will be developed by the A&E firm which designs the irradiation facilities in which the sources are to be used. These items will be developed pursuant to NRC guidelines to allow facility licensing.

March 1982 *

GUIDELINES FOR APPLICATIONS FOR
REGISTRATION OF SEALED SOURCES

This guide has been prepared to assist manufacturers/distributors in the preparation of applications for registration of the design for sealed sources containing radioactive material. The objectives are:

- o To identify and explain the elements of an application that are necessary to demonstrate the adequacy of the sealed source design from the standpoint of health and safety.
- o To facilitate the consistent, effective and timely review of applications by the U.S. Nuclear Regulatory Commission (NRC) and Agreement States.
- o To facilitate the preparation by reviewing agencies, NRC or Agreement States, of registration sheets in a prescribed format.

Applications for registration of sealed sources should contain the following three sections:

- o A. Summary Data
- o B. Descriptive Data
- o C. Health and Safety Data

Guidelines for these three sections are presented below and are followed by Section D -- Specifications and Style.

A. SUMMARY DATA

This section can normally be presented on one page and should contain key summary data as follows:

1. Date: Give the date of submission.

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2. Sealed Source Type: Insert the short name commonly used by the manufacturer/distributor to identify the source.

3. Model: Insert the model number(s) or series number(s) used by the vendor to identify the sealed source.

4. Applicant: Give the name and complete mailing address of the organization submitting the application and indicate whether it is the manufacturer or distributor or both. Also give the name, title and telephone number of the person to be contacted for further information.

5. Other Companies Involved: Give the name and address of any other companies directly involved in the manufacture or distribution of this sealed source. For example, if the applicant distributes a device manufactured by the XYZ Company list the XYZ Company, Mfr., and give the mailing address.

6. Isotope and Maximum Activity: List the isotope(s) approved for use in a sealed source and the maximum acceptable activity level in terms of curies or millicuries for each approved isotope. If depleted uranium is used for shielding, show the number of grams of depleted uranium used.

7. Leak Test Frequency: State the recommended frequency for testing the sealed source for possible leakage of radioactive material. (More detailed testing information will be presented in Section C.)

8. Principal Use: Select from the attached list of principal uses (Exhibit 1) the term which most accurately describes the principal or predominant use intended for the sealed source or device.

9. Custom Source: Indicate by a "Yes" or "No" whether the sealed source is a custom source. If the answer is "Yes", present the basis for this determination. Sealed sources specifically designed and constructed according to the personal order of a single specific license applicant may be considered "CUSTOM" sealed sources for the purpose of a review tailored

to the single applicant. Sealed sources designed and constructed as off-the-shelf items or for use by more than a single license applicant shall not be deemed applicable to custom reviews and shall not be considered for a custom review and registration.

10. Custom User: If this is a custom source, give the name and address of the custom user.

B. DESCRIPTIVE DATA

This section should include the following:

1. Summary Description: Provide a precise, yet concise, description of the sealed source, including information on the chemical and physical form of the radioactivity, the materials used in the capsule construction, capsule dimensions and the methods for fabrication and sealing of the capsules. State the American National Standards Institutes (ANSI) classification designation of the source. Do not include information which has been determined to be "proprietary data." (See Exhibit 2, "Proprietary Data," for definition and guidance on the handling of proprietary data.)

2. Labeling: Describe the information to be engraved, etched or imprinted on a sealed source and the type of location of warning labels. The label for a sealed source should include the words: "CAUTION - RADIOACTIVE MATERIAL," manufacturer's name or trademark, model number or unqiue serial number, radionuclide, activity, assay date, and the radiation symbol. Where labeling the source is impracticable, a tag containing the above information should be attached to the source, unless the attachment of such a tag is also impracticable. When a sealed source is permanently mounted in a device, source labeling is not required, provided the device is labeled as specified above.

3. Diagram: Insert a small drawing of the sealed source showing the materials of construction, dimensions, method of sealing, and relationships of major components. Do not include information which has been determined to be "proprietary data." The diagram should be no larger than 4" by 6" and should be suitable for reproduction for use in a registration sheet.

4. Conditions of Normal Use: Describe the planned use of the sealed source and identify the environment and operating conditions expected during normal use. Include descriptions of the types of users, location of use, possibilities of use as a component in other products, and circumstances of normal use. Indicate the expected useful life of the source. Describe also the probable effects of severe conditions, including accidents and fires, and possible diversion from intended use.

5. Supporting Detail: Provide additional descriptive information which may be helpful in conveying to the reviewer a clear understanding of the sealed source and its detailed characteristics. This should include a design package containing engineering drawings of the sealed source, identifying all methods of construction, dimensions, methods of fabrication and method of sealing the source capsule(s).

If the information presented in the application contains data which the applicant considers to be proprietary data, such data should be clearly marked so that it can be handled appropriately. In addition, the letter transmitting the application should call attention to the inclusion of proprietary data. See Exhibit 2, "Proprietary Data," for definition and guidance on the handling of proprietary data.

Provide references to other pertinent documents, including previous applications and registration sheets.

C. HEALTH AND SAFETY DATA

This section should include the following:

1. Safety Analysis Summary: Provide a paragraph which summarizes the important facts pertaining to safety and the results of the safety analysis performed by the manufacturer/distributor. Include references to the appropriate ANSI, NBS or NRC standards used in the safety analysis.

2. Manufacturing and Distribution Controls: Describe the manufacturing and distribution controls applicable to the sealed source, giving attention to the following:

   a. Quality Assurance and Control: Describe the quality control procedures to be followed in the fabrication of production lots of the sources, as applicable, and the quality control standards for maintaining source design specifications.

   Describe the assay method used to determine the radioactive content of the sealed source. This method shall be traceable to a national standard.

   Each manufacturer, assembler, or distributor shall perform a leak test on each source by applying procedure(s) in the current ANSI Standard entitled, "Classification of Sealed Radioactive Sources." Acceptability of source leakage shall be indicated by removal of less than 0.005 microcuries.

   b. Description of Manufacturer's Recommended Maintenance, Servicing, and Testing Requirements for Use: Describe the manufacturer's recommendation for leak testing, unpacking, handling and disposal of the sealed source and specify availability of these services.

   The normal leak test interval is six months. In the event the manufacturer, assembler, or distributor requests that a sealed source, upon transfer to the user, be considered for a leak test interval greater than six months, sufficient information shall be submitted to demonstrate that such a longer interval is justified as a result of operating experience with identically sealed sources or similarly designed and constructed sealed sources used in similar conditions.

c. Manufacturer's Instructions to Users: Manufacturers or distributors of sealed sources distributed under these registration procedures should provide users with a copy of pertinent radiological safety and operating instructions for the source.

3. <u>Manufacturer's Safety Analysis of Sealed Source Review</u>: Each application for a sealed source review shall include a section which contains the manufacturer's Safety Analysis Report. This report shall contain, but not be limited to, the following information.

a. Safety Analysis: The analysis should determine the ability of the final design to withstand the normal condition of handling, use, and storage including such factors as abrasion, corrosion, vibration, impact, puncture, and the probable effects on containment of abnormal conditions such as fire or explosion.

b. Prototype Testing and Evaluation: Submit the following information:

- Maximum radiation levels at 5 and 30 centimeters from any external surface of the source averaged over an area not to exceed 100 square centimeters, and the method of measurement or calculation.
- Results of tests performed on prototype sources that establish the integrity of the source construction and seal under the most adverse conditions of use to which the source is likely to be subjected. These prototype tests should, insofar as possible, reflect the actual conditions of use and, as a minimum, shall meet the designated usage classification according to the current ANSI standard entitled "Sealed Radioactive Sources, Classification," provided the means for assigning such a classification is described.

c. Additional Information: Submit any additional information, including results of experimental studies and tests, which will facilitate a determination of the safety of the sealed source.

D. SPECIFICATIONS AND STYLE

Review, handling and filing of applications can be facilitated by observance of the following guidelines on specifications and style.

1. Physical Specifications

All pages in an application should be numbered consecutively. Text pages should preferably be printed on two sides with the image printed head to head.

If revisions are necessary subsequent to submission of an application, revised pages should be submitted. Each revised page should be numbered and show the date of revision. The revised portion of the page should be marked by a bold verticle line in the margin opposite the binding margin. If supplemental pages are submitted as part of the revision they may be numbered 13a, 13b, etc.

The preferred paper size is 8½ x 11 inches. If a larger size is used, the sheet, after reduction, should not exceed 11 x 17 inches, including a 2-inch margin at the left for binding. The finished copy when folded should not exceed 8½ x 11 inches.

A margin of no less than one inch should be maintained on the top, bottom and binding side of each sheet.

All drawings should have a drawing number, revision number, company name, title, date, and sheet number.

Type of paper, color of paper and ink, type font and style, and printing or reproduction method should be suitable for microfilming.

2. Style and Composition

The applicant should strive for clear, concise presentation of the information provided in the application. Confusing or ambiguous statements and unnecessarily verbose descriptions do not contribute to expeditious technical review. Claims of adequacy of designs or design methods should be supported by technical bases, i.e., by an appropriate engineering evaluation or description of actual tests. Terms as defined in the NRC regulations and American National Standards guides must be used.

Appendices may be used to include detailed information omitted from the main text for clarity. Examples of such information are summaries of the manner in which the applicant has treated matters addressed in NRC regulatory guides, supplementary information regarding calculational methods or design approaches used by the applicant or its agents, and lists of references mentioned in the text.

All physical tests of sealed source and devices should be supported by photographs in the appendices.

Where numerical values are stated, the number of significant figures given should reflect the accuracy or precision to which the number is known. Where possible, estimated limits of error or uncertainty should be given. Significant figures should not be dropped or rounded off if, by doing so, subsequent conclusions are inadequately supported.

Abbreviations should be consistent throughout the application and should be consistent with generally accepted usage. Any abbreviations, symbols, or special terms unique to the proposed sealed source or device not in general usage should be defined in each section of the application where they are used.

Drawings, diagrams, sketches, and charts should be used where the information can be presented more adequately or conveniently by such means. Due concern should be taken to ensure that all information presented in drawings is legible, symbols are defined, and drawings are not reduced to the extent that visual aids are necessary to interpret pertinent items of information presented in the drawings.

Revised

STANDARD LIST
PRINCIPAL USES OF SEALED SOURCES AND DEVICES

CODE	
A	Industrial Radiography
B	Medical Radiography
C	Medical Teletherapy
D	Gamma Gauges
E	Beta Gauges
F	Oil Well Logging
G	Portable Moisture Density Gauges
H	General Neutron Source Applications
I	Calibration Sources (Activity greater than 30mCi)
J	Gamma Irradiator, Category I
K	Gamma Irradiator, Category II
L	Gamma Irradiator, Category III
M	Gamma Irradiator, Category IV
N	Ion Generators, Chromatography
O	Ion Generators, Static Eliminators
P	Ion Generators, Smoke Detectors
Q	Thermal Generator
R	Gas Sources
S	Foil Sources
T	Other
U	X-Ray Fluorescence
V	General Medical Use

EXHIBIT 1

DEFINITIONS FOR STANDARD LIST
PRINCIPAL USES OF SEALED SOURCES AND DEVICES

CODE	
A	Industrial Radiography -- The examination of the structure of materials by nondestructive methods, utilizing sealed sources of radioactive material.
B	Medical Radiography -- The process of producing x-ray or gamma-ray images to assist in the determination of medical diagnoses.
C	Medical Teletherapy -- The treatment of disease with gamma radiation from a controlled source of radiation located at a distance from the patient.
D	Gamma Gauges -- The use of gamma radiation to measure or control thickness, density, levels, interface location, radiation leakage, or chemical composition.
E	Beta Gauges -- The use of beta radiation to measure or control thicknesss, density levels, interface location, radiation leakage, or chemical composition.
F	Oil Well Logging -- The lowering and raising of measuring devices or tools which may contain radioactive sources into well bases or cavities for the purpose of obtaining information about the well and/or adjacent formations.
G	Portable Moisture Density Gauges -- Portable gauges which use a radioactive sealed source to determine/measure moisture content or density of material. This includes hand-held or dolly-transported devices/sources.
H	General Neutron Source Applications -- All applications, excluding reactor start-up, which use a neutron source.
I	Calibration Sources (Activity greater than 30mCi) -- Sources of a known purity and activity which are used to determine the variation in accuracy of a measuring instrument and to ascertain necessary correction factors.

EXHIBIT 1 (continued)

CODE	
J	Gamma Irradiator, Category I -- An irradiator in which the sealed source(s) is completely contained in a dry container constructed of solid materials, the sealed source is shielded at all times, and human access to the sealed source(s) and the volume(s) undergoing irradiation is not physically possible in its design configuration.
K	Gamma Irradiator, Category II -- All applications which are panoramic and use dry source storage for irradiation of biologic or other materials.
L	Gamma Irradiator, Category III -- Applications which are self contained and use a wet source storage for irradiation of biologic and other materials.
M	Gamma Irradiator, Category IV -- Applications which are panoramic and use a wet source storage for irradiation of biologic and other materials.
N	Ion Generators, Chromatography -- Process of using an ion generating source to determine the chemical composition of material.
O	Ion Generators, Static Eliminators -- Process of using ion generating sources to eliminate static electricity on a surface or a surrounding area.
P	Ion Generators, Smoke Detectors -- Process of using ion generating sources to detect gases and particles created by combustion.
Q	Thermal Generator -- Process of using the heat of a radioisotope to produce energy.
R	Gas Sources -- Sealed sources containing radioactive gas such as krpton-85 or hygrogen-3.
S	Foil Sources -- Sources which are constructed using thin metal foil. The radioactive material may be secured to the foil in a number of ways, for example: plating, laminating, or cold welding.
T	Other -- All other uses or applications not covered in other categories.

EXHIBIT 1 (continued)

CODE	
U	<u>X-Ray Fluorescence</u> -- Sources and/or devices utilizing radioactive material which excites the atoms of samples which, in turn, emit characteristic x-rays and thereby provide a means for sample analysis.
V	<u>General Medical Use</u> -- This category includes diagnostic sources and devices such as bone mineral analyzers and therapeutic sources and devices such as interstitial needles, therapeutic seeds, and opthalmic applicators.

EXHIBIT 1 (continued)

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   The wording set forth below shall be placed at the bottom of the front cover and title page, or first page of text if there is no front cover or title page:

   "TRADE SECRET OR PRIVILEGED OR CONFIDENTIAL COMMERCIAL OR FINANCIAL INFORMATION"

EXHIBIT 2

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which is claimed to be proprietary in accordance with (10 CFR 2.790(b)) (10 CFR 9.5) (10 CFR Part 21) and is exempt from mandatory public disclosure to 10 CFR Part 9.

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SOURCE EFFICIENCY CALCULATIONS FOR ^{137}Cs IRRADIATORS

by

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PACIFIC NORTHWEST LABORATORIES
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SUMMARY

The efficiency of radiation sources used for radiation treatment is important to the overall economics of irradiators. This report examines design optimization for a Cesium-137 (^{137}Cs) radiation source. The initial source design used is currently being fabricated at the Waste Encapsulation and Storage Facility (WESF). Although the current source was intended for long term storage and isolation of the cesium, it has potential for use as an irradiator.

Modifications to the source which were examined include:

- Source Diameter - The optimum cylinder diameter is 1 in.
- Wall Thickness - The optimum stainless steel wall thickness is about 0.1 in. Thicker walls will reduce source efficiency, while thinner walls increase the likelihood of wall failure.
- Cesium Product Form - A cesium-titanate ceramic combines the advantages of good source efficiency with low solubility and low leachability.

The cost to modify the present WESF facility does not appear to justify these design changes. However, new facilities could incorporate this design to obtain optimum source efficiency.

CONTENTS

INTRODUCTION

Radiation treatment can be used beneficially in the treatment of food(1) and the thermal radiation of sewage sludge(2) and medical products.(3) Potential sources of radiation include electron accelerators, ^{60}Co, which is produced in reactors and ^{137}Cs, which is recovered from the reactor fuel wastes.

The Waste Encapsulation and Storage Facility (WESF) currently operated by Rockwell Hanford Operations produces sealed capsules of either radioactive cesium chloride or strontium fluoride.(4) The purpose of the WESF plant is to encapsulate the radioactive cesium and strontium which is recovered from the aqueous wastes generated from the reprocessing of reactor fuel elements at the Hanford Site. The encapsulation of these elements is an initial step in the management of these nuclear fuel wastes.

The radioactive compounds are placed in capsules designed for safe long term storage. The desired characteristics for the capsule are good heat dissipation, thick capsule walls, and a size large enough to reduce the number of capsules needed to contain the required quantity of compound. The cesium product form selected was based on chemical process characteristics.

Since the original CsCl capsule was designed for storage rather than an irradiator, it seemed possible that the capsule design could be improved for use as an irradiator. The economics of irradiators are dependent on the efficiency of the radiation source; hence, a significant increase in the source efficiency would result in a significant improvement in irradiator economics. Because of this, studies were made for the Nuclear By-Product Utilization Program sponsored by the Department of Energy. This paper describes the types of design changes examined and gives the effect of these changes on the source efficiency.

CONCLUSIONS

The results of these source efficiency calculations indicate that an increase in the efficiency of the WESF cesium capsules is possible. This would require changes in the cylinder diameter and wall thickness. To reduce the possibility of environmental contamination when the capsules are used in irradiators, it may be desirable to change the cesium compound form especially if capsule wall thickness is decreased. Compound form change would reduce slightly the source efficiency. The net efficiency change would be an increase from 60% for the present design to 72% for the titanate ceramic in a 1-in. diameter source with 0.1-in. wall.

To evaluate the cost-effectiveness of the proposed changes, the extra expense of more sources with increased handling and fabrication costs and the cost of WESF facility modifications need to be examined. Also, the technical feasibility of such process changes must be considered. At this point, the costs that would be incurred in increasing source efficiency by 1/5 (60% to 72%) do not appear to be justified.

Before WESF production is committed to a new Cs capsule design and product form, additional work in capsule design and cost analysis is recommended. Capsule design should be made compatible with irradiator facility design. This design analysis effort would have to evaluate not only the dose rate to the material undergoing irradiation, but also biological shielding necessary to protect the operators and the public.

PRESENT CAPSULE DESIGN

The WESF cesium capsule is a double-walled stainless steel cylinder. A view of the capsule is shown in Figure 1 and the capsule characteristics are given in Table 1.

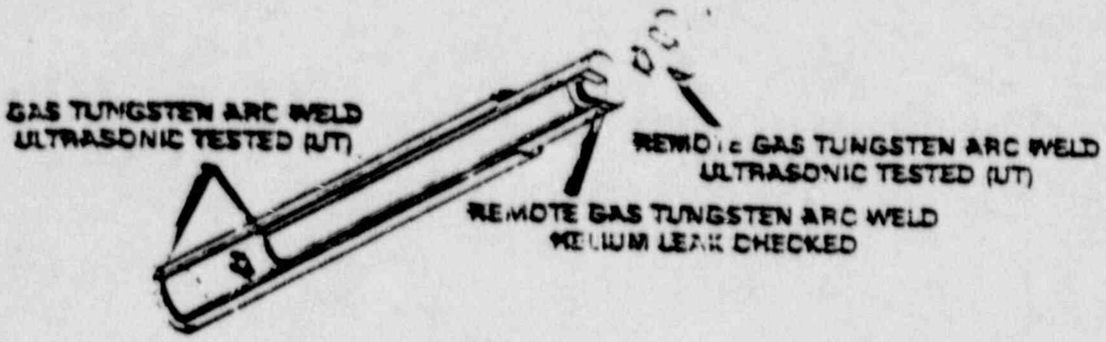

FIGURE 1. WESF Capsule

TABLE 1. WESF Cesium Chloride Capsule Characteristics

DIMENSIONS	
Inner Capsule:	316-L stainless steel 0.095-in. wall thickness 2.250-in. outside diameter 19.725-in. total length 0.400-in. total cap thickness
Outer Capsule:	316-L stainless steel 0.109-in. wall thickness 2.625-in. outside diameter 20.775-in. total length 0.400-in. total cap thickness
DENSITY CsCl	2.47 g/cm^3
QUANTITY OF ^{137}Cs	48-49 wt%
IMPURITIES	4-6 wt%
HEAT GENERATION	300 watts
RADIOACTIVITY	70,000 Ci ^{137}Cs

Dosimetry measurements were performed on one of the WESF cesium capsules to serve as a benchmark for the calculations. The experimental dose rates are listed in Table 2. These measurements were made with thermoluminescent dosimeters (TLD's). TLD-700 (LiF) chips were placed at the midplane of the capsule. A 10.3-minute exposure was made and a control set of TLD's was used to give a true 10.3-minute exposure.

TABLE 2. Measured Radial Dose Rates from Cesium Chloride Source

Distance from Surface of Source, cm	Dose Rate, R/hr
20	302,000
30	168,900
50	55,300

THEORY

The computer code used for dose rate calculations was QAD-P5A.(5) This is a "point-kernel" code. It provides the uncollided gamma flux density, dose rate, and energy deposition rate which result from a point-by-point representation of a volume distributed radiation source. The volume source is divided into a large number of point isotropic sources and the traversal distances through each material are calculated for each point source. Based on the energy dependent attenuation for each material, the uncollided gamma flux density at a receiver point from a source point is calculated(6) by

$$\phi(R) = S \frac{e^{-\mu(E)R}}{4\pi R^2} \tag{1}$$

where

S = source strength for the source point (γ/sec)

$\mu(E)$ = linear attenuation coefficient at the initial energy

$e^{-\mu(E)R}$ = material attenuation factor: i.e., the probability that a gamma of energy E travels a distance R without a collision.

$4\pi R^2$ = geometric attenuation for point source.

Our version of QAD-PSA uses the gamma interaction coefficients of Storm and Israel(7) and Berger's(8) energy fluence-to-exposure conversion factors.

It is important to realize that $\phi(R)$ in equation (1) is the uncollided flux density. This approximates the total flux density only when scattered gammas are completely removed from the radiation beam which is effectively realized only for thin shields. For thick shields, the probability increases that multiple collisions will occur with gammas scattering to the receiver point giving flux densities higher than those calculated by Equation (1).

Calculation of the scattered flux density is complex and it is handled by introducing a buildup factor which increases the flux density to account for scattered gamma rays. This buildup factor is a function of the shield material, gamma energy, and the quantity (i.e., flux density or dose) being calculated. Hence

$$B = \frac{[\text{property of total flux density at } R]}{[\text{same property of uncollided flux density at } R]}$$

Now

$$\phi_T(R) = B(\mu R)S\,\frac{e^{-\mu(E)R}}{4\pi R^2} \qquad (2)$$

where

$B(\mu R)$ = flux density buildup factor

$\phi_T(R)$ = total flux density.

Similar techniques are used in QAD-PSA where the buildup factors are based on the Goldstein and Wilkins moments method of infinite homogeneous media.(9) The Goldstein-Wilkins data are fitted by a polynomial expansion whose simplified form is

$$B_i(\mu R) = a_0 + a_1\mu R + a_2(\mu R)^2 + a_3(\mu R)^3 \qquad (3)$$

where

$B_i(\mu R)$ = the buildup factor for the ith energy group at μR mean free paths.

a_0, a_1, a_2, a_3 = energy dependent coefficients from polynomial fits to Goldstein-Wilkins data.

When alternate layers of different materials are part of a shield, additional problems occur. Because the gamma ray spectrum changes with penetration in a medium (due to absorption and scattering), the emergent spectrum is dependent on the arrangement of the shielding layers. QAD-P5A allows the selection of one of eight different buildup factors:

- H_2O Dose Buildup Factor
- Al Dose Buildup Factor
- Fe Dose Buildup Factor
- Pb Dose Buildup Factor
- H_2O Energy Absorption Buildup Factor
- Al Energy Absorption Buildup Factor
- Fe Energy Absorption Buildup Factor
- Pb Energy Absorption Buildup Factor.

For the capsule itself, the iron buildup factor was selected. The actual buildup factor used is then determined from a polynomial fit for the total μR using the constants for iron in equation (3). In cases where the dose rate was calculated for the capsule in water, an additional case was made using water buildup factors. The iron buildup factor was then used for points within the capsule while the buildup factor for points in the water region was found by

$$B = B_r^{Fe} + (B_R^W - B_r^W) \qquad (4)$$

where

B_r^{Fe} = iron buildup factor at the outside capsule radius

B_R^W = water buildup factor at R in water region

B_r^W = water buildup factor at the outside capsule radius.

While equation (4) is a very rough approximation, it is useful and reasonably accurate for similar cases. It is much simpler than other techniques used to obtain multiple region buildup factors. The only cases in which this technique is important are the water media dose rates. For air, the buildup is small and the efficiency calculations are taken at the surface of the source.

Considerable detail has been provided in the discussion of buildup because it is important to the efficiency results that follow; the effect of the buildup factor is also significant. At the outside of the present capsule design, the buildup factor is approximately 1.5 and with the capsule in water at 100-cm depth it is 37. Thus at the surface of the capsule, one-third of the dose is from scattered gamma rays while after 100 cm of water all but 3% of the dose is from scattered gamma rays. In addition, the buildup factor influences the source efficiency which tempers the effect of any change in capsule design.

QAD-PSA accepts boundary surface equations which fit the form

$$AX^2 + XX_0 + BY^2 + YY_0 + CZ^2 + ZZ_0 - K = r \qquad (5)$$

As only cylinders and planes are required to model the capsule geometry, an exact representation is possible. The model used for all designs consisted of three regions

- source region (e.g. CsCl at the expected density)
- capsule wall (stainless steel whose thickness equals the total wall thickness)
- outside medium (water or air).

The point source distribution was chosen such that increasing the number of source points resulted in no significant change in the calculated dose rates. This ensures that the error in dose due to the finite distribution of source kernels is less than 5%.

CALCULATIONS

The calculated dose rates from the present WESF capsule are given in Figures 2 to 4. In Figure 2, the dose rates in air and water are plotted for radial distances up to 100 cm from the center line of the source. In Figures 3 and 4 the dose rates are plotted along several radial and axial traverses, again in both water and air.

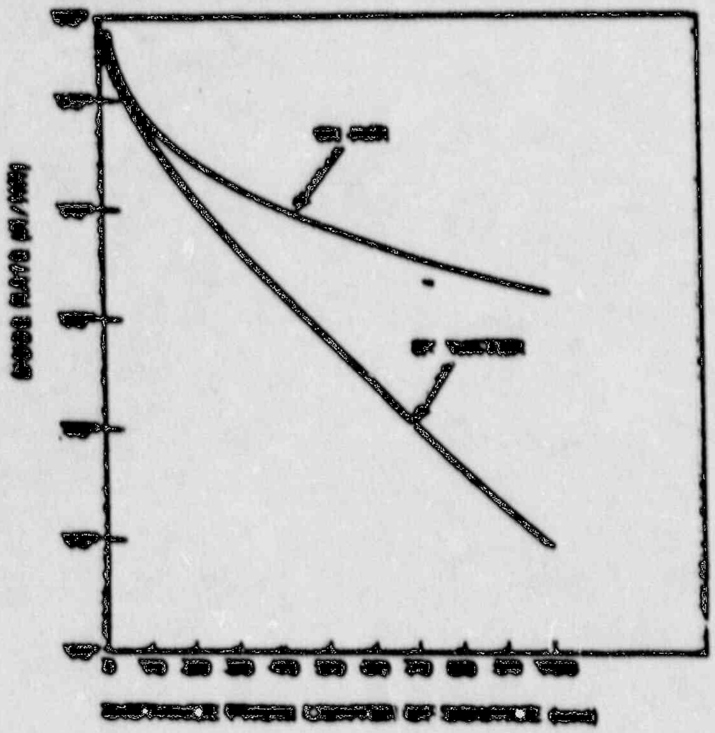

FIGURE 2. Dose Rate From Cesium Chloride Source - Midplane

The dosimetry measurements from Table 2 agree quite well with the calculated dose rates. The experimental dose rates are listed in Table 3 along with the calculated values from Figure 2 air values. Due to the interpolation required for the 10-cm value, a 10% error is possible at this point.

Additional comparisons were made with dose rates estimated by RHO (Rockwell Hanford Operations formerly Atlantic Richfield Hanford Co.,)[4] and from data in the Cesium Data Sheets.[10] The RHO curves are difficult to interpolate accurately for a distance as small as 100 cm, but the calculated dose rates in Table 3 are very close. In the Cesium Data Sheets a spherical source of 300 thermal watts with 1 cm of iron shielding gives a dose rate of approximately 10^7 mrad/hr at 100 cm. For the WESF geometry with 1 cm of iron shielding QAD-P5A calculates a dose rate of 1.4×10^7 mrad/hr at 100 cm. Some difference can be attributed to reduced self shielding for the WESF cylindrical capsule compared to the spherical model used in the Cesium Data Sheets.

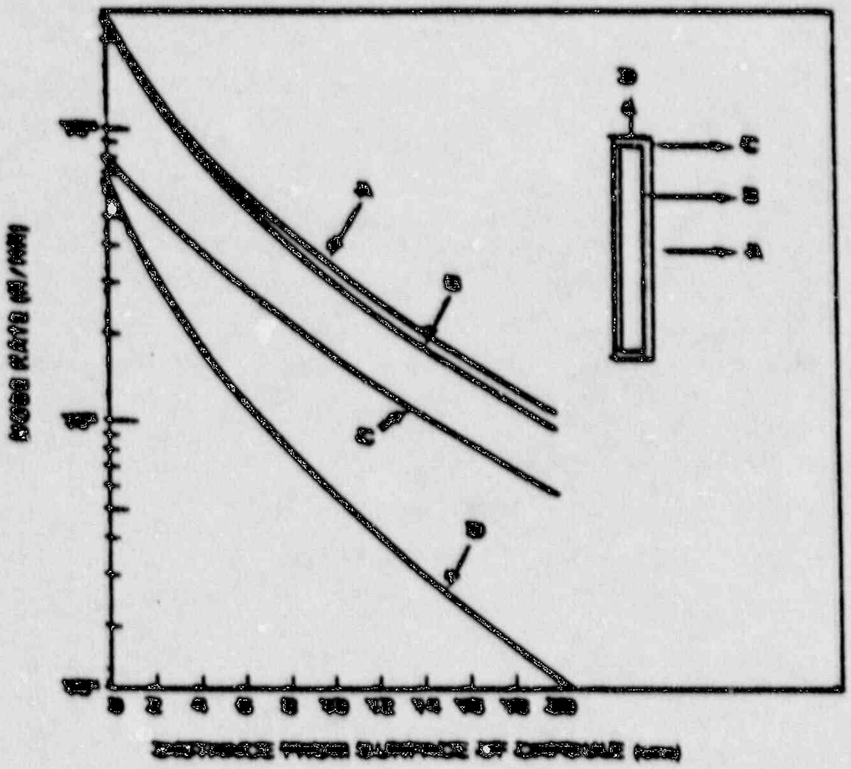

FIGURE 3. Dose Rate From Cesium Chloride Source In Water

DESIGN MODIFICATIONS

The term efficiency is used to describe and compare the different source designs. For these calculations, efficiency is defined as the ratio of the dose rate at the capsule surface with attenuation in the source region and capsule wall to the dose rate at the surface without attenuation in the source region and capsule wall. This definition was originally used in similar studies at Sandia Laboratories. There is no buildup factor applied to the dose rate calculation with no attenuation since no attenuation implies that there are no scattered gamma rays. If the buildup factor was not applied to the calculation made with attenuation, the result would be the efficiency for the uncollided gamma rays. Because the scattered gammas also contribute significantly to the dose rate, and in turn the source efficiency, the buildup factor must be included. For simple comparisons between various source designs, one might suspect the buildup factor would not be needed; however, changes in efficiency caused by modifications in the self shielding or wall attenuation are magnified when the buildup factor is not used. Also, it should be noted that the use of the dose buildup factor may tend to slightly overestimate the biological effectiveness since real efficiency is more related to energy transmission than to dose rate.

Two characteristics of the capsule affect its efficiency as an irradiator. These are the geometry of the capsule and the form of the cesium product. The source geometry parameters include the diameter, wall thickness, and shape. Included in the analysis to ascertain the potential improvements in capsule design were:

- the effect of wall thickness
- the effect of cylinder diameter
- the effect of an annular cylinder design
- the effect of a plate source design.

Decreasing the wall thickness would obviously result in reduced attenuation in the wall. Because long term storage is not required in an irradiator, thinner walls are possible, but this might require a change in

TABLE 3. Dose Rates From Cesium Chloride Sources

Distance from Surface of Source, in	Measured, R/hr	Calculated, R/hr
20	302,000	300,500
30	158,300	160,500
50	55,300	51,000

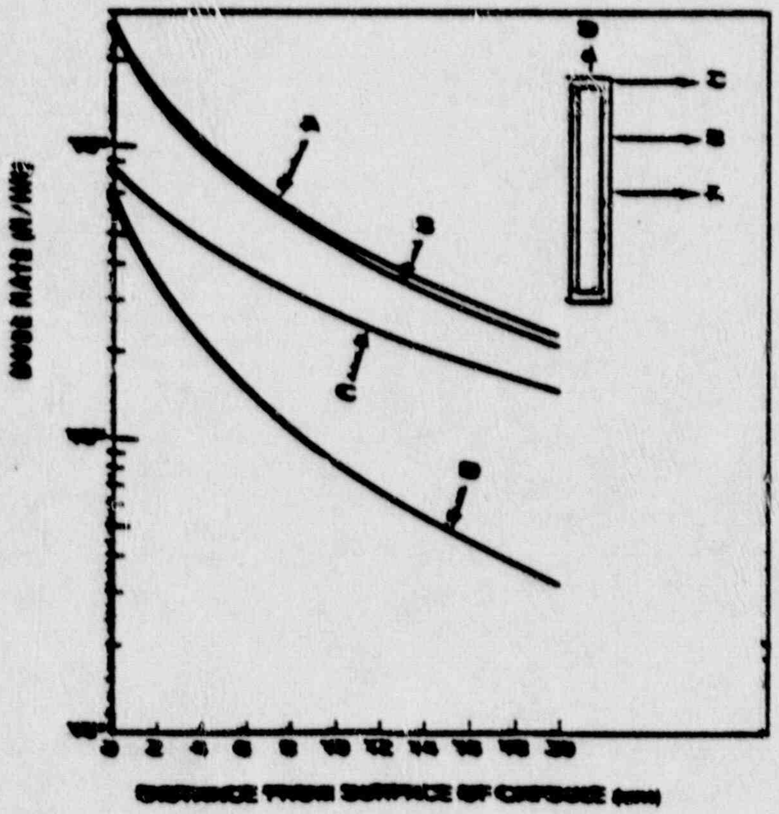

FIGURE 4. Dose Rate From Cesium Chloride Source in Air

product form because of the relatively high water solubility of CsCl. Reducing the cylinder diameter would result in reduced self shielding (or attenuation within the source material itself). Similarly, it was thought that making the source an annular cylinder with a void space in the center would place source material closer to the capsule surface and reduce self shielding. Also such a design would require fewer production changes to WESF. Finally, changing the source to a flat plate might make irradiator design simpler.

The efficiencies for cylinders with various radii and wall thicknesses are listed in Table 4. The highest efficiencies are possible with cylinders having the smallest diameter and the thinnest wall as expected. However, since the number of capsules required for a given volume of compound varies inversely with the square of the diameter, an effective lower limit is reached for the smallest practical diameter. In addition, fabrication and source handling are easier with a few large sources rather than a large number of small irradiators. Evaluation of these factors indicates that the cost effective design is a cylinder with a diameter of approximately 1 in. with 0.1-in. wall thickness. Reducing the diameter to 0.5 in. results in only a 4% increase in efficiency but requires the handling and fabrication of four times as many sources. A minimum wall thickness of 0.1-in. is necessary for encapsulation.

TABLE 4. Cylinder Efficiencies

Source CsCl Diameter, in.	Wall Thickness, in.	Efficiency, %	Efficiency Increase Over Present Design, %
2.05	0.204	60	0[a]
2.05	0.100	69	15
1.0	0.100	77	28
1.0	0.204	67	12
0.5	0.204	71	18

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a. Present Capsule Design

Due to the reduction in self shielding which might be possible with an annular cylinder source geometry, calculations were also made for annular cylinders (2.625-in. OD, 0.2-in. clad); these results are summarized in Table 5. Apparently efficiency is not increased by using an annular geometry. This is probably due to an increase in the absorption in the wall from the larger wall-to-source volume ratio with thin annular regions. The ratio of source-to-clad volume decreases faster with a reduction in radius for annular regions than for cylindrical regions.

TABLE 5. Annular Cylinder Efficiency

Radial Thickness, in.	Efficiency, %
1.0	60
0.5	60
0.3	61

Calculations for a plate source geometry, summarized in Table 6, are based on a 40-cm square plate with a 0.1-in. SS wall thickness. The edge effects for the finite-sized geometry are more important for plate sources than for cylinders. The value at the wall surface approximates the efficiency at any location for an infinite plate source. Since any practical irradiator will be finite in size with a limiting distance over which irradiation will occur, these efficiency values are lower limits. As much as a 10% efficiency increase is possible for smaller irradiators. Compared with cylindrical sources, the efficiencies show no worthwhile increase, thus no significant gains are realized by switching to a plate source geometry. The only reason to change the design to a plate source would be to simplify irradiator fabrication, but this is offset by the major WESF equipment modifications which would be required.

TABLE 6. Plate Source Efficiency

Source Thickness, cm	Efficiency, %
0.5	38
1.0	30
2.0	28

Sandia Laboratories has an ongoing program on the thermoradiation of sewage sludge using radioactive cesium.(2,11) Since the product form used in WESF (CsCl) has a high solubility in water (2 kg/liter), a small leak in a source capsule could cause contamination. Sandia has expressed interest in isolating the radioactive cesium in a high temperature/low leachability compound such as a zeolite or stable ceramic. Table 7 lists the compounds of interest, the bulk density, and the cesium density for each. The bulk densities are from a Sandia Laboratory report(11) except for cesium chloride which is from RHO estimates(4) for the WESF capsule.

TABLE 7. Compound Density

Compound	Density	
	g compound/cm^3	g Cs/cm^3
$Cs_2Ti_4O_9$	4.0	1.8
$Cs_2Nb_4O_{11}$	4.7	1.5
$Cs_2Zr_4O_9$	4.5	1.5
$Cs_8(Al_8Si_{40}O_{96})$	2.5	0.7
CsCl	2.47	1.95

Table 8 lists the efficiencies and dose rates at the clad surface for sources of the various compounds. The source geometry is the same for all compounds (diameter of 1.0 in. with 0.1 in. SS clad) the highest efficiency occurs with zeolite and cesium chloride. In all cases the dose rate is normalized to the present WESF cesium content. In comparing these dose rates, the titanate has the highest surface dose rate except for cesium chloride. The low cesium density in the $Cs_8(Al_8Si_{40}O_{96})$ zeolite reduces the dose rate considerably and thus would require many additional sources for the same level of irradiation.

TABLE 8. Source Efficiency

Compound	Efficiency, %	Dose Rate, R/hr At Clad Surface
$Cs_2Ti_4O_9$	72	1.45×10^5
$Cs_2Nb_4O_{11}$	69	1.25×10^5
$Cs_2Zr_4O_9$	70	1.28×10^5
$Cs_8(Al_8Si_{40}O_{96})$	77	0.62×10^5
CsCl	77	1.75×10^5

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ANSI N542(1972)

Compliance with the tests shall be determined by the ability of the sealed source to maintain its integrity after each test is performed. Methods of testing sources for integrity after testing are set out in Appendix A.

A source with more than one encapsulation shall be deemed to have complied with a test if it can be demonstrated that at least one encapsulation has maintained its integrity after the test.

4.1.2 Classification of Radionuclides According to Radiotoxicity (Table 2). This Table, taken from a current ICRP report, classifies radionuclides into four groups according to relative radiotoxicity.

If the radionuclide is not listed in Table 2, the source shall be considered on an individual basis by the supplier, the user, the regulatory authority, or governmental advisory agency.

4.1.3. Activity Level (Table 3). This Table establishes a maximum activity of sealed sources for each of the four radiotoxicity groups in Table 2, without further evaluation. Sources containing more than the maximum activity shall be subject to further evaluation of the specific usage and design. The activity shall be determined at the time of manufacture of the sealed source.

Table 3 also defines the properties of the radioactive material within the sealed source as "leachable and/or reactive" and "non-leachable and nonreactive." The physical and chemical form and the geometrical shape of the radioactive material used to determine these properties shall be the same as the physical, chemical and geometrical form of the radioactive material within the sealed source.

4.1.4 Sealed Source Performance Requirements for Typical Usage (Table 4). Table 4 is based on current practice and typical environments in which a sealed source or source-device will be used. Average environment includes normal and abnormal use (taking into account reasonable accidental risks), but does not include exposure to fire or explosion. For sealed sources normally mounted in devices, consideration was given to the additional protection afforded the sealed source by the device when the Class number for a particular usage was assigned. Thus, for all usages shown in Table 4, the Class numbers specify the tests to which the sealed source shall be subjected, except that for the category ion generators, the complete source-device combination may be tested.

Obviously, Table 4 does not cover all source usage situations. If the particular average environment differs from the values shown, or if the source usage is not shown, the specifications of the source shall be considered on an individual basis by the supplier, the user, and the regulatory authority. The numbers shown in Table 4 refer to the Class numbers used in Table 1.

4.1.5 Leak Test Methods (Appendix A). Appendix A lists currently acceptable leak test (integrity test) methods.

4.1.6 Quality Assurance and Control (Appendix B). To assure that production sources will have performance characteristics equal to the tested prototypes used in classifying the sources, a good Quality Assurance and Control program is necessary. Appendix B is included as a guide to aid a manufacturer in establishing a specific program.

4.1.7 Summary of ANSI N44.1—1973. Integrity and Test Specifications for Selected Brachytherapy Sources (Appendix C).

4.1.8 Summary of ANSI N540—1975. Classification of Self-Luminous Light Sources (Appendix D).

4.1.9 IAEA Tests for Special Form Radioactive Material. Excerpts from IAEA Safety Series No. 6—Regulations for the Safe Transport of Radioactive Materials, 1973 Revised Edition (Appendix E).

4.2 Fire, Explosion or Corrosion. Table 4 does not consider exposure of the source-device to fire, explosion or corrosion. In the evaluation of sealed sources and source-device combinations, the manufacturer and user must consider the probability of fire, explosion and corrosion and the possible results. Factors which should be considered in determining the need for actual testing are:

(1) consequence of loss of activity;
(2) quantity of active material contained in the source;
(3) radiotoxicity;

(4) chemical and physical form and the geometrical shape of the radioactive material;
(5) environment in which it is used; and
(6) protection afforded the source or source-device combination.

4.3 **Radiotoxicity and Solubility.** Except as required in Section 4.2 radiotoxicity of the radionuclide shall be considered only when the activity of the sealed source exceeds the value shown in Table 3. If the activity exceeds this value, the specifications of the source must be considered on an individual basis. If the activity does not exceed the values shown in Table 3, Table 4 may be used without further consideration of either radiotoxicity or solubility.

5. Procedure To Establish Classification and Performance Requirements

5.1. Establish radiotoxicity group from Table 2.

5.2 Determine amount of activity allowable from Table 3.

5.3 If the desired quantity does not exceed the allowable quantity of Table 3, an evaluation of fire, explosion, and corrosion probabilities shall be made. If no significant probability with respect to fire, explosion, and corrosion exists, the required classification for the source (performance requirements) may be taken directly from Table 4. If a significant probability does exist, the factors listed in 4.2 shall be evaluated with particular attention to the temperature and impact requirements.

5.4 If the desired quantity exceeds the allowable quantity of Table 3, an evaluation of fire, explosion or corrosion probability and a separate evaluation of the specific source usage and source design shall be made.

5.5 After the required classification of the source for the particular application or usage has been established, the performance test conditions can be obtained directly from Table 1.

5.6 Alternatively, the source may be tested, the source Class determined from Table 1, and some suitable application selected from Table 4. Sources of an established classification may be used in any application having less severe specific performance requirements (classification numbers).

6. Identification

The designation according to section 3 shall be marked on the sealed source or source container or source holder or accompanying document.

7. Testing Procedures for Table 1

7.1 **General.** The testing procedures given in this section present acceptable procedures for determining performance classification numbers. All the test environments provide the minimum requirements. Procedures which can be demonstrated to be at least equivalent are also acceptable. All tests, except the temperature tests, shall be carried cut at ambient temperature.

7.2 Temperature Test [1]

7.2.1 **Equipment.** The heating or cooling equipment shall have a test zone volume of at least five times the volume of the test specimen. If a gas or oil-fired furnace is used for the temperature test, an oxidizing atmosphere shall be maintained throughout the test.

7.2.2 **Procedure.** All tests shall be performed in air except in the low temperature test, when an atmosphere of carbon dioxide is permitted. All test sources shall be held at the maximum (or minimum, for low temperature tests) test temperature for a period of at least 1 h.

Although Table 1 specifies a low temperature of −40°C, "dry ice" may be used as the cooling material. Thus, the low temperature may approach −75°C.

Sources to be subjected to temperatures below ambient shall be cooled to the test temperature in less than 45 minutes.

Sources to be subjected to temperature above ambient shall be heated to the test temperature at least as rapidly as indicated by the following time-temperature table.

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[1] Part of this test for Class 6 is similar in principle to the heating test given in IAEA regulations for the safe transport of radioactive materials.

Chemical Engineers' Handbook

JOHN H. PERRY
Editor of First, Second, and Third Editions

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sodium silicate based
750°F.

TURE MATERIALS

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mber of steels are made
ls A 201 and A 212 are
for −50°F. service.
203 Grade A or B for
for service to −150°F.

m-nickel steels are in-
om −300° to −425°F.
grades used at low tem-
t popular. The original
gher than another metal.
-treatment, and welding,
offsets the higher initial

and Alloys for
e Service*

ale	Recommended Max. Service Temp., °F.
	−50
	−50
	−75
	−150
	−320
47	−425
356, 5454, 5456	−425
copper	−320

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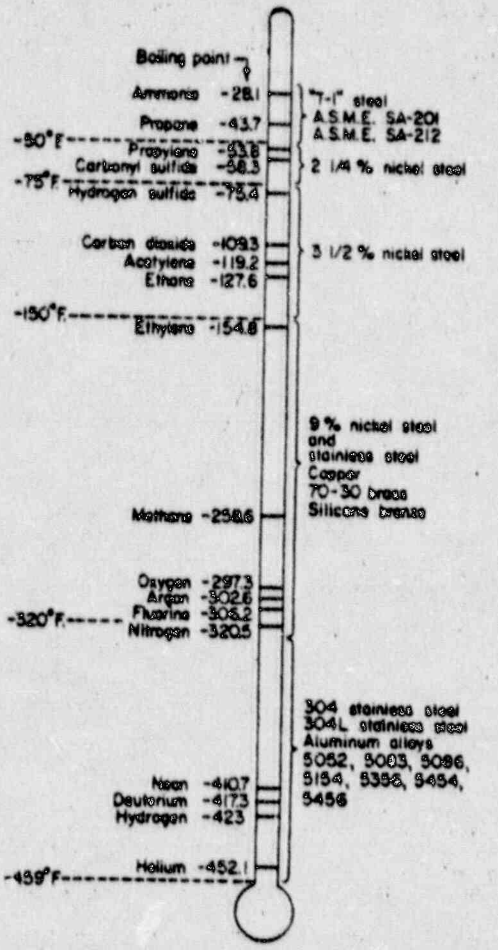


FIG. 23-1. What steels to use as the temperature drops. This chart shows schematically the various steels used for low-temperature applications in environments defined by the normal boiling points of certain liquefied gases.

cost. Sensitization or formation of chromium carbides can occur in several stainless steels during welding, and this will affect impact strength. However, tests have shown that impact properties of types 304 and 304L are not greatly affected by sensitization, but the properties of 302 are impaired at −300°F.

Nickel Steel. Low-carbon 9 per cent nickel steel is a ferritic alloy developed for use in cryogenic equipment operating as low as −320°F. A.S.T.M. specifications A 300 and A 353 cover low-carbon 9 per cent nickel steel (A 300 is the basic specification for low-temperature ferritic steels). Refinements in welding and (A.S.M.E. Code approved) elimination of postweld thermal treatments make 9 per cent steel competitive with many low-cost materials used at low temperatures.

Aluminum. Aluminum alloys have an unusual ability to maintain strength and shock resistance at temperatures as low as −425°F. Good corrosion resistance and relatively low cost make these alloy very popular for low-temperature equipment. For most welded construction the 5000 series aluminum alloys are widely used. These are the aluminum-magnesium and aluminum-magnesium-manganese materials.

Copper and Alloys. With few exceptions the tensile strength of copper and its alloys increases quite rapidly as the temperature goes down. However, copper's low structural strength becomes a problem when constructing large-scale equipment. Therefore, alloys must be used. One of the most successful for low temperatures is silicon bronze. The alloy can be used to −320°F. with safety.

Insulation. Perlite, balsa wood, cellular glass, and mineral wool can all be used at temperatures down to about −260°F.

But still better insulations are needed to cope with liquid hydrogen, oxygen, and helium. Randomly dispersed reflective flakes are not good enough. Two very promising insulations have been developed by Linde.

One is a medium-quality variety consisting of 10 to 30 layers of aluminum foil per inch, separated by 3-micron glass-fiber mat. It is a relatively low-density (2 lb./cu. ft.) low-cost insulation with a thermal conductivity of 0.11 × 10^{-3} B.t.u./(hr.)(ft.)(°F.) between 80° and −297°F. A higher-quality variety consists of 40 to 80 layers/in. of aluminum foil and submicron glass-fiber paper. This is somewhat heavier than the other insulation (4.7 lb./cu. ft.) and more costly, but it has the very low thermal conductivity of 0.025 × 10^{-3} B.t.u./(hr.)(ft.)(°F.) over the same temperature range.

High-temperature Materials. Successful applications of metals in high-temperature process service depend on an appreciation of certain engineering factors. The important alloys for service up to 2000°F. are shown in Table 23-15.

Table 23-15. Important Commercial Alloys for High-temperature Process Service

	Nominal composition, %			
	Cr	Ni	Fe	Other
Ferritic steels:				
Carbon steel	....		bal.	
2¼ chrome*	2¼	....	bal.	Mo
Type 502	5	....	bal.	Mo
Type 410	12	....	bal.	
Type 430	16	....	bal.	
Type 446	27	....	bal.	
Austenitic steels:				
Type 304	18	8	bal.	
Type 321	18	10	bal.	Ti
Type 347	18	11	bal.	Cb
Type 316	18	12	bal.	Mo
Type 309	24	12	bal.	
Type 310	25	20	bal.	
Type 330	15	35	bal.	
Nickel-base alloys:				
Nickel	....	bal.		
Incoloy	21	32	bal.	
Hastelloy B	....	bal.	6	Mo
Hastelloy C	16	bal.	6	W, Mo
60/15	15	bal.	25	
Inconel	15	bal.	7	
80/20	20	bal.		
Hastelloy X	22	bal.	19	Co, Mo
Multimet	21	20	bal.	Co
Rene 41	19	bal.	5	Co, Mo, Ti
Cast irons:				
Ductile iron	....	....	bal.	C, Si, Mg
Ni-Resist, D-2	2	20	bal.	Si, C
Ni-Resist, D-4	5	30	bal.	Si, C
Cast stainless (ACI types):				
HC	28	4	bal.	
HF	21	11	bal.	
HH	26	12	bal.	
HK	26	20	bal.	
HT	15	35	bal.	
HW	12	bal.	28	
Super alloys:				
Inconel X	15	bal.	7	Ti, Al, Cb
A 286	15	25	bal.	Mo, Ti
Stellite 25	20	10	Co-base	W
Stellite 21 (cast)	27.3	2.8	Co-base	Mo
Stellite 31 (cast)	25.2	10.5	Co-base	W

* See Table 23-44 for data on other heat-resistant low- and medium-alloy steels.

Table 23-8. Chemical Resistance of Important Plastics

	Polypropylene polyethylene	CAB*	ABS†	PVC‡	Saran§	Polyester glass¶	Epoxy glass	Phenolic asbestos	Fluorocarbons	Chlorinated polyether (Penton)	Polycarbonate
10% H_2SO_4	Excel.	Good	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.
50% H_2SO_4	Excel.	Poor	Excel.	Excel.	Excel.	Good	Excel.	Excel.	Excel.	Excel.	Excel.
10% HCl	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.
10% HNO_3	Excel.	Poor	Good	Excel.	Excel.	Good	Good	Fair	Excel.	Excel.	Excel.
10% Acetic	Excel.	Good	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.
10% NaOH	Excel.	Fair	Excel.	Good	Fair	Fair	Excel.	Poor	Excel.	Excel.	Excel.
50% NaOH	Excel.	Poor	Excel.	Excel.	Fair	Poor	Good	Poor	Excel.	Excel.	Excel.
NH_4OH	Excel.	Poor	Excel.	Excel.	Poor	Fair	Excel.	Poor	Excel.	Excel.	Excel.
NaCl	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.
$FeCl_3$	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.
$CuSO_4$	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.
NH_4NO_3	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Good	Excel.	Excel.	Excel.
Wet H_2S	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	
Wet Cl_2	Poor	Poor	Excel.	Good	Poor	Poor	Poor	Excel.	Excel.	Excel.	
Wet SO_2	Excel.	Poor	Excel.	Excel.	Good	Excel.	Excel.	Excel.	Excel.	Excel.	
Gasoline	Poor	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.
Benzene	Poor	Poor	Poor	Poor	Fair	Good	Excel.	Excel.	Excel.	Fair	Fair
CCl_4	Poor	Poor	Poor	Fair	Fair	Excel.	Good	Excel.	Excel.	Fair	Poor
Acetone	Poor	Poor	Poor	Poor	Fair	Poor	Good	Poor	Excel.	Good	Good
Alcohol	Poor	Poor	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.	Excel.

Ratings are for long-term exposures at ambient temperatures (less than 100°F.).
* Cellulose acetate butyrate.
† Acrylonitrile butadiene styrene polymer.
‡ Polyvinyl chloride, type I.
§ Chemical resistance of Saran-lined pipe is superior to extruded Saran in some environments.
¶ Refers to general-purpose polyesters. Special polyesters have superior resistance, particularly in alkalies.

brittle and hence do not stand up well under shock and impact. Also these irons are more difficult to machine. Threads must be formed by direct casting. Welding is possible, although Durichlor is more difficult to weld than Duriron. Special welding rods and heat-treatments must be used.

Silicon irons are very resistant to oxidising and reducing environments, and resistance depends on the formation of a passive film. These irons are widely used in sulfuric acid service, since they are unaffected by sulfuric at all strengths, even up to the boiling point.

The Mo-containing iron is especially recommended for hydrochloric acid, although presence of oxidized chlorides such as ferric chloride will cause pitting.

Because they are very hard, silicon irons are good for combined corrosion-erosion service.

To overcome some of the mechanical disadvantages of silicon irons, a number of other cast alloys have been developed. One, **Hastelloy D**, contains about 82 per cent Ni, 3 per cent Cu, 9 per cent Si, 2 per cent Fe. This alloy has superior resistance to mechanical and thermal shock over silicon irons. Machining is easier than with silicon irons. The alloy is recommended for sulfuric acid service, since it is practically unaffected by all concentrations of sulfuric up to and including the boiling point. However, it is not recommended for nitric or chromic acids.

Another group of cast-iron alloys are called **Ni-Resist**. These materials are related to gray cast iron in that they have high carbon contents (3 per cent), with fine graphite flakes distributed throughout the structure. Nickel contents vary from 13.5 to 36 per cent, and some have 6.5 per cent Cu.

Generally, nickel-alloy castings have superior toughness and impact resistance compared with gray irons. The nickel-alloy castings can be welded and machined.

Corrosion resistance of nickel alloys is superior to cast irons, but less than that of pure nickel. There is little attack from neutral or alkaline solutions. Oxidising acids such as nitric are highly detrimental. Cold, concentrated sulfuric acid can be handled.

Ni-Resist has excellent heat resistance, with some grades serviceable up to 1500°F. Also, a ductile variety is available, as well as a hard variety (Ni-Hard).

Stainless Steel. There are more than 70 standard types of stainless steel and many special alloys. These steels are produced in the wrought form (A.I.S.I. types) and as cast alloys (A.C.I.) types. Generally, all are iron-based, with 12 to 30 per cent chromium, 0 to 22 per cent nickel, and minor amounts of carbon, columbium, copper, molybdenum, selenium, tantalum, and titanium. These alloys are very popular in the process industries. They are heat- and corrosion-resistant, non-contaminating, and easily fabricated into complex shapes.

There are three groups of stainless alloys: (1) martensitic, (2) ferritic, (3) austenitic.

The **martensitic** alloys contain 12 to 20 per cent chromium with controlled amounts of carbon and other additives. Type 410 is a typical member of this group. These alloys can be hardened by heat-treatment, which can increase tensile strength from 80,000 to 200,000 lb./sq. in.

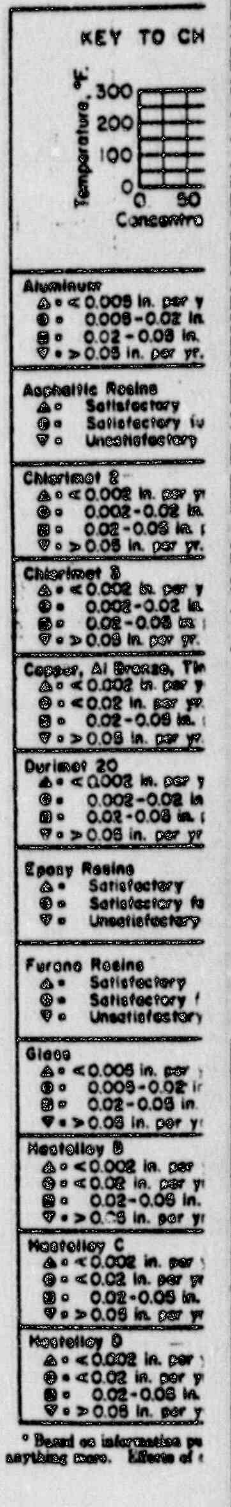
Corrosion resistance is inferior to austenitic stainless steels, and martensitic steels are generally used in mildly corrosive environments (atmospheric, fresh water, and organic exposures).

Ferritic stainless contains 15 to 30 per cent Cr, with low carbon content (0.1 per cent). The higher chromium content improves its corrosive resistance. Type 430 is a typical example. The strength of ferritic stainless can be increased by cold working but not by heat-treatment. Fairly ductile, ferritic grades can be fabricated by all standard methods. They are fairly easy to machine with high-speed equipment. Welding is no problem, although it requires skilled operators.

Corrosion resistance is rated good although ferritic alloys are not good against reducing acids such as HCl. But mildly corrosive solutions and oxidising media are handled without harm. Type 430 is widely used in nitric acid plants. In addition it is very resistant to scaling and high-temperature oxidation up to 1500°F.

Austenitic stainless steels are the most corrosion-resistant of the three groups. These steels contain 16 to

(Continued on page 23-48)



° Based on information pu
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Table 23-6. American Iron and Steel Institute Standard Steels. Compositions*

Introduction. The information presented in the following tables represents the manufacturing practices of the many steel producers which have been reported to the American Iron and Steel Institute and related scientific and technical information which has been obtained from other sources as further described. These practices evolved from the utilization of available raw materials and manufacturing methods to produce steel products to meet the requirements of purchasers' specifications and standards. The latter comprise specifications and standards of individual purchasers, consuming industries, standardization bodies, and specification writing bodies, including the United States government.

Although the institute is not a specification writing body, it participates with standardization bodies and specification writing bodies in the preparation of standards and specifications. Metallurgical engineers employed by steel producers participate by serving on committees of bodies which issue standards and specifications. The results of such participation are ordinarily identified by the name of the body or branch of the government, such as, for example, "A.S.T.M. Standard Specifications," "A.P.I. Standard," "A.S.M.E. Specification," "S.A.E. Standard," "A.A.R. Specification," "American Standard," "Military Specifications" and "Federal Specifications." The technical aspects of standards and specifications are prominent in the A.I.S.I. manual.

In some of the sections of the manual, some of the numbered grades of steel are identified as "A.I.S.I. Standard Steels." They are standards which embrace only the chemical composition ranges and limits of such grades. The history of such grade numbers can be traced from the standardization efforts of the Automobile Manufacturers Association early in this century to the present work by the Society of Automotive Engineers and the institute. Other bodies, including the United States government, have contributed information on properties and uses of steel products which has aided the establishment of numbered grades as standards. Among others, individual purchasers, consumers and engineers have, by their advice and recommendations, participated in the evolution of A.I.S.I. standard steels.

* Based on information from American Iron and Steel Institute's "Steel Products Manual."

Table 23-6a. American Iron and Steel Institute Standard Steels: Ladle Chemical Ranges and Limits for Basic Open-hearth and Acid Bessemer Carbon Steels

A.I.S.I. No.	Chemical composition limits, %			
	C	Mn	P, max.	S, max.
C 1008	0.10 max.	0.25/0.50	0.040	0.050
C 1010	0.08/0.13	0.30/0.60	.040	.050
C 1011	0.08/0.13	0.60/0.90	.040	.050
C 1012	0.10/0.15	0.30/0.60	.040	.050
C 1015	0.13/0.18	0.30/0.60	.040	.050
C 1016	0.13/0.18	0.60/0.90	.040	.050
C 1017	0.15/0.20	0.30/0.60	.040	.050
C 1018	0.15/0.20	0.60/0.90	.040	.050
C 1019	0.15/0.20	0.70/1.00	.040	.050
C 1020	0.18/0.23	0.30/0.60	.040	.050
C 1021	0.18/0.23	0.60/0.90	.040	.050
C 1022	0.18/0.23	0.70/1.00	.040	.050
C 1023	0.20/0.25	0.30/0.60	.040	.050
C 1024	0.19/0.25	1.35/1.65	.040	.050
C 1025	0.22/0.28	0.30/0.60	.040	.050
C 1026	0.22/0.28	0.60/0.90	.040	.050
C 1027	0.22/0.29	1.20/1.50	.040	.050
C 1029	0.25/0.31	0.60/0.90	.040	.050
C 1030	0.28/0.34	0.60/0.90	.040	.050
C 1031	0.28/0.34	0.30/0.60	.040	.050
C 1033	0.30/0.36	0.70/1.00	.040	.050
C 1035	0.32/0.38	0.60/0.90	.040	.050
C 1036	0.30/0.37	1.20/1.50	.040	.050
C 1037	0.32/0.38	0.70/1.00	.040	.050
C 1038	0.35/0.42	0.60/0.90	.040	.050
C 1039	0.37/0.44	0.70/1.00	.040	.050
C 1040	0.37/0.44	0.60/0.90	.040	.050
C 1041	0.36/0.44	1.35/1.65	.040	.050
C 1042	0.40/0.47	0.60/0.90	.040	.050
C 1043	0.40/0.47	0.70/1.00	.040	.050
C 1045	0.43/0.50	0.60/0.90	.040	.050
C 1046	0.43/0.50	0.70/1.00	.040	.050
C 1049	0.46/0.53	0.60/0.90	.040	.050
C 1050	0.48/0.55	0.60/0.90	.040	.050
C 1051	0.45/0.56	0.85/1.15	.040	.050
C 1052	0.47/0.55	1.20/1.50	.040	.050
C 1053	0.48/0.55	0.70/1.00	.040	.050
C 1055	0.50/0.60	0.60/0.90	.040	.050
C 1060	0.55/0.65	0.60/0.90	.040	.050
C 1070	0.65/0.75	0.60/0.90	.040	.050
C 1078	0.72/0.85	0.30/0.60	.040	.050
C 1080	0.75/0.88	0.60/0.90	.040	.050
C 1084	0.80/0.93	0.60/0.90	.040	.050
C 1085	0.80/0.93	0.70/1.00	.040	.050
C 1086	0.80/0.93	0.30/0.50	.040	.050
C 1090	0.85/0.98	0.60/0.90	.040	.050
C 1095	0.90/1.03	0.30/0.50	.040	.050

26 per cent chromium, 6 to 22 per cent nickel. Carbon is kept low (0.08 per cent) to minimize carbide precipitation. These alloys can be work-hardened, but heat-treatment will not cause hardening. Tensile strength in the annealed condition is about 85,000 lb./sq. in., but work hardening can increase this to 300,000 lb./sq. in. Austenitic stainless steels are tough and ductile.

They can be fabricated by all standard methods. But austenitic grades are not easy to machine—they work-harden and gall. Rigid machines, heavy cuts and high speeds are essential. Welding, however, is readily performed, although welding heat may cause chromium carbide precipitation, which depletes the alloy of some chromium and lowers its corrosion resistance. For mild service, this is not serious, but for severe corrosive service, the carbides must be put back into solution by heat-treatment. To avoid precipitation, special stainless steels stabilized with titanium, columbium, or tantalum have been developed (types 321, 347, 348). Another approach to the problem is the use of low-carbon steels such as types 304L and 316L, with 0.03 per cent maximum carbon.

Type 302 is the basic alloy of this group. Types 304 and 304L are low-carbon versions of 302. Types 316, 316L, and 317, with 2.5 to 3.5 per cent molybdenum, are the most corrosion-resistant.

In the stainless group, nickel greatly improves corrosion resistance over straight chromium stainless. Even so, the chromium-nickel steels, particularly the 18-8 alloys, perform best under oxidizing conditions, since resistance depends on an oxide film on the surface of the alloy. Reducing conditions and chloride ions destroy this film and bring on rapid attack. Chloride ions, combined with high tensile stresses, will cause stress-corrosion cracking.

Austenitic stainless steels have excellent resistance to nitric acid at practically all concentrations and temperatures. Most nitric acid plants are constructed with type 304. To handle sulfuric acid without inhibitors, stainless steel (type 316) can be used only below 5 per cent and above 85 per cent at temperature below the boiling point.

Cast stainless alloys are widely used in pumps, valves, and fittings. These casting are designated under the Alloy Casting Institute system. All corrosion-resistant alloys have the letter C plus a second letter (A to N)

Table 23-6b. American Iron and Steel Institute Standard Steels: Ladle Chemical Ranges and Limits for Basic Open-hearth Resulfurized Carbon Steels*

A.I.S.I. No.	Chemical composition limits, %			
	C	Mn	P, max.	S
C 1108	0.08/0.13	0.50/0.80	0.040	0.08/0.13
C 1109	0.08/0.13	0.60/0.90	.040	0.08/0.13
C 1110	0.08/0.13	0.30/0.60	.040	0.08/0.13
C 1113	0.10/0.16	1.00/1.30	.040	0.24/0.33
C 1115	0.13/0.18	0.60/0.90	.040	0.08/0.13
C 1116	0.14/0.20	1.10/1.40	.040	0.16/0.23
C 1117	0.14/0.20	1.00/1.30	.040	0.08/0.13
C 1118	0.14/0.20	1.30/1.60	.040	0.08/0.13
C 1119	0.14/0.20	1.00/1.30	.040	0.08/0.13
C 1120	0.18/0.23	0.70/1.00	.040	0.08/0.13
C 1125	0.22/0.28	0.60/0.90	.040	0.08/0.13
C 1126	0.23/0.29	0.70/1.00	.040	0.08/0.13
C 1132	0.27/0.34	1.35/1.65	.040	0.08/0.13
C 1137	0.32/0.39	1.35/1.65	.040	0.08/0.13
C 1138	0.34/0.40	0.70/1.00	.040	0.08/0.13
C 1139	0.35/0.43	1.35/1.65	.040	0.12/0.20
C 1140	0.37/0.44	0.70/1.00	.040	0.08/0.13
C 1141	0.37/0.45	1.35/1.65	.040	0.08/0.13
C 1144	0.40/0.48	1.35/1.65	.040	0.24/0.33
C 1145	0.42/0.49	0.70/1.00	.040	0.04/0.07
C 1146	0.42/0.49	0.70/1.00	.040	0.08/0.13
C 1151	0.48/0.55	0.70/1.00	.040	0.08/0.13

* For easy machining.

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Addendum to
SAND 82-1492

WESF CESIUM-137 GAMMA RAY SOURCES

The following information is provided to amplify and clarify material presented in SAND82-1492 and is based, in part, upon data obtained since the publication of the document.

Irradiator Classification Category:

As discussed in the Registration Application, most contemplated cesium-137 irradiator designs require intermittent exposure of the source capsules to a water environment during (1) initial loading, (2) periodic recharging and/or (3) operation where source is not in use. However, some facilities such as the TransPortable Cesium Irradiator (TPCI) will not require such exposure to water and will operate as self-contained, dry source storage systems.

Because most irradiator designs require source exposure to water, Category III of ANSI N542 is appropriate for these facilities. In these systems, the cesium-137 is self-contained within the double encapsulation of the WESF capsules and, through irradiator design, is isolated from the environment in an irradiation chamber. The sources are occasionally exposed to water so Category III - self-contained, wet source storage is appropriate even though in most designs the sources will only be exposed to water for a small fraction (less than 1 percent) of their useful lives in these facilities. In a facility such as TPCI, Category I, self-contained, dry source storage is the appropriate classification. It should be noted that the ANSI N542 performance test requirements are identical for sources used in Categories II, III and IV irradiators.

Puncture Test Requirement:

The ANSI N542 requirement for puncture testing for sources in Category III irradiators is Class 4 (50 gms from 1 m). Paragraph 3.2.5 on page 25 incorrectly cites 10 gms in the first line. However, note that the test conducted and survived by the WESF capsule involved dropping the capsule itself (approximately 7,600 gms) onto a steel rod from 4.57 meters.

Vibration Test Requirement:

Currently contemplated irradiator designs will restrain the WESF capsules rigidly in a source plaque which is housed in a massive concrete structure or an approved shipping cask

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(as in TPCI). In these environments, vibration of the sources is extremely unlikely. During shipment to the facilities, the sources will be transported in approved regulatory containers and would experience only those low-level vibrations experienced by a truck on a highway which can be transmitted through the cask to the capsules. However, SNLA has conducted a vibration test of a WESF capsule (loaded with non-radioactive CsCl) according to the Category III ANSI N542 requirements (30 min, 25-500 Hz @ 5 g peak amplitude). Preliminary visual results indicate no effect of this loading on the capsule integrity. Helium leak checks of the welds are currently being conducted [Phone conversation - McMullen/Kenna - 8/4/83].

Temperature Test Requirement:

The static temperature test requirement for Category III irradiator sources is -40°C for 20 minutes. As noted on page 22, due to the internal heat generation through decay of the cesium-137 within the WESF capsule (approximately 300 watts), it is very difficult to achieve a -40°C condition on the external surface of the outer capsule. In fact, with a doubly encapsulated WESF cesium chloride capsule generating 300 watts of decay heat, an external still air temperature of -200 to -400°F is required to cool the outer capsule surface to -40°C.

Quality Assurance:

Fabrication of the WESF cesium chloride sources is accomplished by Rockwell-Hanford Operations under strict procedures. The 316L stainless steel capsule tubing is procured according to Rockwell specification HWS-8835 outlining physical, chemical, mechanical and dimensional parameters which must be met. Rockwell specifications H2-66760 and H2-66761 outline procedures for inner and outer capsule fabrication while Rockwell document SDWM-OCD-003 establishes welding criteria for capsule welds. With regard to encapsulation of the cesium chloride at WESF, Rockwell has published an "Operating Specifications Document for B Plant and WESF." Three pertinent specifications are PSD-B-257-00053 (Rev. D-0) which defines process parameters and purity levels for preparation of the molten cesium chloride; PSD-B-257-00054 (Rev. D-0) which outlines procedures for capsule welding, leak checking, decontamination and calorimetric analysis; and PSD-B-257-00055 (Rev. D-0) which establishes procedures for storage of the capsules in the WESF pool.

Capsule Destructive Analysis:

As discussed on page 26, ORNL did destructively analyze a cesium WESF capsule. This capsule was produced and filled with cesium chloride by DOE/Rockwell-Hanford in 9/75, was stored in the WESF pool from 9/75 to 8/78, was shipped in an NRBK 43 cask to SNLA in 8/78, was stored in a SNLA pool from 8/78 to 5/79, was loaded and used in the Sandia Irradiator for Dried Sewage Solids from 5/79 to 8/81 and was shipped to ORNL for destructive analysis on 8/81. During the 2 years of use in the SIDSS, the capsule was one of 15 arranged in a source plaque and resided in air for the major part of the time. The results of the ORNL analysis are presented in SAND83-0928. The measured inner capsule outer surface temperature upon opening at ORNL was 127°C (Table II, page 15). The outer capsule exterior surface temperature was 104°C. The source contained about 60,000 Ci of cesium-137 (page 24). [redacted]: ([illegible] surface temperatures are in °F and not °C as stated, and ([illegible] inner capsule contained 61.8 kCi cesium-137, not 71.8 kCi. Metallographic, SEM, microprobe, gas analysis and mechanical tests were conducted on the sectioned capsule and, as the report states on page 26, "the cesium chloride appears to be a benign resident within the capsule to this point in time." In effect, no notable corrosion phenomena were observed and no effect on mechanical properties due to any contact with CsCl was discovered after 8 years in various operational environments.

Recycle of Cesium:

The comments on page 32, paragraph 4.1 referring to recycling of cesium chloride pertain to operating procedures at Rockwell-Hanford by which newly encapsulated sources can be reopened, the CsCl remelted and reintroduced into the process for subsequent filling of another capsule. These procedures would be followed if, typically, one of the weld tests (leak check or ultrasonic test) indicated a substandard weld on one of the capsules.

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Product Irradiators in Non-Agreement States

All of the irradiators listed below are wet storage type. All of the irradiators contain cobalt-60.

	Licensee	Location of Irradiator	Qua~~l~~ntity (Ci)	Use
1.	Radiation Sterilizers	Schaumberg, IL	5,000,000	Medical products
2.	Radiation Sterilizers	Westerville, OH	5,000.000	Medical products
3.	Appliant Radiant Energy Corp.	Lynchburg, VA	400,000	Materials
4.	Radiation Technology	Rockaway, NJ	2,000,000	Products, wood
5.	Isomedix	Northborough, MA	2,000,000	Materials, food
6.	Isomedix	Vega Alta, PR	3,000,000	Medical products
7.	Isomedix	Groveport, OH	2,000,000	Materials
8.	Isomedix	Morton Grove, IL	500,000	Products
9.	Isomedix	Whippany, NJ	2,000,000	Materials
10.	Isomedix	Parsippany, NJ	2,000,000	Materials
11.	Ethicon	Somerville, NJ	2,000,000	Medical products
12.	Becton-Dickinson	North Canaan, CT	1,500,000	Medical products
13.	Permagrain Products	Karthaus, PA	2,000,000	Products

Product Irradiators in Agreement States

All of the irradiators listed below are wet storage type. All of the irradiators contain cobalt-60. The specific products which may be irradiated

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were not specified except for food irradiation.

	License	Location of Irradiator	Quality (Ci)	Food
14.	Process Technology	West Memphis, AR	2,500,000	Yes for export
15.	Sherwood Medical	Northfolk, NE	3,000,000	No
16.	Becton-Dickinson	Broken Bow, NE	3,000,000	No
17.	Ethicon	San Angelo, TX	1,500,000	No
18.	Johnson & Johnson	Sherman, TX	3,000,000	No
19.	American Convertors	El Paso, TX	10,000,000	No
20.	Surgikose	Arlington, TX	4,000,000	No
21.	Sherwood Medical	Commerce, TX	3,000,000	No
22.	Neutron Products	Dickinson, MD	750,000	Yes - R & D
23.	Neutron Products	Dickinson, MD	1,500,000	Yes - R & D
24.	COBE Laboratories	Lakewood, CO	5,000,000	No
25.	Radiation Sterilizers	Tustin, CA	3,000,000	Yes
26.	Becton-Dickinson	Oxnard, CA	3,000,000	No
27.	International Nutronics	Irvine, CA	1,250,000	No
28.	International Nutronics	Palo Alto, CA	750,000	No
29.	Buckeye Cellulose	Huntsville, AL	3,000,000	No
30.	Sherwood Medical	Deland, FL	3,000,000	No

31.	Radiation Sterilizers	Decatur, GA (under construction)	5,000,000	No
32.	Isomedix	Columbus, MS	2,000,000	No
33.	Process Technology	Haw River, NC	1,200,000	No
34.	Isomedix	Spartanburg, SC	4,00,000	No
35.	Becton-Dickinson	Sumter, SC	3,000,000	No

Accidents

At the Isomedix facility in Parsippany, NJ, a worker entered a hot cell irradiation facility while the radiation source was in an unshielded configuration. The licensee's procedure required surveys to be made prior to entering the hot cell. In addition, a radiation alarm was supposed to be activated if the door was opened and the sources were in an unshielded configuration. The alarm was not automatically engaged if the door was opened; the radiation monitor which would activate the alarm was external to the cell. The procedures also required that a visual check be made through the hot cell viewing window to determine that the radiation source was in a shielded position prior to entering into the cell. Because of fatigue and human error, the worker failed to follow the procedures, entered into the cell, and received a dose of about 300 rem.

At the Radiation Technology facility in Rockaway, NJ, the interlock system which was intended to sound an alarm if the door was opened while the radiation source was in an unshielded configuration was inactivated by removal of the door. The licensee continued operations even though the safety system had been rendered inoperable. A worker, unaware that the radiation source was unshielded, entered into the irradiation source while the radiation source was exposed. The worker failed to make a survey because he thought the radiation source was in the shielded position. The worker received a dose of about 300 rem.

Fires

At the Becton-Dickinson facility in Broken Bow, Nebraska, the source rack was prevented from returning to the shielded position by product carrier boxes which had bent out corner edges and flaps that extended into the travel path of the source rack. After about 10 hours, several fires occurred in the product and product boxes. The first were extinguished by the sprinkler system in the irradiation room. After approximately 24 hours, the source rack was successfully lowered to its shieleded position. There was no damage to the source pencil; there was some damage to the source pass conveyor that caused several days of down time.

An event similar to the Broken Bow event occurred at the Becton-Dickinson facility at North Canaan, Connecticut.

Contamination

A spill and release of water contamination with cobalt-60 occurred at the International Nutronics facility in Dover, New Jersey. A pump malfunction caused pool water, which had been contaminated due to a leaking source, to be released into the facility and into areas adjacent to the building. The licensee discontinued all irradiator operations. The facility and surrounding area are in the process of decontamination pursuant to an NRC order.

Physics-chemical Character of ^{137}CsCl-316L Stainless Steel

A. Currently-used WESF capsules were designed for water storage, not as gamma radiation sources.

B. Chromium-nicked steels perform best under oxidizing conditions, since resistance (to corrosion) depends on an oxide film on the surface of the alloy. Reducing conditions, and chloride ions, destroy this film and bring on rapid attack. Chloride ions, combined with high tensile stresses, will cause stress-corrosion cracking. This is true in aqueous solution and needs to be studied in the dry system at elevated temperature.

C. The effects of loss of capsule integrity can be significant due to the high solubility of CsCl and the specific activity of ^{137}Cs. For instance:

1. At about 30°C. the solubility of CsCl is 200 grams per 100 grams of water.
2. Less than 0.003 mg of ^{137}Cs would contaminate a 30,000 gallon storage pool to levels beyond those permitted for release to an unrestricted area.
3. If a resin column is considered a point source, about 4 mg. of absorbed ^{137}Cs would result in a gamma radiation level of 1 R/hr. at 1 meter.

D. A long-term (≈5 years) compatability testing program on WESF capsules is underway at PNL.

1. The chloride is about 90% Cs and 10% chlorides of potassium. rubidium, sodium, calcium, and barium.
2. Theoretical considerations indicate that certain impurities in CsCl could have a significant effect on corrosion rate.
3. Early results (≈6 months aging) at 450°C interface temperature show 316L corrosion rates of about 0.008 inches/year.
4. Definite conclusions must await the results of the longer-term tests.
5. These tests are more indicative of effects in dry-storage, dry-irradiation facilities.

E. A Cesium-137 demonstration at Sandia (dry-storage, dry-irradiation) has operated for more than five years irradiating grapefruit, mangoes, and sewage sludge. This experience will be factored into licensing decisions.

F. The use of WESF capsules in wet-storage, dry-irradiation would impose thermal cycling effects.

1. The CsCl-stainless interfacial termperature undergoes about a 140°C change between water storage and use in air.
2. This temperature differential causes an expansion (and contraction) of about 0.014 inches of the circumference at the interface.
3. If a capsule is used for 1 half-life of Cs-137 (≈30 years), and is cycled daily between water storage and dry irradiation, 10,000 cycles are imposed.
4. The effects of thermal cycling on capsule integrity are not known. The unknown effects include:

a. The effects of thermal (expansion and contraction) cycling on corrosion rate at the inner capsule-CsCl interface.

b. The effects of thermal cycling and pool water purity, e.g. chloride content, on the outer capsule.

c. The effects of temperature and/or thermal cycling on capsule physical stability, swelling, etc.

Safety Precautions

The uncertainties that accrue with long-term use of capsules containing ^{137}CsCl may be offset to a degree by constraints on facility design and operation. These constraints must be based on the properties of the material being irradiated as well as the mode of irradiator operation. For,

A. Dry-Irradiation/Wet Storage, constraints may include:
   1. Periodic sampling and destructive testing of capsules.
   2. Specification of limits on storage pool water quality for both radioactive and non-radioactive constituents.
   3. Water impervious storage pool liners.
   4. Shielded cleanup systems for removal of radioactive contaminants in storage pool water.
   5. Approved plans for operation of cleanup systems.
   6. Approved plans for disposal of radioactively-contaminated resins.

B. Dry-Irradiation/Dry-Storage, constraints may include:
   1. Periodic sampling and destructive testing of capsules.
   2. Ventilation leakage control.
   3. Specification of limits on radioactive constituents in ventilation off-gas.
   4. High efficiency filtration system requirements.

5. Approved plans for operation of filtration systems.
6. Approved plans for disposal of radioactively contaiminated filters.

C. Commodities, the constraints would be determined by the properties of the commodity, which may vary widely. For example, irradiation of:

1. Fruits, Nuts, and Field Crops may cause imposition of cleanliness standards to prevent stems, leaves, dust, and dirt from impeding transport through the irradiator or movement of the source racks.
2. Bulk Grains may require that the dust loading of air be controlled to prevent explosion.
3. Crated or Boxed Commodities may require no constreints other than those currently imposed on medical product irradiators.

pped Apples

food is a miracle, but it may not be smart ◆BY DAVID HOLZMAN

pest.

HE IRRADIATION THAT ZAPPED the apples in my pie at the National Press Club last month could have killed me many times over. Yet the pie and the rest of the lunch it came with—appetizers of beef and pork, and chicken kebab in a bed of rice—tasted perfectly normal. The chicken was so tender you could cut it with a fork.

Irradiation of food—exposing raw meat, vegetables and fruit to radioactive zapping by cesium 137 or cobalt 60, two elements that are waste products of the nuclear weapons and energy industries—is said by its promoters to kill hazardous germs and pests, to prevent or at least delay spoilage without refrigeration, and to leave the food tastier than it would be after other preserving processes.

The atmosphere in the conference rooms where lunch was served was like a restaurant: gleaming white tablecloths, clinking tableware and animated chatter by a crowd of 150, including members of the news media, officials of food trade associations, congressional staffers and representatives of some Third World embassies and the Inter-American Development Bank. Only the television crews, the speeches and the displays of elderly irradiated foodstuffs looking pristine next to their decrepit nonirradiated counterparts were reminders that this was no ordinary afternoon of leisurely lunching in pleasant surroundings.

The affair was a determined celebration of a food-processing method that has billed itself as up-to-date and "the last word" for 30 years, but has yet to win legalization or find a significant market. The lunchers were there as guests of Miramar Industries of McLean, a manufacturer of irradiation equipment, which had staged the invitation-only lunch on behalf of Rep. Sid Morrison (R-Wash.) and Sen. Slade Gorton (R-Wash.). On this January day Morrison had for the third time introduced his bill to promote irradiation as a food preservation process.

Gorton explained to guests that irradiation could supplant dangerous pesticides like EDB, which the Environmental Protection Agency banned last September. Irradiation, he said, could kill such insect pests as "the codling moth in apples." Washington state is the nation's leading apple grower and, needless to say, the apples in the pies at lunch had come from there.

Irradiation could also eliminate salmonella in red meat, poultry and fish, Gorton said. High doses of irradiation can actually sterilize meat so that, vacuum-packed, it will keep for years at room temperature. Seven years ago, Sen. Tom Harkin (D-Iowa) told lunch guests, he had been served 10-year-old irradiated bacon by the Department of the Army. "I'm still here," he joked, "and it tasted like it had been produced that day."

If the food at the Miramar lunch tasted as if it had been processed only the day before, however, it may have been because it *had* been processed only the day before, most of it at an irradiation plant in New Jersey. Moreover, the food had received only low doses suitable for killing insects, not zaps strong enough to extend shelf life.

As guests took first tentative bites of zapped strawberries, nuts and dried fruit on the tables, Morrison extolled other virtues of irradiation. No longer, he said, would American housewives need to cook pork until it resembled "an old army boot"—irradiation would eliminate the trichinosis worm.

Morrison made a point of introducing Rep. Melvin Price (D-Ill.), forme chairman of the House Armed Services Committee. Price, said Morrison, was one of irradiation's oldes friends, a man who for years ha "pushed for what we call beneficia use . . . of some of the byproducts o the defense establishment."

The "byproducts" Morrison wa talking about is the radioactive garbage of nuclear weapons manufacturing, specifically cesium 137. Morrison's district includes the Hanfor Federal Nuclear Reservation, wher much of the nation's nuclear weaponry is manufactured and where increasing quantities of radioactiv trash are piling up. "Every congressman who has ever come out of tha district has wanted to do somethin with that nuclear garbage," says Robert Alvarez of the Environmenta Policy Institute.

ALMOST ANYTHING tha would the quell the pork-borne threa of trichinosis, which lodges painfull and incurably in human muscle tissue, would probably seem a great ide to those at the National Pork Producers Council. Dave Meisinger, th Pork Producers' director of researc and education, says trichinosis is rar in this country but when it strike the headlines are grabbers. The nam of pork becomes mud.

In early 1982, just after the Por Producers had set a goal of eradicating the trichina parasite by 1987, Bil McMullen of the Department o Energy approached them, promotin food irradiation. "I had never eve heard of irradiation," Meisinger say "I said, 'Sure, we're interested.'"

Trichinosis breaks out amon swine when infected wild animals in

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David Holzman is a Washington science writer.

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vade the farm, said Dr. George Wilson of the American Meat Institute. Infected slaughterhouse scraps in feed spread the parasite farther. Wilson said a new immunological technique named ELISA (Enzyme-Linked Immuno-Sorbed Assay), which singles out infected carcasses, has a number of advantages over irradiation. Among other things, offending farms can be traced and forced to clean up their operations. Moreover, ELISA is cheaper, an estimated 10 to 15 cents per hog or less, a distinct saving from the 27 to 95 cents per hog that Department of Energy experts project irradiation might cost.

Nevertheless, the Pork Producers endorsed Morrison's food irradiation bill, and continue to testify favorably on it. Why? Irradiation may do more than fight trichinosis, says Ray Hankes, chairman of the Pork Producers' trichina-safe pork task force. "Even at triple the cost, if we can extend shelf life and cut down on refrigeration, there may be some merchandising techniques that we are not even aware of that this may allow us to explore."

Meisinger concedes, though, that irradiation is less promising than ELISA and another trichinosis detection system called the Stomacher method. "Those I feel a lot more strongly about because they have been shown to work economically and effectively," he says.

SOME TECHNOLOGIES catch fire in the private sector and blaze their way into the homes of America. Food irradiation, however, has smoldered for 30 years in various federal agencies with little interest shown by private food companies. The major federal program now resides not in the Department of Agriculture, the usual home of the government's food-related activities, but in the so-called byproducts utilization program of the Department of Energy. How it got there and where it is going is a classic study in bureaucracy.

Food irradiation research began in earnest under Eisenhower's Atoms for Peace Program. The Army of that day hoped to convince the Food and Drug Administration that meat sterilized by radiation was a safe, tasty alternative to C-rations, but much of the research was flawed and in 1968 the FDA rescinded its earlier approval of irradiated bacon.

Meanwhile, a new irradiation program had sprung up in another corner of the bureaucracy. In 1972, Jacek (Jack) Sivinski had finished his work on irradiation for NASA. Later that decade, two spacecraft would land on Mars and analyze the soil for signs of life.

Sivinski's assignment had been to investigate irradiation as a method of sterilizing spacecraft to ensure that Earthling microbes did not contaminate Martian soil samples. As matters turned out, NASA chose another sterilization technique, but Sivinski was loath to disband his team of irradiation biologists. He thought he saw a solution. The Clean Water Act had just been passed, and mountains of municipal sludge would soon be piling up. U.S. policy at the time called for reprocessing the spent fuel rods from nuclear power plants to recapture plutonium for use as reactor fuel. But reprocessing also begins to separate out other radioactive elements, including a lot of highly radioactive cesium 137. Siviniski thought up a use for it: killing the sewage pathogens so that sterile sludge could fertilize the gardens of America.

The Atomic Energy Commission, now the Department of Energy,

Continued on page 22

Irradiation, from page 17

launched the program. But President Carter spoiled Sivinski's plans when he banned reprocessing in order to keep plutonium out of civilian hands. A side effect of the Carter decision was to leave cesium locked in the spent fuel rods.

Some cesium was available, however, from reprocessing of nuclear weapons waste. Sivinski got his operation moved to the weapons end of the DOE; there its major goal became food irradiation. The byproducts utilization program was born.

In 1981, Sivinski left the Department of Energy for a job at CH2M Hill, a major environmental engineering consulting firm (The "CH2M" stands for Cornell, Howland, Hayes and Merrifield, founders of one of the two firms that parented the present corporation). Despite the new job, Sivinski's role changed little. He directs the CH2M's irradiation research, which is all under contract to his old office at DOE, and he continues to trot around the globe preaching irradiation. "The developing countries, those are the ones who need to go into irradiation to balance their payments, for quarantine requirements, and to feed their hungry," he says.

ROBERT MORRIS, a postharvest food technologist at the Agency for International Development, is a pale, intense man who can hardly contain his enthusiasm for food irradiation. He can foresee his agency conducting training programs to teach Third World nations to run irradiators. This is all still a dream, because AID as yet has no policy on food irradiation, but Morris is doing his best to promote it to his supervisors. "I see my role as a lobbyist for the developing countries," he says.

Morris was introduced to food irradiation in April 1982, shortly after Jack Sivinski paid a visit to his boss. His initial skepticism soon disappeared. "My close association with the wisdom of Jack Sivinski helped persuade me," he says. ". . . I have *irradiation is a waste of money. To others, it's only one of several technologies, in most cases not the best of them. And to most experts on Third World hunger, it's just the latest in a series of high-tech solutions that always fail.*

WARNING signs, above, posted near plates of irradiated food warned guests at the Miramar lunch that what they were about to eat, while safe, was commercially illegal.

APPLE-farming Rep. Sid Morrison of Washington, left, sees irradiated apples as good news for the orchards of the United States.

traveled with Jack, and have been in close contact with DOE since April 1982."

Again and again the same names show up in the unfolding irradiation story. Recently AID gave $100,000 to the National Food Processors Association, an industry irradiation backer, to investigate the role food irradiation might play in President Reagan's Caribbean Basin Initiative. Now DOE is lending CH2M Hill's Sivinski to head up the study.

IN THE PAST, despite the best efforts of Morrison, Gorton and their like-minded colleagues, food irradiation never generated much enthusiasm on Capitol Hill. One staffer from a relevant committee says that Morrison is the only congressman who ever called about it. Nor did the FDA show much inclination to legalize irradiation for general use on food.

Then, in 1983, a series of events helped launch a new irradiation drive. First, Margaret Heckler, a longtime booster, became secretary of health and human services, the agency that oversees FDA. (Heckler had for 16 years represented the Massachusetts congressional district that was home to the Army's original irradiation research. "She dragged me to the Natick labs in 1979, looking desperately for a way to keep them opened," recalls Carol Tucker Foreman, who was an assistant agriculture secretary under Carter.)

Then the hazards of EDB, a widely used agricultural pesticide, came to public attention and the Environmental Protection Agency prepared to ban its use. Irradiation's proponents seized the opportunity to promote irradiation as a substitute. In November, Morrison introduced the first version of his bill.

Meanwhile, Heckler began pressing the FDA to legalize irradiation. "Now is the time to move forward with this promising technique," Heckler proclaimed to the National Food Processors Association when a draft of proposed legalizing regulations finally appeared in the Federal Register in February 1984.

A YEAR LATER, though, the FDA still hasn't issued final regulations. And when and if it does, that doesn't mean that Safeway shelves will sag under the weight of all that irradiated food. To some in the food industry, irradiation is a waste of money. To others, it's only one of several competing technologies, in most cases not the best of them. And to most experts on Third World hunger, it's simply the latest in a series of high-tech solutions that always fail because the roots of hunger lie in unequal distribution of wealth, not agricultural shortage.

Regarding salmonella in chicken, Dr. Kenneth May of Holly Farms has this to say: "Why should multimillions of dollars be spent to solve a problem that's easily solvable in anyone's kitchen? Salmonella is destroyed by heating to 140 degrees for five minutes."

For fruit and vegetables, irradiation doesn't always work and has strong competitors, says Agriculture's Dr. Milton Cuyer: dips to kill the pests, worm detection devices, biological pest control and fly-free zones such as parts of the Rio Grande Valley, where fruit flies have been wiped out.

Irradiation does work well on apples. And apples need irradiation, so the story goes, because Washington state faces a glut and needs new markets abroad. Japan, a major potential customer, bars Washington apples because—say the Japanese—they are infested by the codling moth. Morrison, an apple farmer, is personally concerned.

This summer the byproducts utilization program will send a demonstration irradiator to the orchards of Washington. But all for naught, according to officials of several apple trade associations. "In my opinion it's a maneuver by the Japanese government to protect their own apple industry," says Ken Pollard of the Western New York Apple Growers Association, in one typical comment. "After World War II, when MacArthur was there, we shipped an awful lot of apples to Japan and there was never any codling moth problem."

Reducing postharvest losses in the Third World is still a laudable goal, but irradiation is not the way to do it, say experts on agriculture in developing countries. The scale is wrong. To pay for themselves irradiators must process massive quantities of food. But Third World farmers are minuscule producers. "It's bizarre to think of them trundling two tons of grain from their shack to an irradiator and back," says Cliff Lewis of the World Bank.

Keeping bugs and bacteria out of grain is easy, says Dr. Joan Gussow, professor of nutrition at Columbia University, citing Nicaragua's latest grain preservation technology: "Very simple storage bins made of concrete plastered over woven straw that hold the grain off the ground to keep it dry."

UNTIL FOOD irradiation came along, cesium 137 was nothing but a troublesome piece of garbage for the nuclear industry. It is highly radioactive, with a 30-year half-life, and there was nothing to do with it but bury it with the rest of the nuclear waste.

Unfortunately, though, the reprocessing that would extract it from the rest of the garbage is expensive, so expensive that despite President Reagan's invitation to private industry to develop the technology, there have been no takers. Jerry Brubaker of the house subcommittee on energy conservation and power explains why: "If you take the value of all the recovered products, you still can't come close to equaling the cost of reprocessing."

That leaves the nuclear weapons program as the only source of the isotope; currently the byproducts utilization program, which markets it, has none available. Cobalt 60, the alternative irradiation isotope, comes only from 11 Canadian reactors; people like Siviniski worry that if food irradiation begins on a large scale, cobalt will be in similar short supply. Meanwhile, despite the cesium shortage, the price the byproducts utilization program charges for it is only one-tenth what Canadian cobalt sells for; it's a price that comes nowhere near recouping production costs, raising questions as to whether some sort of special promotion is under way. The program's William Remini responded that the price was set years ago. "I don't think it's ever been the policy of the U.S. government to change its price based on supply and demand," he said. ■

ORIGEN

PWR, 3.3% ^{235}U, 36.4 MW, 27,000 MWD, 73 DAYS

At. Wt. 130 → 140 $> 10^{-3}$ GRAM-ATOMS / MT U

		Decay	Half-life
52 Te 130	1.82×10^{-1}	STABLE	
Te 132	3.36×10^{-2}	β^-	78 HR.
53 I 131	5.51×10^{-2}	β^-	8 DAY
I 133	1.38×10^{-2}	β^-	21 HR.
I 135	4.12×10^{-3}	β^-	6.7 HR.
54 Xe 131	2.80×10^{-1}	STABLE	
Xe 132	4.99×10^{-1}	STABLE	
Xe 133	8.04×10^{-2}	β^-	5.3 DAYS
Xe 134	9.14×10^{-1}	STABLE	
Xe 135	1.62×10^{-3}	β^-	9.2 HRS.
Xe 136	1.33×10^{0}	STABLE	
55 Cs 133	7.09×10^{-1}	STABLE	
Cs 134	6.96×10^{-3}	β^-	2.1 YRS
Cs 135	2.13×10^{-1}	β^-	3×10^{6} YRS
Cs 137	7.50×10^{-1}	β^-	30.2 YRS.

$_{52}Te^{132} \xrightarrow[78\text{ HR.}]{\beta^-} {}_{53}I^{132} \xrightarrow[2.3\text{ HR}]{\beta^-} {}_{54}Xe^{132}$ (STABLE)

$_{53}I^{131} \xrightarrow[8\text{ DAY}]{\beta^-} {}_{54}Xe^{131}$ (STABLE)

$_{53}I^{133} \xrightarrow[21\text{ HR.}]{\beta^-} {}_{54}Xe^{133} \xrightarrow[5.3\text{ DAYS}]{\beta^-} {}_{55}Cs^{133}$ (STABLE)

$_{53}I^{135} \xrightarrow[6.7\text{ HR.}]{\beta^-} {}_{54}Xe^{135} \xrightarrow[9.2\text{ HR.}]{\beta^-} {}_{55}Cs^{135} \xrightarrow[3 \times 10^{6}\text{ YRS}]{\beta^-} {}_{56}Ba^{135}$ (STABLE)

$_{54}Xe^{133} \xrightarrow[5.3\text{ DAY}]{\beta^-} {}_{55}Cs^{133}$ (STABLE)

$_{54}Xe^{135} \xrightarrow[9.2\text{ HR.}]{\beta^-} {}_{55}Cs^{135} \xrightarrow[3 \times 10^{6}\text{ YR}]{\beta^-} {}_{56}Ba^{135}$ (STABLE)

$_{55}Cs^{133}$ (STABLE)

$_{55}Cs^{134} \xrightarrow[2.1\text{ YR}]{\beta^-} {}_{56}Ba^{134}$ (STABLE)

$_{55}Cs^{135} \xrightarrow[3 \times 10^{6}\text{ YR.}]{\beta^-} {}_{56}Ba^{135}$ (STABLE)

$_{55}Cs^{137} \xrightarrow[30.2\text{ YRS}]{\beta^-} {}_{56}Ba^{137}$ (STABLE)

A|99

ORNL-4186

Contract No. W-7405-eng-26

ISOTOPES DEVELOPMENT CENTER

CESIUM-137 DATA SHEETS

S. J. Rimshaw

E. E. Ketchen

Isotopes Division

DECEMBER 1967

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

A/100

CONTENTS

$${_{92}U^{235}} + {_0n^1} \longrightarrow {_{54}Xe^{135}} + {_{38}Sr^{99}} + 2\,{_0n^1}$$

$${_{54}Xe^{135}} + {_0n^1} \xrightarrow[(2\times10^6)]{} {_{54}Xe^{136}} + {_0n^1} \xrightarrow[(280b)]{} {_{54}Xe^{137}} \xrightarrow[3.9\,\text{min.}]{\beta^-} {_{55}Cs^{137}} \xrightarrow[30\,\text{YRS.}]{\beta^-} {_{56}Ba^{137}}\ \text{(STABLE)}$$

A SMALL EFFECT DUE TO LOW ABSORPTION CROSS-SECTION OF $_{54}Xe^{136}$

$${_{54}Xe^{135}} \xrightarrow[9.2\,\text{HR.}]{\beta^-} {_{55}Cs^{135}} \xrightarrow[3\times10^6\,\text{YR.}]{\beta^-} {_{56}Ba^{135}}\ \text{(STABLE)}$$

$${_{54}Xe^{133}} \xrightarrow[5.3\,\text{DAY}]{\beta^-} {_{55}Cs^{133}}\ \text{(STABLE)}$$

present and the entire nucleus starts to oscillate. If enough energy is available, the nucleus is deformed sufficiently that the protons concentrate at either end and cause the nucleus to split into two fragments which fly apart at high velocities because of the mutual repulsion of the protons.

The liquid-drop model predicts that fission should be symmetric. Chemical analysis of the fission products shows that this is practically never true and that the fission products actually fall into a light group at a mass number of about 95 and a heavy group at a mass number of about 140. The fission yield of a particular fission product is defined as the percentage of fissions that lead to the formation of that particular product. Except in very rare cases, each fission produces two fission products, so that the total fission yield is normally assumed to be 200 per cent. Since most fission fragments are radioactive, decaying by negative beta emission, the fission yield is given in terms of the mass number of the nucleus which is not affected by beta decay. Figure 2-3 gives the fission product yield for U^{235}, Pu^{239}, and U^{233}. Note that the curves are very similar.

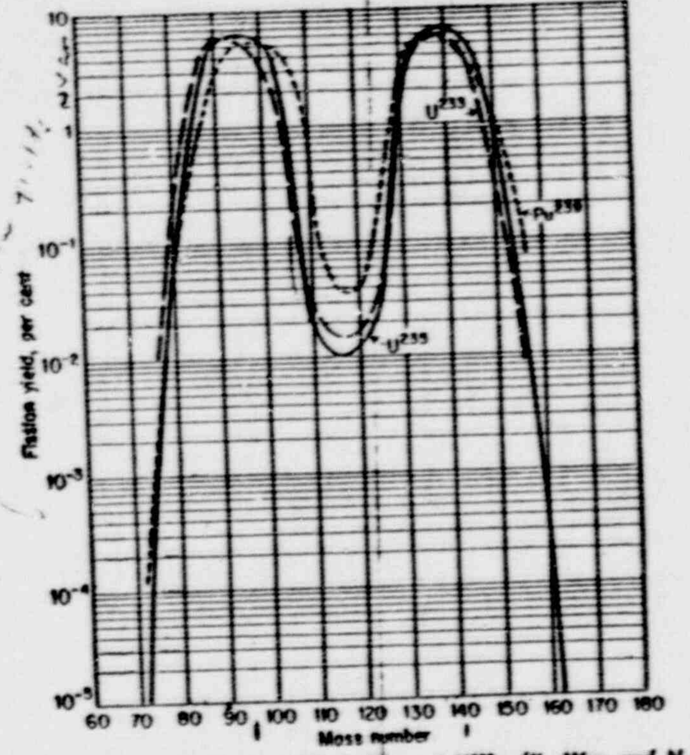


FIG. 2-3. Fission product yields for U^{235}, Pu^{239}, and U^{233}. (K. Way and N. Dismuke, *Fission Product Yields*, ORNL-280 and AECD-2817, 1949.)

The data given in Fig. 2-3 are for fission by thermal neutrons. The distribution of fission products will be affected by the energy of the incident particles, and for fast neutrons the dip in the curve is less pronounced, indicating that symmetric fission is more likely.

Fission Product Decay. An understanding of the instability of fission fragments may be obtained by referring to the neutron-proton numbers for stable nuclides as shown in Fig. 1-3. For the compound nucleus U^{236}, the ratio of neutrons to protons is 1.57. This is considerably higher than the neutron-proton ratios of about 1.3 to 1.4 for stable nuclides of atomic number from 30 to 60. When the uranium nucleus fissions, this neutron excess or proton deficiency is relieved partially by the emission of fission neutrons and partially by radioactive decay of the fission fragments. All radioisotopes decay in such a way as to become more stable, and since beta decay is equivalent to the transformation of a neutron into a proton within the nucleus, substantially all the fission fragments are beta emitters.

Let us consider a typical fission reaction for U^{235}. The most probable fission fragments are strontium and xenon, atomic numbers 38 and 54, respectively, and we shall assume that two neutrons are also emitted on fission, so that a typical fission equation is

$$ {}_{92}U^{235} + {}_{0}n^{1} \rightarrow {}_{38}Sr^{94} + {}_{54}Xe^{140} + 2\,{}_{0}n^{1} \tag{2-13}$$

Both products of this reaction are radioactive since the heaviest stable isotope of strontium is Sr^{88} and of xenon Xe^{136}. The further decay of each fission fragment is termed a fission chain, and usually involves about three successive beta disintegrations. The fission chains for the products from Eq. (2-13) are

$$ {}_{38}Sr^{94} \xrightarrow{\beta^-} {}_{39}Y^{94} \xrightarrow{\beta^-} {}_{40}Zr^{94} \text{ (stable)} \tag{2-14}$$

$$ {}_{54}Xe^{140} \xrightarrow[16\text{ sec.}]{\beta^-} {}_{55}Cs^{140} \xrightarrow[66\text{ sec.}]{\beta^-} {}_{56}Ba^{140} \xrightarrow[12.8\text{ days}]{\beta^-} {}_{57}La^{140} \xrightarrow[40\text{ hr.}]{\beta^-} {}_{58}Ce^{140} \text{ (stable)} \tag{2-15}$$

A large number of fission chains have been identified in the fission products. In addition to being beta emitters, many of the fission products are also gamma emitters, and for this reason suitable radiation shielding must be used when handling fission products. In many cases the fission products are considered to be a nuisance since they act as a poison for the nuclear chain reactor, but the fission products do serve as a valuable source of radioisotopes, particularly those with a high neutron-proton ratio.

Prompt Neutrons. As we have seen, the immediate fission fragments are radioactive since they contain a greater neutron-proton ratio than stable nuclei. Since the energy from fission is about 200 Mev, which considerably exceeds the binding energy of a neutron, it is not surprising that the high neutron excess of the fission fragments is partially relieved by the

CESIUM-137 DATA SHEETS

I. FUEL FORM (as processed)

REFERENCE COLUMN

A. CESIUM CHLORIDE (CsCl) HALF-LIFE: 30.0 y

1

1. Composition

a. Radionuclidic Abundance

The cesium product contains >99% cesium and <1% rubidium.

2, 3

Isotope	% Abundance	HALF LIFE
^{133}Cs	43.4	STABLE
^{135}Cs	20.1	3×10^6 YR.
^{137}Cs	36.5	30.2 YR.

MOST CESIUM COMES FROM FISSION PRODUCT Cs AND THE DECAY OF F.P. XENON.

These values were obtained from a mass spectrometric analysis of fission product cesium.

The above isotopes are produced in about equal quantities during fission, but ^{135}Xe, which is A ~~the~~ precursor of ^{137}Cs, has a cross section of 2.7×10^6 barns and a considerable amount of this isotope burns up during reactor operation.

b. Radiochemical Purity

2, 3

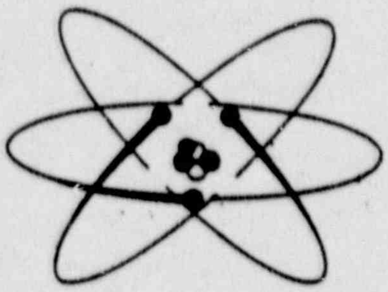
The principal radionuclide other than ^{137}Cs is ^{134}Cs ($T_{1/2} = 2.1$ y), whose content increases with increasing irradiation dose, as shown in the following table.

Calculated Ratios of ^{134}Cs/^{137}Cs Activities as a Function of Irradiation Dose to the Fuel in Megawatt-Days per Ton

Irradiation dose, Mwd/ton	Ratio of ^{134}Cs/^{137}Cs
1,000	0.036
2,000	0.072
4,000	0.145
6,000	0.218
8,000	0.29
10,000	0.36
15,000	0.54
20,000	0.72

RADIOLOGICAL HEALTH HANDBOOK

Compiled and edited
by the
Bureau of Radiological Health
and the
Training Institute
Environmental Control Administration



Revised Edition
January 1970

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service
Consumer Protection and Environmental Health Service
Rockville, Maryland 20852

For sale by the Superintendent of Documents, U.S. Government Printing Office
Washington, D.C. 20402 - Price $4.00

GAMMA RADIATION LEVELS FOR ONE CURIE OF SOME RADIONUCLIDES*

Nuclide	Γ†	Nuclide	Γ†	Nuclide	Γ†
Actinium-227	~2.2	Gold-198	2.3	Potassium-43	5.6
Antimony-122	2.4	Gold-199	~0.9	Radium-226	8.25
Antimony-124	9.8	Hafnium-175	~2.1	Radium-228	~5.1
Antimony-125	~2.7	Hafnium-181	~3.1	Rhenium-186	~0.2
Arsenic-72	10.1	Indium-114m	~0.2	Rubidium-86	0.5
Arsenic-74	4.4	Iodine-124	7.2	Ruthenium-106	1.7
Arsenic-76	2.4	Iodine-125	~0.7	Scandium-46	10.9
Barium-131	~3.0	Iodine-126	2.5	Scandium-47	0.56
Barium-133	~2.4	Iodine-130	12.2	Selenium-75	2.0
Barium-140	12.4	Iodine-131	2.2	Silver-110m	14.3
Beryllium-[illegible]	~0.3	Iodine-132	11.8	Silver-111	~0.2
Bromine-[illegible]	14.6	Iridium-192	4.8	Sodium-22	12.0
Cadmium-115m	~0.2	Iridium-194	1.5	Sodium-24	18.4
Calcium-47	5.7	Iron-59	6.4	Strontium-85	3.0
Carbon-11‡	5.9	Krypton-85	~0.04	Tantalum-182	6.8
Cerium-141	0.35	Lanthanum-140	11.3	Tellurium-121‡	3.3
Cerium-144	~0.4	Lutecium-177	0.09	Tellurium-132	2.2
Cesium-134	8.7	Magnesium-28	15.7	Thulium-170	0.025
Cesium-137	3.3	Manganese-52	18.6	Tin-113	~1.7
Chlorine-38‡	8.8	Manganese-54	4.7	Tungsten-185	~0.5
Chromium-51	0.16	Manganese-56	8.3	Tungsten-187	3.0
Cobalt-56	17.6	Mercury-197	~0.4	Uranium-234	~0.1
Cobalt-57	0.9	Mercury-203	1.3	Vanadium-48	15.6
Cobalt-58	5.5	Molybdenum-99	~1.8	Xenon-133	0.1
Cobalt-60	13.2	Neodymium-147	0.8	Ytterbium-175	0.4
Copper-64	1.2	Nickel-65	~3.1	Yttrium-88	14.1
Europium-152	5.8	Niobium-95	4.2	Yttrium-91	0.01
Europium-154	~6.2	Osmium-191	~0.6	Zinc-65	2.7
Europium-155	~0.3	Palladium-109	0.03	Zirconium-95	4.1
Gallium-67	~1.1	Platinum-197	~0.5		
Gallium-72	11.6	Potassium-42	1.4		

---

* Jaeger, R. G., *et al.*, *Engineering Compendium on Radiation Shielding, Vol. 1*, (New York: Springer-Verlag, 1968), pp. 21-30.

† Γ = R-cm^2/hr-mCi or $\Gamma/10$ = R/hr at 1 m/Ci

‡ *A Manual of Radioactivity Procedures* (National Bureau of Standards Handbook No. 80 [Washington, D.C.: Supt. of Docs., U.S. Government Printing Office, Nov. 1961)], Appendix A, pp. 137-140.

SPECIFIC ACTIVITY

Radionuclide	Half-Life	Curies per gram
Hydrogen-3	12.3y	9.64×10^{3}
Carbon-14	5730y	4.46
Nitrogen-16	7.2s	9.79×10^{10}
Sodium-22	2.60y	6.25×10^{3}
Sodium-24	15.0h	8.71×10^{6}
Phosphorus-32	14.3d	2.85×10^{5}
Sulfur-35	88d	4.24×10^{4}
Chlorine-36	3.1×10^{5}y	3.21×10^{-2}
Argon-41	1.83h	4.18×10^{7}
Potassium-42	12.4h	6.02×10^{6}
Calcium-45	165d	1.76×10^{4}
Chromium-51	27.8d	9.21×10^{4}
Manganese-54	303d	7.98×10^{3}
Iron-55	2.6y	2.50×10^{3}
Manganese-56	2.576h	2.17×10^{7}
Cobalt-57	270d	8.48×10^{3}
Iron-59	45d	4.92×10^{4}
Nickel-59	8×10^{4}y	7.58×10^{-2}
Cobalt-60	5.26y	1.13×10^{3}
Nickel-63	92y	61.7
Copper-64	12.8h	3.83×10^{6}
Zinc-65	245d	8.20×10^{3}
Gallium-72	14.1h	3.09×10^{6}
Arsenic-76	26.5h	1.56×10^{6}
Bromine-82	35.34h	1.08×10^{6}
Rubidium-86	18.66d	8.14×10^{4}
Strontium-89	52d	2.82×10^{4}
Strontium-90	28.1y	141
Yttrium-90	64h	5.44×10^{5}
Yttrium-91	58.8d	2.44×10^{4}
Molybdenum-99	67h	4.72×10^{5}
Technetium-99m	6.0h	5.28×10^{6}
Ruthenium-106	367d	3.36×10^{3}
Iodine-125	60d	1.74×10^{4}
Iodine-130	12.4h	1.94×10^{6}
Iodine-131	8.05d	1.24×10^{5}
Barium-133	7.2y	374
Cesium-134	2.05y	1.30×10^{3}
Cesium-137	30.0y	87.0
Barium-140	12.8d	7.29×10^{4}
Lanthanum-140	40.22h	5.57×10^{5}
Cerium-141	33d	2.81×10^{4}
Cerium-144	284d	3.19×10^{3}
Praseodymium-144	17.3m	7.55×10^{7}
Promethium-147	2.62y	929
Tantalum-182	115d	6.24×10^{3}
Tungsten-185	75d	9.41×10^{3}
Iridium-192	74.2d	9.17×10^{3}
Gold-198	64.8h	2.44×10^{5}
Gold-199	75.6h	2.08×10^{5}
Mercury-203	46.9d	1.37×10^{4}
Thallium-204	3.8y	462
Polonium-210	138.4d	4.49×10^{3}
Polonium-212	304ns	1.75×10^{17}
Radium-226	1602y	0.988
Thorium-232	1.41×10^{10}y	1.09×10^{-7}
Uranium-233	1.62×10^{5}y	9.48×10^{-3}
Thorium-234	24.1d	2.32×10^{4}
Uranium-235	7.1×10^{8}y	2.14×10^{-6}
Uranium-238	4.51×10^{9}y	3.33×10^{-7}
Plutonium-239	2.44×10^{4}y	6.13×10^{-2}

TABLE OF THE ISOTOPES (Continued)

z	Isotope	% nat. abundance	Atomic mass	Lifetime	Modes of decay	Decay energy (MeV)	Particle energies (MeV)	Particle intensities	Gamma energies (KeV)	Gamma intensities	Thermal neutron capture cross-section	I	μ	Q
	$_{55}Cs^{134}$			2.05y	β^-	2.062	0.089	28%	475.34 ± 0.1	15%	134 ± 12b	4	+2.990	+0.43
							0.410	1%	563.20	8%				
							0.662	71%	569.33	14%				
									604.7	98%				
									795.8	88%				
									801.9	9%				
									1038.6	1.1%				
									1167.9	1.9%				
									1365.1	3.4%				
									1400.5	0.08%				
	$_{55}Cs^{135}$			3 × 10⁶y	β^-	0.210	0.210	100%			8.7 ± 0.5b	7/2	+2.729	+0.049
	$_{55}Cs^{136}$			13d	β^-	2.54	0.341	93%	66.9 ± 0.2	13.7%		5	±3.70	
							0.56	2%	86.4	5.6%				
							0.657	7%	153.30 ± 0.2	8.2%				
									164.0 ± 0.2	4.5%				
									176.7 ± 0.2	13%				
									273.8 ± 0.2	12%				
									340.6 ± 0.15	44%				
									818.5 ± 0.2	100%				
									1048.1 ± 0.2	80%				
									1235.4 ± 0.2	20%				
	$_{55}Cs^{137}$			30.23 ±0.16y	β^-	1.176	0.511	94%	661.630 ± 0.03	84.8 ± 0.5%	110 ± 33 mb	7/2	+2.838	+0.050
							1.176	6%						
	$_{55}Cs^{138}$			32.2m	β^-	4.83	1.49	0.5%	66.7 ± 0.3			3	±0.5	
							2.20	10%	75.41 ± 0.4	0.081				
							2.39	36%	107.6 ± 0.3	0.06				
							2.53	5%	112.27 ± 0.25	0.16				
							2.62	16%	137.89 ± 0.15	1.58				
							2.94	12%	165.75 ± 0.2	3.17				
							3.4	21%	191.83 ± 0.3	0.5				
									212.04 ± 0.3					
									227.60 ± 0.18	1.56				
									324.53 ± 0.2	0.43				
									364.1 ± 0.3					
									408.67 ± 0.2	5.3				
									421.39 ± 0.3	0.43				
									462.51 ± 0.18	35.49				
									516.53 ± 0.2	0.65				
									546.71 ± 0.15	12.69				
									871.61 ± 0.18	6.07				
									1009.65 ± 0.18	37.86				
									1146.88 ± 0.2	1.49				
									1203.4 ± 0.8					
									1343.33 ± 0.3	1.64				
									1435.74 ± 0.10	73% 100				
									[illegible] ± 0.20	1.33				

TABLE OF THE ISOTOPES (Continued)

z	Isotope	% nat. abundance	Atomic mass	Lifetime	Modes of decay	Decay energy (MeV)	Particle energies (MeV)	Particle intensities	Gamma energies (KeV)	Gamma intensities	Thermal neutron capture cross-section	I	μ	Q
									80.997 ± 0.006 160.66 ± 0.06 223.43 ± 0.26 276.43 ± 0.26 303.09 ± 0.21 356.26 ± 0.15 384.09 ± 0.20	55.3 ± 4.2 1.01 ± 0.05 0.75 ± 0.04 11.6 ± 0.17 29.9 ± 0.29 ≡100 14.9 ± 0.26				
	$_{56}$Ba^{134}	2.42%	133.9043								0.16 ± 0.02b (Ba^{135m}) 2 ± 2b (Ba^{135})	0		
	$_{56}$Ba^{135m}			28.7h	IT	0.268			268	100%				
	$_{56}$Ba^{135}	6.59%	134.9056								5.8 ± 0.9b	3/2	+0.8365	+0.18
	$_{56}$Ba^{136m}			0.32s	IT	2.04			163.7 818 1050					
	$_{56}$Ba^{136}	7.81%	135.9044								10 ± 1mb (Ba^{137m}) 0.4 ± 0.4b (Ba^{137})			
	$_{56}$Ba^{137m}			2.55m	IT	0.6616			661.630 ± 0.030					
	$_{56}$Ba^{137}	11.32%									5.1 ± 0.4b	3/2	+0.9357	+0.28
	$_{56}$Ba^{138}	71.66%	137.9050								0.35 ± 0.15b	0		
	$_{56}$Ba^{139}			82.9m	β^-	2.34	0.95 2.23 2.38	<1% 27% 72%	165.85 ± 0.010 1270 1430	27%	4 ± 1b			
	$_{56}$Ba^{140}			12.8d	β^-	1.05	0.47 0.58 0.89 1.01 1.02	34% 11% ~3% 33% 19%	13.85 29.97 132.7 162.7 304.9 423.8 437.6 537.3	30% 1.7% 8% 7% 3% 5% 29%	1.6 ± 0.3b			
	$_{56}$Ba^{141}			18m	β^-	3.0	2.0 2.4 2.6 2.8 3.0		113.08 ± 0.05 180.70 ± 0.09 190.33 ± 0.05 276.94 ± 0.06 304.18 ± 0.04	21 0.7 100 52 57				

TABLE OF THE ISOTOPES (Continued)

z	Isotope	% nat. abundance	Atomic mass	Lifetime	Modes of decay	Decay energy (MeV)	Particle energies (MeV)	Particle intensities	Gamma energies (KeV)	Gamma intensities	Thermal neutron capture cross-section	I	μ	Q
									343.66 ± 0.04 389.66 ± 0.07	31 3.1				

CESIUM CHLORIDE (CsCl)

REFERENCE COLUMN

The ratio of ^{134}Cs to ^{137}Cs activities will decrease by a factor of ~2.0 every two years after discharge of the fuel from the reactor.

2. Specific Power (9 YEARS AFTER ENCAPSULATION) 2, 3, 4

a. 0.120 watt/g of pure CsCl
0.119 watt/g of 99% CsCl-1% RbCl SEE CALC. ON REVERSE

b. 25.1 curies of ^{137}Cs per gram of pure CsCl SEE CALC. ON FACING PAGE.
24.85 curies of ^{137}Cs per gram of 99% CsCl-1% RbCl

3. Radiation

a. Alpha particles - None

b. Beta particles 5

Nuclide	Max E, Mev	Avg E, Mev	Abundance, %	w/kilocurie	Particles w^{-1} sec^{-1}
^{137}Cs	1.18	0.42	8	1.23	0.619×10^{12}
	0.52	0.19	92		7.11×10^{12}

c. Gamma 5

Nuclide	Energy, Mev	Abundance, %	w/kilocurie	Particles w^{-1} sec^{-1}
^{137m}Ba	0.662	83.5	3.61	6.46×10^{-12}
Total gamma power			3.61	
Total beta power			1.23	
Total gamma and beta power			4.84	

d. Bremsstrahlung 5

Bremsstrahlung radiation from the 1.18-Mev beta particles is negligible compared to the shielding required for the 0.662-Mev gamma photons.

The bremsstrahlung radiation from the 0.52-Mev beta particles will be similar to that from the 0.545-Mev beta particles of ^{90}Sr calculated in Ref. 5. These values are given in Appendix 1, Table 1.

The bremsstrahlung radiation from the 1.18-Mev beta particles of ^{137}Cs will be similar to that from the 0.968-Mev beta particles of ^{170}Tm calculated in Ref. 5.

$$1\text{ Mev} = 4.44 \times 10^{-20}\text{ KW-hr.}$$

$$\frac{25.0\ \cancel{\text{Ci}}\ \text{Cs}^{137}}{\text{g CsCl}} \times \frac{3.7 \times 10^{10}\ \text{Dis}}{\cancel{\text{sec}}.\ \cancel{\text{Ci}}} \times \left[(0.42)(0.08) + (0.19)(0.92)\right] \frac{\cancel{\text{Mev}}}{\cancel{\text{Dis}}} \times 4.44 \times 10^{-20}\ \frac{\cancel{\text{KW}}\text{-}\cancel{\text{hr}}}{\cancel{\text{Mev}}} \times$$

$$1000\ \frac{\text{W}}{\cancel{\text{KW}}} \times \frac{3600\ \cancel{\text{sec}}}{\cancel{\text{hr}}} = 2.5 \times 3.7 \times 2.08 \times 4.44 \times 3.6 \times 10^{-4} = 0.0308\ \frac{\text{WATTS}}{\text{G. CsCl}}\ \beta$$

$$(2.5)(3.7)(0.662)(\overset{9}{\cancel{0.855}})(4.44)(3.6) \times 10^{-3} = 0.0879\ \frac{\text{WATTS}}{\text{G CsCl}}\ \gamma$$

TOTAL POWER $0.0308 + 0.0979 = 0.129$ WATTS / G. CsCl

ACTIVITY-MASS RELATIONSHIP - SPECIFIC ACTIVITY

The specific activity (SpA) of a radioactive nuclide (disintegrations per unit time)/(unit mass), is calculated from the basic equation:

$$SpA = \lambda N = \frac{(\ln 2)\ N}{T_{\frac{1}{2}}}.$$

Where: N = number of radioactive atoms per unit mass, and

$T_{\frac{1}{2}}$ = half-life.

This basic equation can be transformed as follows:

by definition: $N = 6.0225 \times 10^{23}$/atomic mass

$Ci = 3.7 \times 10^{10}$ DISINTEGRATIONS/SECOND

$$\text{Substituting}: \quad SpA = \frac{0.69315\ N}{T_{\frac{1}{2}}\ (\text{secs})} = \frac{0.69315}{T_{\frac{1}{2}}} \times \frac{6.0225 \times 10^{23}}{\text{atomic mass}} \times \frac{1}{3.7 \times 10^{10}} = \text{Ci/gm.}$$

This equation is satisfactory when the half-life of the nuclide is expressed in seconds. If, however, the half-life is expressed in other units (such as minutes, hours, days, or years), a separate time conversion is required for each. By substituting the appropriate time conversion factors the following five equations can be obtained.

$$\text{curies/gram or SpA } (T_{\frac{1}{2}} \text{ in secs}) = \frac{1.128 \times 10^{13}}{(T_{\frac{1}{2}})(\text{atomic mass})} \quad (1)$$

$$\text{curies/gram or SpA } (T_{\frac{1}{2}} \text{ in mins}) = \frac{1.880 \times 10^{11}}{(T_{\frac{1}{2}})(\text{atomic mass})} \quad (2)$$

$$\text{curies/gram or SpA } (T_{\frac{1}{2}} \text{ in hrs}) = \frac{3.134 \times 10^{9}}{(T_{\frac{1}{2}})(\text{atomic mass})} \quad (3)$$

$$\text{curies/gram or SpA } (T_{\frac{1}{2}} \text{ in days}) = \frac{1.306 \times 10^{8}}{(T_{\frac{1}{2}})(\text{atomic mass})} \quad (4)$$

$$\text{curies/gram or SpA } (T_{\frac{1}{2}} \text{ in yrs}) = \frac{3.578 \times 10^{5}}{(T_{\frac{1}{2}})(\text{atomic mass})} \quad (5)$$

Example: Calculate the specific activity of ^{131}I whose half-life is 8.05d. Using equation (4) and the mass number as the atomic mass, make the appropriate substitutions:

$$SpA = \frac{1.306 \times 10^{8}}{8.05 \times 131} = 1.24 \times 10^{5}$$

The following specific activities were calculated from the above equations, using half-lives from The Table of Isotopes.[1] Integer mass numbers were used rather than actual masses, except for ^{3}H where the exact mass was used. (It should be noted that these specific activities are for pure forms of the nuclides only.) More extensive tables of specific activities are available.[2]

---

[1] Lederer, C. M., Hollander, J. M., and Perlman, I., The Table of Isotopes, (6th ed.; New York: John Wiley & Sons, Inc., 1967).

[2] Goldstein, G., and Reynolds, S. A., "Specific Activities and Half-Lives of Common Radionuclides," Nuclear Data A, Vol. 1, No. 5 (July 1966), pp.435-452.

CESIUM CHLORIDE (CsCl)

REFERENCE COLUMN

These values are given in Appendix 1, Table 2. More accurate values of the bremsstrahlung spectrum can be calculated from the computer program given in Ref. 5.

e. Neutron – none

4. Critical Mass

Cesium-137 and ^{137}Ba are not fissionable.

5. Compatibility With Materials of Containment

A CsCl source which was encapsulated in stainless steel and contained 1540 curies of ^{137}Cs showed no signs of reaction after being opened nine years later. The source operated at slightly above ambient room temperature. 6

6. Thermophysical Properties

a. Density

Density of CsCl versus temperature 7

Temperature, °C	Density, g/cm^3 cubic	fcc	liquid
25	3.999		
100	3.952		
200	3.897		
300	3.827		
400	3.758		
469	3.709	3.153	
500		3.141	
600		3.095	
645		3.072	2.792

Density of molten CsCl in g/cm^3

$\rho = 3.4782 - (1.0650 \times 10^{-3}\ t)$ 8

(temperature range 670-905°C)

b. Coefficients of thermal expansion

$4.78 \times 10^{-5}/°C$ 9

(temperature range 50-150°C)

CESIUM CHLORIDE (CsCl)

REFERENCE COLUMN

c. Specific heat and enthalpy

(1) Specific heat in cal g^{-1} °C^{-1} 10

CsCl (solid, alpha)

$7.59 \times 10^{-2} + (7.29 \times 10^{-6}\ T) - (2.71 \times 10^{2}\ T^{-2})$

(temperature range 385.2-740.5°K)

CsCl (solid, beta)

$4.78 \times 10^{-3} + (1.05 \times 10^{-4}\ T) - (5.29 \times 10^{2}\ T^{-2})$

(temperature range 753.7-904.9°K)

CsCl (liquid)

$8.23 \times 10^{-2} + (2.54 \times 10^{-5}\ T)$

(temperature range 923.6-1168.0°K)

(2) Enthalpy in calories/mole 10

CsCl (solid, alpha)

$H_T - H_{273.15} = -3705 + 12.78\ T + (6.14 \times 10^{-4}\ T^{2}) + (4.56 \times 10^{4}\ T^{-1})$

(temperature range 385.2-740.5°K, ±0.5%)

CsCl (solid, beta)

$H_T - H_{273.15} = 1365 + 0.805\ T + (8.82 \times 10^{-3}\ T^{2}) + (8.91 \times 10^{4}\ T^{-1})$

(temperature range 753.7-904.9°K, ±0.5%)

CsCl (liquid)

$H_T - H_{273.15} = -0.69 + 13.86\ T + (2.14 \times 10^{-3}\ T^{2})$

(temperature range 923.6-1168.0°K, ±0.5%)

d. Temperature of phase transformations

(1) Melting point - 645°C 11

(2) Boiling point - 1300°C 11

(3) Melting temperatures of CsCl-$BaCl_2$ mixtures 12

Mole % CsCl	Temperature, °C
50	725
60	630
70	614
80	590
90	599
100	657
86.5	568 eutectic ?

CESIUM CHLORIDE (CsCl)

REFERENCE COLUMN

e. Latent heats of phase transformations

ΔH transition (469°C) 1.8 kcal/mole, cubic to fcc 7

ΔH fusion 4.9 ± 0.1 kcal/mole 13

ΔH vaporiation 38.2 ± 2.0 kcal/mole 13

f. Vapor pressure

$$\log P_{mm} = 11.346 - \frac{11.076 \times 10^3}{T} + \frac{5}{2} \log \frac{1000}{T}$$ 14

(T is in °K)

g. Thermal conductivity

$cal\ cm^{-1}\ sec^{-1}\ °C^{-1}$	Temperature, °C	15
0.001456	46	
0.001365	89	

Determined on 87.9% theoretically dense pellets of CsCl.

h. Thermal diffusivity

cm^2/sec	Temperature, °C
0.0050	46
0.0047	89

Calculated by dividing the thermal conductivity by the product of the specific heat and density.

i. Viscosity

η, centipoise	Temperature, °C	16
1.45	667	
1.06	727	
0.84	777	
0.68	827	
0.56	877	
0.52	897	

j. Surface tension

dyn/cm	Temperature, °C	17
89.2	664	
81.9	771	
73.7	881	
66.4	979	
61.6	1035	
56.3	1080	

*

CESIUM CHLORIDE (CsCl)

REFERENCE COLUMN

k. Total hemispherical emittance

l. Spectral emissivity
A value of 0.9 can be assumed.

m. Crystallography
cubic, body centered
a = 4.113 kX — 18

n. Solubilities — 18

g per 100 g water	Temperature, °C
162.3	0.7
197.2	29.9
229.4	60.2
259.6	89.5
290.0	119.4

o. Diffusion rates

7. Mechanical Properties

a. Hardness

b. Crush strength

8. Chemical Properties

a. Heat and free energy of formation, entropy

(1) Heat of formation
$\Delta H^\circ_f = -106.3$ kcal/mole — 11

(2) Free energy of formation
$\Delta F^\circ_f = -99.6$ kcal/mole — 11

(3) Entropy
$S^\circ_{298} = 23.9$ eu — 11

b. Chemical reactions and reaction rates (oxygen, nitrogen, water, steam, hydrogen, liquid metals, other) — 18

(1) Oxygen - no reaction

(2) Nitrogen - no reaction

(3) Water - soluble

(4) Inorganic acids - soluble

*

REFERENCE COLUMN

9. Biological Tolerances

19

Maximum permissible body burdens and maximum permissible concentrations of ^{137}Cs in air and in water were taken from Ref. 19 and are given in the table on the following page.

10. Shielding Data

Gamma dose rates from ^{137}Cs power sources of 100, 200, 500, 1000, 2000, 5000, 10,000, and 20,000 watts with iron, lead, and uranium shielding are given in Figs. 1-8. Extra shielding is required when ^{134}Cs impurity is present in the ^{137}Cs source because of the higher energy gamma photons of ^{134}Cs compared to ^{137}Cs.

Maximum Permissible Body Burdens and Maximum Permissible Concentrations for Radionuclides in Air and in Water for Occupational Exposure[19]

Radionuclide and type of decay		Organ of reference (critical organ underscored)	Max. permissible burden in total body, q(μc)	Maximum permissible concentrations, μc/cm^3			
				For 40-hr week		For 168-hr week	
				Water	Air	Water	Air
$_{55}Cs^{137}$ (β^-, γ, e^-)	(Sol)	Total Body	30	4×10^{-4}	6×10^{-8}	2×10^{-4}	2×10^{-8}
		Liver	40	5×10^{-4}	8×10^{-8}	2×10^{-4}	3×10^{-8}
		Spleen	50	6×10^{-4}	9×10^{-8}	2×10^{-4}	3×10^{-8}
		Muscle	50	7×10^{-4}	10^{-7}	2×10^{-4}	4×10^{-8}
		Bone	100	10^{-3}	2×10^{-7}	5×10^{-4}	7×10^{-8}
		Kidney	100	10^{-3}	2×10^{-7}	5×10^{-4}	8×10^{-8}
		Lung	300	5×10^{-3}	6×10^{-7}	2×10^{-3}	2×10^{-7}
		GI (SI)*		0.02	5×10^{-6}	8×10^{-3}	2×10^{-6}
	(Insol)	Lung			10^{-8}		5×10^{-9}
		GI (LLI)*		10^{-3}	2×10^{-7}	4×10^{-4}	8×10^{-8}

---

*The abbreviations GI, SI, and LLI refer to gastrointestinal tract, small intestine, and lower large intestine, respectively.

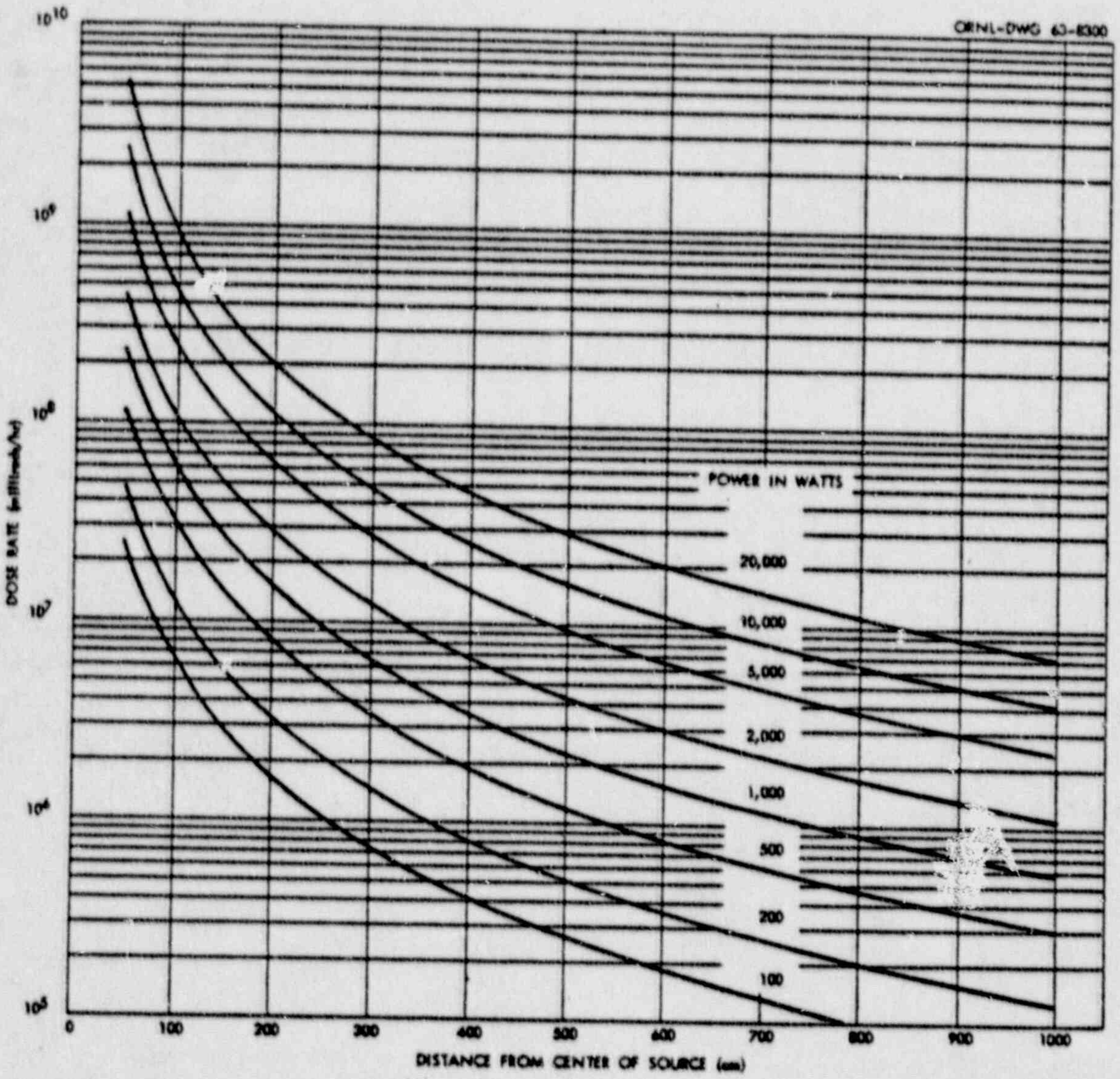


Fig. 1. Gamma Dose Rates from Unshielded Isotopic Power Sources of Cesium-137 as a Function of Distance from Center of Source.

ORNL DWG 63-8468

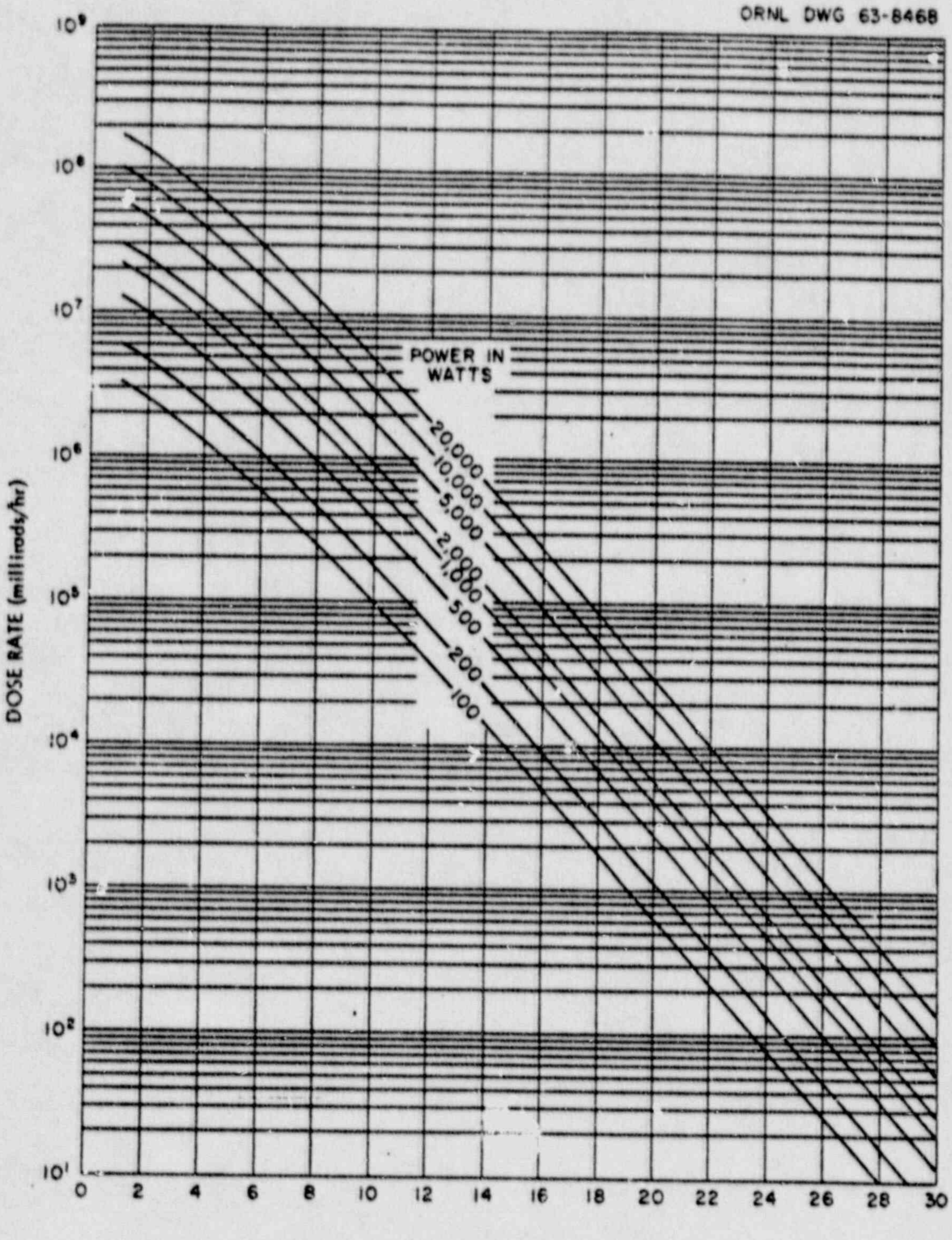


Fig. 2. Gamma Dose Rates from Iron-Shielded Isotopic Power Sources of Cesium-137. Center of source to dose point separation distance = 100 cm.

ORNL DWG 63-8469

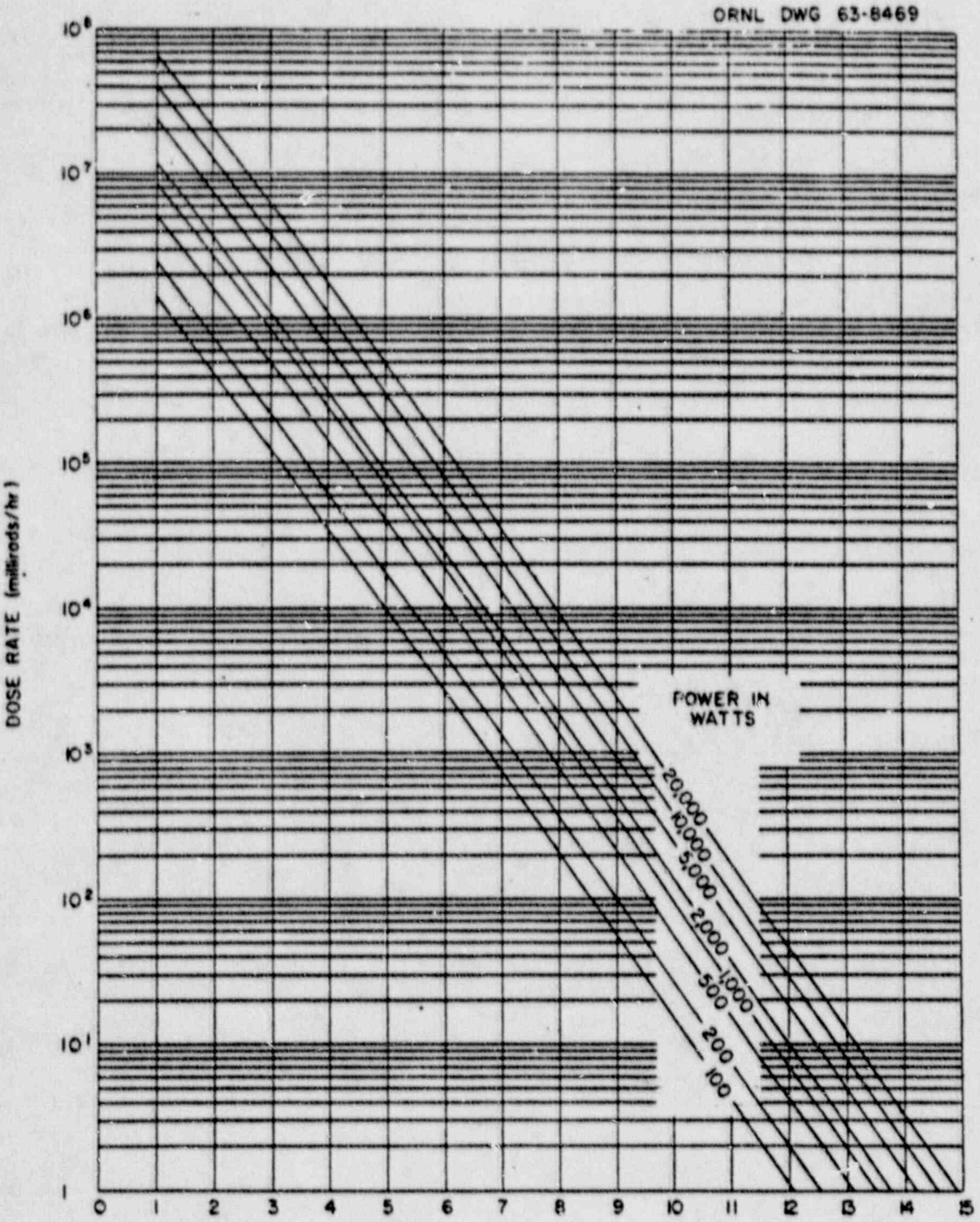


Fig. 3. Gamma Dose Rates from Lead-Shielded Isotopic Power Sources of Cesium-137. Center of source to dose point separation distance = 100 cm.

*

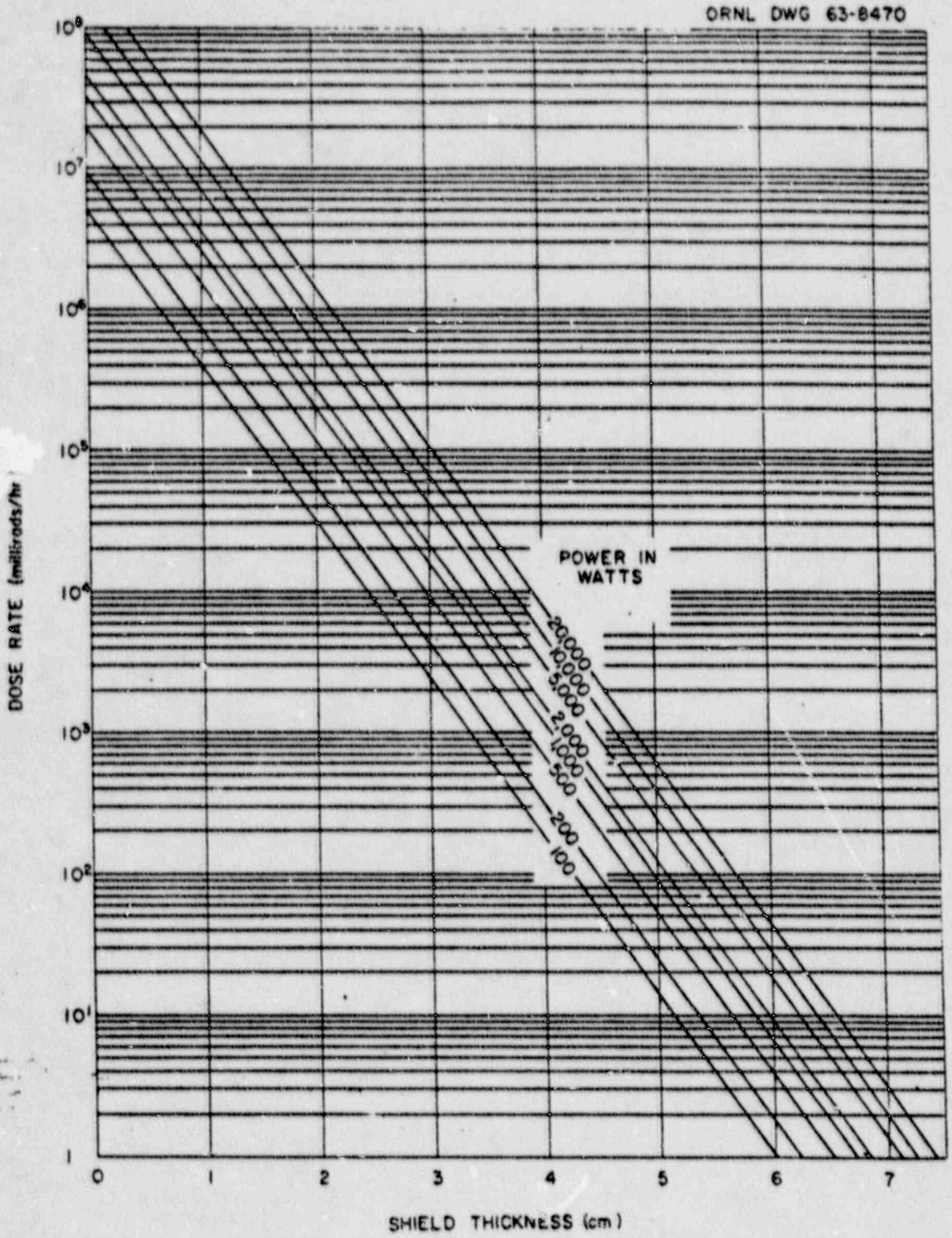


Fig. 4. Gamma Dose Rates from Uranium-Shielded Isotopic Power Sources of Cesium-137. Center of source to dose point separation distance = 100 cm.

REFERENCE COLUMN

C. CESIUM BOROSILICATE GLASS ($Cs_2O \cdot B_2O_3 \cdot 4.2SiO_2$)

1. Composition

a. Radionuclidic Abundance

The cesium product contains >99% cesium and <1% rubidium. The radionuclidic abundances are given in Section I.A.1.

b. Radiochemical Purity

The principal radiochemical impurity in the ^{137}Cs fission product is ^{134}Cs ($T_{1/2} = 2.1$ y), whose content in aged wastes is 2-5% of the ^{137}Cs activity. The ratio of ^{134}Cs to ^{137}Cs activities will decrease by a factor of ~2.0 every two years as a result of decay.

2. Specific Power

a. 0.065 watt/g of pure cesium glass — 4
0.064 watt/g of 99% cesium glass-1% rubidium glass

b. 13.6 curies of ^{137}Cs per gram of cesium glass — 2, 3, 4
13.5 curies of ^{137}Cs per gram of 99% cesium glass-1% rubidium glass

3. Radiation

The radiation is given under Section I.A.3.

4. Critical Mass

Cesium-137 and ^{137}Ba are not fissionable.

5. Compatibility With Materials of Containment

6. Thermophysical Properties

a. Density

3.1 grams/cm^3 – density of pure cesium glass — 23

3.1 grams/cm^3 – density of 99% cesium glass-1% rubidium glass

b. Coefficient of thermal expansion

7.0×10^{-6}/°C for 1.24 Rb_2O-1.08 B_2O_3-7.64 SiO_2 glass — 24

CESIUM BOROSILICATE GLASS ($Cs_2O \cdot B_2O_3 \cdot 4.2SiO_2$)

REFERENCE COLUMN

c. Specific heat and enthalpy

(1) Specific heat in cal g^{-1} °C^{-1}

0.148 at 298°C — 25

(2) Enthalpy in calories/mole

d. Temperature of phase transformations

(1) Softening point – 1200-1300°C — 23

(2) Boiling point

e. Latent heats of phase transformations

f. Vapor pressure

At 1200°C cesium glass will volatilize at the rate of 35 mg of $Cs_2O \cdot B_2O_3$ per cm^2/hr. — 23

g. Thermal conductivity — 26

cal cm^{-1} sec^{-1} °C^{-1}	Temperature, °C
0.00367	125
0.00424	209
0.00600	352
0.00792	448

These values are for Pyrex glass.

h. Thermal diffusivity

0.0117 cm^2/sec at 298°C

This value was calculated by dividing the the thermal conductivity by the product of specific heat and density.

i. Viscosity

j. Surface tension

k. Total hemispherical emittance

l. Spectral emissivity

m. Crystallography

Glass shows no long-range X-ray structure.

n. Solubilities

The leach rate of cesium glass is 0.2 mg cm^{-2} day^{-1} of $Cs_2O \cdot B_2O_3$. — 23

REFERENCE COLUMN

c. Diffusion rates

7. Mechanical Properties

a. Hardness

5-7 on Mohs scale — 27

b. Crush strength

3500-7000 kg/cm^2 at room temperature — 27 (silicates)

8. Chemical Properties

a. Heat and free energy of formation, entropy

(1) Heat of formation of $Cs_2O \cdot B_2O_3 \cdot 4.2SiO_2$

$\Delta H^\circ_f = -1317$ kcal/mole — 22, 25

(estimated from the ΔH°_f's of the individual oxides and the Na-B compounds)

(2) Free energy of formation

$\Delta F^\circ_f = -1237$ kcal/mole — 22, 25

(calculated by $\Delta F^\circ_f = \Delta H^\circ_f - T\Delta S^\circ_f$)

(3) Entropy

$S^\circ_{298} = 95.4$ eu — 22

(estimated by summing the individual oxides)

b. Chemical reactions and reaction rates (oxygen, nitrogen, water, steam, hydrogen, liquid metals, other) — 23

(1) Oxygen – no reaction

(2) Nitrogen – no reaction

(3) Water – 0.2 mg cm^{-2} day^{-1} of cesium

(4) Inorganic acids – soluble

9. Biological Tolerances

Maximum permissible body burdens and maximum permissible concentration of ^{137}Cs in air and water are given under the CsCl Source Form, Section I.A.9. — 19

10. Shielding Data

The shielding requirements are presented under the CsCl Source Form, Section I.A.10.

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Handbook

OF

Chemistry and Physics

CESIUM (L. *caesius*, sky blue), Cs; at. wt. 132.905; at. no. 55; m.p. 28.5°C; b.p. 690°C; sp. gr. 1.873 (20°C) valence 1. Cesium was discovered spectroscopically in 1860 in mineral water from Dürkheim, by Bunsen and Kirchhoff. Cesium, an alkali metal, occurs in *lepidolite*, *pollucite* (a hydrated silicate of aluminum and cesium) and other sources. It is isolated by electrolysis of the fused cyanide. The metal is characterized by a spectrum containing two bright lines in the blue along with several others in the red, yellow and green. It is silvery-white, soft, and ductile. It is the most electropositive and most alkaline element. Cesium, gallium and mercury are the only three metals that are liquid at room temperature. Cesium reacts explosively with cold water, and reacts with ice at temperatures above −116°C. Cesium hydroxide, the strongest base known, attacks glass. Because of its great affinity for oxygen the metal is used as a "getter" in radio tubes. It is also used in photo-electric cells, as well as a catalyst in the hydrogenation of certain organic compounds. The metal has recently found application in ion propulsion systems. Although these are not useable in the earth's atmosphere, 1 lb. of cesium in outer space theoretically will propel a vehicle 140 times as far as the burning of the same amount of any known liquid or solid. Its chief compounds are the chloride and the nitrate. The present price of cesium is about 100 dollars to 150 dollars/lb., depending on quantity and purity.

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Formerly Professor of Chemistry at Case Institute of Technology

In collaboration with a large number of professional chemists and physicists whose assistance is acknowledged in the list of general collaborators and in connection with the particular tables or sections involved.

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PHYSICAL CONSTANTS OF INORGANIC COMPOUNDS (Continued)

No.	Name	Synonyms and Formulae	Mol. wt.	Crystalline form, properties and index of refraction	Density or spec. gravity	Melting point, °C	Boiling point, °C	Solubility, in grams per 100 cc		
								Cold water	Hot water	Other solvents
c282	Cesium	Cs	132.905	silv met or hex	1.873^{20}	28.5	690	d		s liq NH_3
c283	acetate	$CsC_2H_3O_2$	191.95	deliq		194		$945.1^{-2.5}$	$1345.5^{88.5}$	
c284	aluminum sulfate	$CsAl(SO_4)_2.12H_2O$	568.19	col, cub, 1.4587	1.97	117		0.34^0	42.50^{100}	s dil al
	amide	$CsNH_2$	148.93	wh need	3.44	262 ± 1		d		s liq NH_3
	azide	CsN_3	174.93	col need, deliq		310		224.2^0		1.037^{16} al; i eth
c287	benzoate	$CsC_7H_5O_2$	254.03					204.5	305.5	
c288	borofluoride	$CsBF_4$	219.71	rhomb, 1.350	3.20	550 d		1.6^{17}	ca 30^{100}	s dil NH_3
c289	borohydride	$CsBH_4$	147.75	wh, cub, 1.498	2.404			v s		sl s al; i eth, bz
c290	bromate	$CsBrO_3$	260.81	hex, ca 2.15	4.109	ca 420 d		3.66	5.3	
c291	bromide, mono-	CsBr	212.81	col, cub, 1.6984	4.44, liq 3.04	636	1300	124.3^{25}	v s	s a
c292	bromide, tri-	$CsBr_3$	372.63	rhomb		180				
c293	dibromochloride	$CsBr_2Cl$	328.18	yel-red, rhomb		191	150, − Br_2	s		d al, acet
c294	bromochloride iodide	CsIBrCl	375.17	yel-red, rhomb		235	d 290	s		s al
c295	bromoiodide di-	$CsIBr_2$	419.63	rhomb	4.25	248	d 320	4.61		s al
c296	carbonate	Cs_2CO_3	325.82	col cr, deliq		d 610		260.5^{15}	v s	11^{19} al; s eth
c297	carbonate, hydrogen	$CsHCO_3$	193.92	rhomb		175 − ½H_2O		209.3^{15}	v s	s al
c298	chlorate	$CsClO_3$	216.36	sm cr	3.57			6.28	76.5	s al
c299	perchlorate	$CsClO_4$	232.36	rhomb, at 219 cub, 1.4752, 1.4788, 1.4804	3.327^4	d 250		2.00	28.57	0.093 al; 0.7878 al; 0.150 acet
c300	chlorobromide	$CsBrCl_2$	283.73	glossy-yel, rhomb		205		s		d al, eth
c301	chloride	CsCl	168.36	col, cub, deliq, 1.6418	3.988	646	1290	162.22	250.10	33.7 MeOH; v s al; i acetone
c302	chloroiodide	$CsICl_2$	330.74	cr, trig	3.86	230	d 290	s		s al
c303	chloroaurate	$CsAuCl_4$	471.68	yel, monocl				0.5	27.5	s al
c304	chlorobromide, di-	$CsBrCl_2$	283.73			205				
c305	chlorodibromide	$CsBr_2Cl$	328.18	yel		191				
c306	chloroiodide, di-	$CsICl_2$	330.73	gn cr, rhomb	3.86	230	d 290	s		s al
c307	chloroplatinate	Cs_2PtCl_6	673.62	yel, cub	4.197 ± 0.005	d 570		0.024	0.377	i al
c308	chlorostannate	Cs_2SnCl_6	597.23	wh, cub	3.33					
c310	chromate	Cs_2CrO_4	381.80	yel pr, rhomb	4.237			71.4	95.5	
c311	chromium sulfate	Cesium chromium alum. $Cs[Cr(H_2O)_6](SO_4)_2.6H_2O$	593.21	vit cr	2.064	116		5.6		
c312	cyanide	CsCN	158.92	very sm wh cr	3.03			v s	v s	
c313	fluoride	CsF	151.90	cub, deliq, 1.478 ± 0.002	4.115	682	1251	367^{18}		191 MeOH; i Diox, Pyr
c314	fluoride	$CsF.1\frac{1}{2}H_2O$	178.93					366.5		
c315	fluorogermanate	Cs_2GeF_6	452.39	isotrop cr, reg oct	4.10			sl s	v s	sl s a
c316	fluosilicate	Cs_2SiF_6	407.89	wh, cub	3.372			0.6	sl s	i al
c317	fluotellurite	$CsTeF_5$	355.50	col need				d	d	s HF soln
c318	formate	$CsCHO_2$	177.93		1.016				2012	

*

PHYSICAL CONSTANTS OF INORGANIC COMPOUNDS (Continued)

No.	Name	Synonyms and Formulae	Mol. wt.	Crystalline form, properties and index of refraction	Density or spec. gravity	Melting point, °C	Boiling point, °C	Solubility, in grams per 100 cc: Cold water	Hot water	Other solvents
	Cesium									
c319	formate	$CsCHO_2.H_2O$	195.94			41, $-H_2O$				
c320	gallium selenate	$CsGa(SeO_4)_2.12H_2O$	704.73	cub cr				4.16		
c321	gallium sulfate	$CsGa(SO_4)_2.12H_2O$	610.93	col cub, 1.46495		2.113		1.21		0.0028 75% al
c322	hydride	CsH	133.91	wh cr, cub	3.41	d		d	d	d a; i org solv
c323	hydrofluoride	$CsF.HF$	171.91	monocl, deliq		160		v s		v s a; i al
c324	hydrogencarbide	$CsHC_2$	157.94	trop cr		300		d		
c325	hydroxide	$CsOH$	149.91	lt yel, deliq	3.675	272.3		395.5		s al
c326	iodate	$CsIO_3$	307.81	wh, monocl	4.85			2.6		
c327	metaperiodate	$CsIO_4$	323.81	wh rhomb pl	4.259			2.15	s	
c328	iodide	CsI	259.81	rhomb, deliq, 1.7876	4.510	621	1280	44°	160	s al
c329	iodide, penta-	CsI_5	767.43	bl, tricl		73				
c330	iodide, tri-	CsI_3	513.63	blk, rhomb	4.47	207.5		sl s	sl s	s al
c331	iodotetrachloride	$CsICl_4$	401.63	pale or needles	3.374	226	d	sl s	sl s	
c332	iron (II) sulfate	$Cs_2SO_4.FeSO_4.6H_2O$	621.87	lt grn, monocl, 1.500, 1.504, 1.509	2.791	ca 70		101.1 (anhyd)		
c333	iron (III) sulfate	$CsFe(SO_4)_2.12H_2O$	567.08	pa-vit cr, 1.484	2.061	ca 90		s	s	
c334	magnesium sulfate	$Cs_2SO_4.MgSO_4.6H_2O$	590.34	col, monocl, 1.486, 1.483	2.674					
c335	permanganate	$CsMnO_4$	251.84		3.597	d 320		0.097	1.27	
c336	mercury bromide(ic)	$CsBr.2HgBr_2$	933.63	rhomb				0.807		sl s al
c337	mercury chloride(ic)	$CsCl.HgCl_2$	439.88	col, cub or rhomb, 1.792				1.64		i abs al
c338	nitrate	$CsNO_3$	194.91	col, hex or cub, 1.55, 1.56	3.685 liq 2.71	414	d	9.16	196.8	s acet, sl s al
c339	nitrate, hydrogen	$CsNO_3.HNO_3$	257.92	col		100				
c340	nitrate, dihydrogen	$CsNO_3.2HNO_3$	320.93	col pl		32–36				
c341	nitrite	$CsNO_2$	178.91	yel cr				v s	v s	
c342	oxalate	$Cs_2C_2O_4$	353.83		3.230			282.0		
c343	oxide	Cs_2O	281.81	or need	4.36	d 400; m.p. 490 (in N_2)		v s	d	s a
c344	oxide, per	Cs_2O_2	297.81	pa yel need	4.25	400	650, $-O_2$	s	d	s a
c345	oxide, tri-	Cesium oxide, sesqui- Cs_2O_3	313.81	choc br cr, cub	4.25	400		d		s a
c346	phthalate, hydrogen	$CsHC_8H_4O_4$	298.03	rhomb	2.178					
c347	polonium chloride	Cs_2PoCl_6	688.53	cub, 1.86	3.82					
c348	rhodium sulfate	$CsRh(SO_4)_2.12H_2O$	644.12	yel, oct	2.238	110–111		sl s		
c349	rhodium sulfate	$CsRh(SO_4)_2.12H_2O$	644.12	or cr	2.22	111		sl s	s	
c350	salicylate	$CsC_7H_5O_3$	270.02					196.2	1522	
c351	selenate	Cs_2SeO_4	403.77	rhomb, deliq, 1.5989, 1.6060, 1.5994	4.4528			244.8		
c352	sulfate	Cs_2SO_4	361.87	col rhomb, or hex, 1.560, 1.564, 1.566	4.243	1010	tr hex 600	167°	220	i al, acet
c353	sulfate, hydrogen	$CsHSO_4$	229.97	col rhomb pr	3.352	d		s		
c354	sulfide	$Cs_2S.4H_2O$	369.94	wh cr, deliq				v s	v s	
c355	sulfide, di-	Cs_2S_2	329.94	dk red, amorph		460	>800	hyg		
c356	sulfide, di-	$Cs_2S_2.H_2O$	347.96	[illegible]				s		
c357	sulfide, hexa-	Cs_2S_6	458.19	br red		186				
c358	sulfide, penta-	Cs_2S_5	426.13		2.806	210				
c359	sulfide, tetra-	Cs_2S_4	394.06	yel		d 160				
c360	sulfide, tri-	Cs_2S_3	362.00	yel leaf		217	780			
c361	tartrate, hydrogen	$CsHC_4H_4O_6$	281.99	wh, rhomb cr				9.7	100	
c362	tartrate	$Cs_2C_4H_4O_6$	413.88	col, trig	3.0			v s d	v s	
c363	vanadium sulfate	Cesium vanadium alum- $VCs(SO_4)_2.12H_2O$	593.15	red, cub, 1.4780	2.033	82	$-12H_2O$, 230; d 300	0.404	sl s	

*

CESIUM-137 SOURCE MATERIAL FOR AN IRRADIATOR

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Large quantities of cesium-137 are being recovered from fission product waste at the U.S. Department of Energy Waste Encapsulation and Storage Facility (WESF) near Richland, Washington. The recovered cesium-137 chloride is doubly encapsulated for long-term storage and is available as a rugged, reliable source form suitable for use in gamma irradiators.

The 30.17-year half-life, 25 Ci/g specific activity and 0.66 MeV gamma rays combine to make cesium-137 chloride a good gamma ray source material. Commercial experience with cesium-137 chloride sources during the past 25 years and the rigorous tests which the WESF capsule has passed give confidence to the reliability of this source form.

GRAM QUANTITIES @
LOW TEMPERATURE!!

Introduction

Cesium-137 is considered a prime candidate for the gamma source to be used in sludge irradiators because it is a long-lived gamma emitter and is a by-product of nuclear fission. Large quantities of cesium-137 have been present in stored fission product waste since the initiation of plutonium-239 production during World War II, and much larger quantities are potentially available from the operation of nuclear power reactors. An additional impetus for the use of cesium-137 is the potential alleviation of the fission product waste disposal problem by removing a long-lived isotope from the mixture of radioisotopes resulting from fission.

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The first significant quantity of cesium-137 used in a gamma source was isolated at Oak Ridge National Laboratories (ORNL) in 1952. Since then, about 4.0 megacuries of cesium-137 in the cesium chloride source form have been encapsulated in several thousand stainless-steel capsules at ORNL. These sources have been in use for as much as 25 years in commercially supplied irradiators, teletherapy units, and radiography units.

Because of the benefits obtained by the removal of long-lived heat-producing radionuclides from high-level waste tanks, the U. S. Department of Energy (DOE) Hanford site near Richland, Washington, began isolating and separately packaging cesium-137 and strontium-90 for storage in 1974. A total of 38 megacuries of cesium-137 have been processed and encapsulated in stainless-steel capsules at the Hanford Waste Encapsulation and Storage Facility (WESF), and an additional 121 megacuries will be encapsulated by 1990. These capsules are available for purchase at the DOE-approved price of $0.10/Ci, FOB, Hanford.

Characteristics of Cesium Chloride Source Form

The nuclear characteristics of cesium-137 are a bane to the waste disposers but a blessing to the gamma source users. It has a conveniently long half-life (30.17 years) and a reasonably good specific activity in the chloride form (25 Ci/g) for use in gamma sources. Only 35 percent of the cesium produced in fission is the radioactive cesium-137, and it decays with the emission of beta particles and 0.66 MeV gamma rays to stable barium. A by-product of the decay process is the generation of radiation energy at the rate of 0.12 w/g of cesium chloride (4.84 kW/MCi). The radiation energy is converted to heat by the absorption of the emitted radiation within the source (2.67 kW/MCi) and in the irradiated material and irradiator structure (2.17 kW/MCi).

The decay of cesium-137 assures a steady replacement market to maintain the gamma field intensity in an irradiator. The relatively long half-life is on the side of the irradiator owner, however, since he will need to add only 2.2 percent of the original activity each year on the average. An irradiator handling the sludge of a moderate-sized city would have an initial loading of about 5 megacuries and would require an average annual addition of 110,000 Ci of cesium-137, which is the amount contained in two WESF capsules. The additions, of course, can be made less frequently; for example, 500,000 Ci in eight WESF capsules every 4.5 years. The original capsule loading should be good for at least one half-life, or 30 years.

The physical characteristics of cesium chloride are on the side of gamma irradiator utilization for the most part, but there are some less desirable traits that must be considered in designing the radiation source. Being a monovalent alkali metal, cesium in the chloride form is quite soluble in water, just as sodium chloride is soluble. The cesium chloride source form is, however, effectively isolated from contact with water or other external environment by highly reliable encapsulation as indicated in a subsequent section of this paper. Phase changes include a solid phase transition at 460°C involving a 17-percent volume increase and melting at 640°C.

The theoretical density of cesium chloride is 3.97 g/cm^3, but the practical density of cold-pressed cesium chloride pellets is 3.3 g/cm^3. In practice, cesium chloride is tamped into capsules to achieve a density of 2.5 g/cm^3 to leave free space for expansion if the capsule should be heated above 460°C. The same safety factor is provided in WESF cans since they are filled by pouring molten cesium chloride.

Cesium Chloride Encapsulation

The time-tested technique for highly reliable containment of radioactive source material is double encapsulation, which is simply placing one sealed capsule inside another. The drawing of the Hanford WESF capsule shown in Figure 1 exemplifies this technique. One end of each capsule is inert gas, tungsten-arc welded (GTA), and submitted to ultrasonic testing in the shop. After the inner capsule is filled with cesium chloride, the cap is positioned and a remote weld by the GTA process is completed. After this weld is helium leak checked, the inner capsule is cleaned to remove excessive cesium-137 contamination and is inserted into the outer capsule. The final seal weld of the cap to the outer capsule is then made and ultrasonically tested for the required penetration and freedom from defects, and the capsule is decontaminated and stored in a water-filled pool.[1]

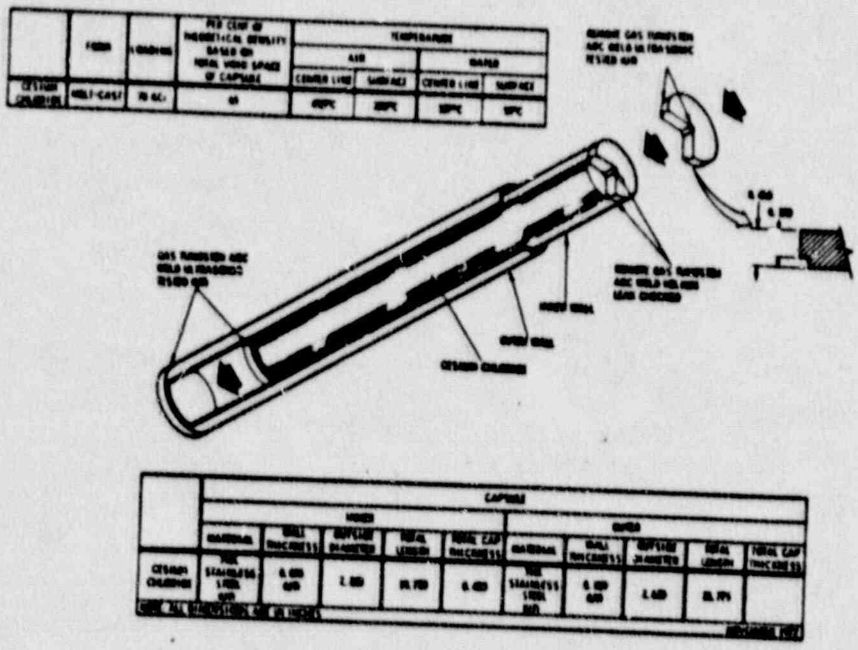

Figure 1. Hanford Waste Encapsulation and Storage Facility Capsules

These routine procedures represent, of course, the culmination of a great deal of previous development work, design, and testing.

After extensive tests of many alloys, Type 316L stainless steel was selected as the most resistant to the possible corrosive effects of cesium chloride. The capsule wall thicknesses were designed for potential stresses, and the weld joint was carefully designed for good weld penetration and excellent weld strength. Many weld tests were made and specimens sectioned to determine the precise welding parameters needed to achieve the required weld penetration. In addition, the welding conditions are regularly checked on test specimens during the actual remote operations.

These operations are typically performed in cells equipped with master-slave manipulators and high-density glass viewing windows in shielding walls 3- to 4-feet thick. Operations are segregated as much as possible in separate cells, and the operations of cesium chloride powder handling, inner capsule welding, and outer capsule welding are done in progressively cleaner cells so that the final capsule is protected as much as possible from external contamination. The capability of safely handling megacurie quantities of dry cesium chloride has been demonstrated during the past 25 years at ORNL and Hanford. This experience provides a high degree of confidence in the capability of irradiator facilities with similar protective features as those on our remote processing cells to prevent the release of any activity from rugged, doubly encapsulated sources.

Cesium Chloride Source Safety

The considerations involved in defining source safety are the physical characteristics of the source form, compatibility between the source form and capsule material, compatibility of the capsule with external environments, and resistance of the capsule to potential stresses. The ability of the capsule to protect the source form in the event of exposure to unusual stresses must be considered for the transportation phase as well as the irradiator resident time.

9-YRS. OLD, TWO PELLET
TOTAL ~ 1 INCH LONG
LOW TEMP. HISTORY

Laboratory tests of cesium chloride compatibility with Type 316L stainless steel showed no adverse effects. These results were verified for authentic source material in actual use in 1963 by sectioning a cesium chloride source made by ORNL in 1954. Photomicrographs of the interior surface of the capsule revealed no pitting, cracking, or change in the crystalline structure.[2] An elevated temperature compatibility test of a fully loaded WESF capsule was conducted as ORNL by holding a capsule containing 69,000 Ci of cesium-137 at 380°C for 279 days. The capsule had been loaded with molten cesium chloride at the Hanford site 2-1/2 years prior to the beginning of the test. AND STORED IN WATER IN A POOL. Metallographic examination of the sectioned capsule after the test revealed a general attack of 1-3 mil with pitting to 10 mil deep. These data can be interpreted as good compatibility resistance for an extended extreme temperature condition at the interface of the capsule wall and source material that would not be encountered in expected use situations. 279 DAYS ≈ 6700 HRS.

10 MILS = 254 μm

The WESF cesium chloride capsule has been qualified as a "special form" for shipping by passing the tests for special form radioactive material specified in IAEA Safety Standards, Safety Series No. 6. The tests consisted of an impact test (drop from 9 m onto an unyielding surface), percussion test (impact equivalent to a 1.4 kg weight falling on the capsule from 1 m), bending test, and heat test (heated in air to 800°C, held for 10 minutes, and allowed to cool). Visual inspection of the capsule after the test series revealed no damage, and the capsule showed no leakage when tested by two methods, including a helium leak test with a sensitivity of $< 1.2 \times 10^{-8}$ std cm^3/sec.

Cesium-137 sources, because of the large amount of radioactivity contained, must be shipped in rugged, heavily shielded containers which are designated Type B packaging in U. S. Department of Transportation regulations. Several shipping casks suitable for the transport of WESF capsules have been licensed by the U. S. Nuclear Regulatory Commission after they were shown to pass the tests specified in CFR 49, Part 173.398, for Type B packaging. These tests consist of the free drop test (a free drop from a distance of 9 m onto a flat, essentially unyielding surface), the puncture test (a free drop of 1 m) onto a vertical, cylindrical mild steel bar (15 cm diam), and the thermal test (exposure to a radiant environment of 800°C for 3 min). Either engineering calculations or tests can be used to show that a cask is capable of performing satisfactorily after these tests.

Once the capsules arrive at the irradiator and are installed in the source rack, the additional protection provided to the capsules consists of the thick shielding walls and double, high-efficiency filtration of the exhausted ventilating air. Periodic swipe tests of the capsules are required by regulations in order to detect possible small increases in contamination on the capsule surfaces. If a storage pool is used in the irradiator, the water is monitored for radioactivity and must meet very strict standards before it is discharged.

The ultimate disposal of spent cesium chloride sources used in municipally owned gamma irradiators is not covered by a well-defined policy at the present time other than by burial in a privately owned radioactive waste disposal site. Since the useful resident time of the original sources in an irradiator will probably be approximately 30 years, a firm national policy on ultimate waste disposal in a national disposal site should be in effect by the time these sources are removed from the irradiator. The WESF, of course, was built for the long-term storage of the WESF capsule, and it is conceivable that a policy permitting the return of spent irradiator capsules to that facility could be worked out.

Conclusions

Cesium-137, because of its availability in large quantities as a by-product of fission in nuclear reactors, is an excellent candidate source for use in sewage sludge irradiators. The capsules used to store cesium chloride in the WESF embody the technology gained by extensive development and experience during the past 25 years. The WESF capsule provides a rugged, well-tested containment of cesium chloride that has been shown to be resistant to a series of unusual stresses.

References

1. L. M. Knights, Waste Encapsulation and Storage Facility Operational Safety Analysis Report, ARH-2965, March 1974.

2. K. W. Haff, Examination of Nine-Year-Old Cesium Chloride-137 Source Capsule, Oak Ridge National Laboratory, ORNL-TM-584, August 1963.

DISCUSSION ON PAPER BY LAMB

KENNETH E. ROACH (Southern Science Applications, Inc.): The question I would like to ask is, does the feasibility of the sludge irradiation program depend on the reprocessing of commercial reactor fuels, or has somebody looked at the amount of sludge to be irradiated and how much cesium will be available from all sources for the next 25, 30 years?

EUGENE LAMB (Oak Ridge National Laboratories): Well, it is not dependent on cesium alone. I would say that if it goes gamma, it would be dependent on two gamma sources, cobalt-60 and cesium. Now, as Theo Ouwerkerk pointed out, there is beginning to be a crunch in cobalt-60 and there probably will be for quite some time. It is difficult to predict just how much market penetration will be gained by cesium-137 or even by gamma radiation of sludge at this point. This type of study is underway.

BART CHEZAR (New York State Power Authority): What would be the annual production of cesium-137 in a large nuclear plant, and what percentage of the nuclear waste from the plant would be cesium-137?

LAMB: You just asked me a question I probably could have answered about a year ago, but right now it does not come to mind as to how much is produced in, say, a thousand megawatt reactor. Theo, do you remember?

THEO OUWERKERK (Atomic Energy of Canada, Limited): I have a few figures.

LAMB: Okay.

OUWERKERK: In 1980, 200 or 300 megacuries per year of cesium from the boiling water reactors, and by 1990, 700 to 1,000 megacuries per year of cesium production.

LAMB: Is that the total amount?

OUWERKERK: Yes. You know, certain assumptions are made. I do not know what your power program will do the next couple of years, but if you wanted to do all your sludge with cesium, I think you would need more than a thousand megacuries per year. It depends. It is also a combination of replenishments, although that is only about 2 or 3 percent per year. It is very difficult to predict. On the cobalt side, we figured that just for medical applications (and as you may know, food is coming up now quickly, so it is pretty difficult for us to predict it) 5 years from now we would need about 30 or 40 megacuries of cobalt-60. We really have to boost our production because we are now at about the 50 megacurie per year level, but I think we can make that. If we also have to apply cobalt in the sludge field, we probably need another 50 or so megacuries of cobalt per year for that application.

LAMB: There was another part of your inquiry. Now, I have forgotten --

CHEZAR: What percentage of the total high-level waste is cesium-137? What is its percentage of the total waste product?

LAMB: Of total activity?

CHEZAR: Yes.

LAMB: It is so dependent upon the time after discharge from the reactor. I have not thought in those terms for so long. I cannot answer the question right now.

BILL REMINI (U. S. Department of Energy): An initial look at the market penetration based on the available cesium indicated 10-percent market penetration of all of the sludge. Looking at how much of the total sludge you would deem acceptable for irradiation, that is sludges that are not heavy metal containing, shows that that percentage would climb up. I think we indicated that somewhere around 30 to 40 percent of the good sludges could be immediately handled with the available cesium.

NEIL CASE (Oak Ridge National Laboratories): After 30 years, you still have after-cesium that can be recovered by removal of barium, so there is another source. I do not think we have really considered that very much in all our source inventory evaluations.

HENRY C. SANDKUHL (Energy Systems): I have been given this afternoon three different figures of the ratio of cesium to cobalt, anywhere from five to seven. Is there any one specific number or does it have that range?

LAMB: Well, it depends on the design of the capsule, both the cesium and cobalt. I believe they are based on the WESF capsule, and the WESF capsule was designed for waste storage of cesium, not for efficiency of output of the gamma. A more efficient source can be designed, something in the order of about 1-inch diameter rather than 2-inche diameter. That would probably bring the ratio down, oh, maybe five to one.

SANDKUHL: It depends on the capsule?

LAMB: Yes.

*

5. Physico-chemical Character of ^{137}CsCl-316L Stainless Steel

A. Currently-used WESF capsules were designed for water basin storage, not as gamma radiation sources.

B. Chromium-nicked steels perform best under oxidizing conditions, since resistance (to corrosion) depends on an oxide film on the surface of the alloy. Reducing conditions, and chloride ions, destroy this film and bring on rapid attack. Chloride ions, combined with high tensile stresses, will cause stress-corrosion cracking. <u>This is true in aqueous solution and needs to be studied in the dry system at elevated temperature</u>.

C. The effects of loss of capsule integrity can be significant due to the high solubility of CsCl and the specific activity of ^{137}Cs. For instance:
   1. At about 30°C. the solubility of CsCl is 200 grams per 100 grams of water.
   2. Less than 0.003 mg. of ^{137}Cs would contaminate a 30,000 gal. storage pool to levels beyond those permitted for release to an unrestricted area.
   3. If a resin column is considered a point source, about 4 mg. of adsorbed ^{137}Cs would result in a gamma radiation level of 1 R/hr. at 1 meter.

D. A long-term (~5 years) compatability testing program on WESF capsules is underway at PNL.
   1. The chloride is about 90% Cs and 10% chlorides of potassium, rubidium, sodium, calcium, and barium.
   2. Theoretical considerations indicate that certain impurities in CsCl could have a significant effect on corrosion rate.
   3. Early results (~6 months aging) at 450°C. interface temperature show 316L corrosion rates of about 0.008 inches/year.
   4. Definite conclusions must await the results of the longer-term tests.
   5. These tests are more indicative of effects expected in dry-storage, dry-irradiation facilities.

E. A Cesium-137 demonstration at Sandia (dry-storage,dry-irradiation) has operated for more than five years irradiating grapefruit, mangoes, and sewage sludge. This experience will be factored into licensing decisions.

F. The use of WESF capsules in wet-storage, dry-irradiation would impose thermal cycling effects.
   1. The CsCl-stainless interfacial temperature undergoes about a 140°C. change between water storage and use in air.
   2. This temperature differential causes an expansion (and contraction) of about 0.014 inches of the circumference at the interface.
   3. If a capsule is used for 1 half-life of Cs-137 (~30 years), and is cycled daily between water storage and dry irradiation, 10,000 cycles are imposed.
   4. The effects of thermal cycling on capsule integrity are not known. The unknown effects include:
      a. The effects of thermal (expansion and contraction) cycling on corrosion rate at the inner capsule-CsCl interface.
      b. The effects of thermal cycling and pool water purity,e.g. chloride content, on the outer capsule.
      c. The effects of temperature and/or thermal cycling on capsule physical stability, swelling, etc.

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6. Safety Precautions

The uncertainties that accrue with long-term use of capsules containing ^{137}CsCl may be offset to a degree by constraints on facility design and operation. These constraints must be based on the properties of the material being irradiated as well as the mode of irradiator operation. For,

A. Dry-Irradiation/Wet Storage, constraints may include:
   1. Periodic sampling and destructive testing of capsules.
   2. Specification of limits on storage pool water quality for both radioactive and non-radioactive constituents.
   3. Water impervious storage pool liners.
   4. Shielded cleanup systems for removal of radioactive contaminants in storage pool water.
   5. Approved plans for operation of cleanup systems.
   6. Approved plans for disposal of radioactively-contaminated resins.

B. Dry-Irradiation/Dry-Storage, constraints may include:
   1. Periodic sampling and destructive testing of capsules.
   2. Ventilation leakage control.
   3. Specification of limits on radioactive constituents in ventilation off-gas.
   4. High efficiency filtration system requirements.
   5. Approved plans for operation of filtration systems.
   6. Approved plans for disposal of radioactively contaminated filters.

C. Commodities, the constraints would be determined by the properties of the commodity, which may vary widely. For example, irradiation of
   1. Fruit, Nuts, and Field Crops may cause imposition of cleanliness standards to prevent stems, leaves, dust, and dirt from impeding transport through the irradiator or movement of the source racks.
   2. Bulk Grains may require that the dust loading of air be controlled to prevent explosion.
   3. Crated or Boxed Commodities may require no constraints other than those currently imposed on medical product irradiators.

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COMPARISON OF RADIOACTIVE ISOTOPES
COBALT-60 VERSUS CESIUM-137

There are many forms of energy, such as chemical, electrical, or nuclear energy, which if used properly and with adequate safeguards, promote well-being and provide a multitude of benefits. One of these forms of energy is emitted as gamma radiation from radioisotopes. A major industry in itself, radioisotopes find applications in tracing, activation analyses, radioisotope generators, chemical processing, medical product sterilization, and in food disinfestation and preservation. Two well-known radioisotopes are cobalt-60 (Co-60) and cesium-137 (Cs-137).

The majority of irradiation facilities in operation worldwide are using a Co-60 isotope as the high-energy gamma ray emitting source. There are approximately 130 industrial irradiation facilities with a total installed capacity of 70 million curies (MCi)[1]. Only a few of these irradiators use Cs-137.

At present usage rates, adequate amounts of Co-60 can be produced to meet the demand. However, with the identified beneficial uses of irradiation technology such as sterilizing medical products, disinfecting sewage sludges, controlling the spread of insect pests including the Caribbean and Mediterranean fruit flies, and improving the storage life of many foods, the demand for radioactive isotopes will increase. To meet the demand, additional production capacity for Co-60 and/or Cs-137 will be required. This analysis comparatively evaluates Co-60 and Cs-137 on the basis of isotope characteristics, production and supply, source utilization, isotope demand, and cost.

ISOTOPE CHARACTERISTICS

Cobalt-60

Radioactive Co-60 is produced via a neutron capture reaction that is initiated by placing the stable isotope Co-59 into a nuclear reactor. When an atom of inactive Co-59 assimilates a low or intermediate energy neutron into its nucleus, the resulting "compound" nucleus, $^{60}Co^*$, is unstable and promptly emits a gamma ray (or photon) to relieve the instability:

$$^{1}_{0}n + {}^{59}_{27}Co \rightarrow {}^{60}_{27}Co^* \rightarrow {}^{60}_{27}Co + \gamma.$$

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[1] A curie is the amount of radioactive isotope needed to obtain 37 billion atomic disintegrations per second. A million curies is denoted as a megacurie or MCi.

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The residual nucleus, Co-60, is still radioactive (half-life[2] = 5.27 years) and will subsequently undergo beta (β^-) decay--a process in which the charge of a nucleus is changed without a change in the total number of nucleons[3]. In simple terms, beta decay occurs when one of the neutrons of Co-60 changes into a proton and an electron, and the created electron (β^- particle) is emitted from the nucleus[4].

$$^{60}_{27}\mathrm{Co} \rightarrow {}^{60}_{28}\mathrm{Ni} + {}^{0}_{-1}\beta.$$

The beta decay of Co-60 generates a new nuclide, Ni-60, the nucleus of which initially lies in an excited or energetic state. As the nucleons in Ni-60 rearrange themselves into a more stable configuration, two photons of energy or gamma rays are released. It is this final release of gamma ray energy which finds practical application in product irradiation. The Ni-60 nuclide is thereafter nonradioactive. The complete decay scheme of Co-60 is summarized in Figure 1.

Cesium-137

Radioactive Cs-137 (half-life = 30.17 years) is produced as a fission product of nuclear reactor fuel. Contrary to what might be expected on intuitive grounds, a fissioning nucleus (i.e., uranium-235) rarely splits exactly in half. Instead, the fission fragments tend to fall into two broad groups: a light group with mass numbers from 80 to 110 and a heavy group with mass numbers from 125 to 155. Two of the most abundant fission fragments from these groups are strontium-90 and cesium-137.

Cesium-137 is similar to Co-60 in that it decays by beta emission:

$$^{137}_{55}\mathrm{Cs} \rightarrow {}^{137}_{56}\mathrm{Ba} + {}^{0}_{-1}\beta.$$

However, the beta decay may lead to either of two different isomeric states[5] of barium-137 (Ba-137). One beta decay leads to the ground-state of Ba-137, a stable nuclide, and

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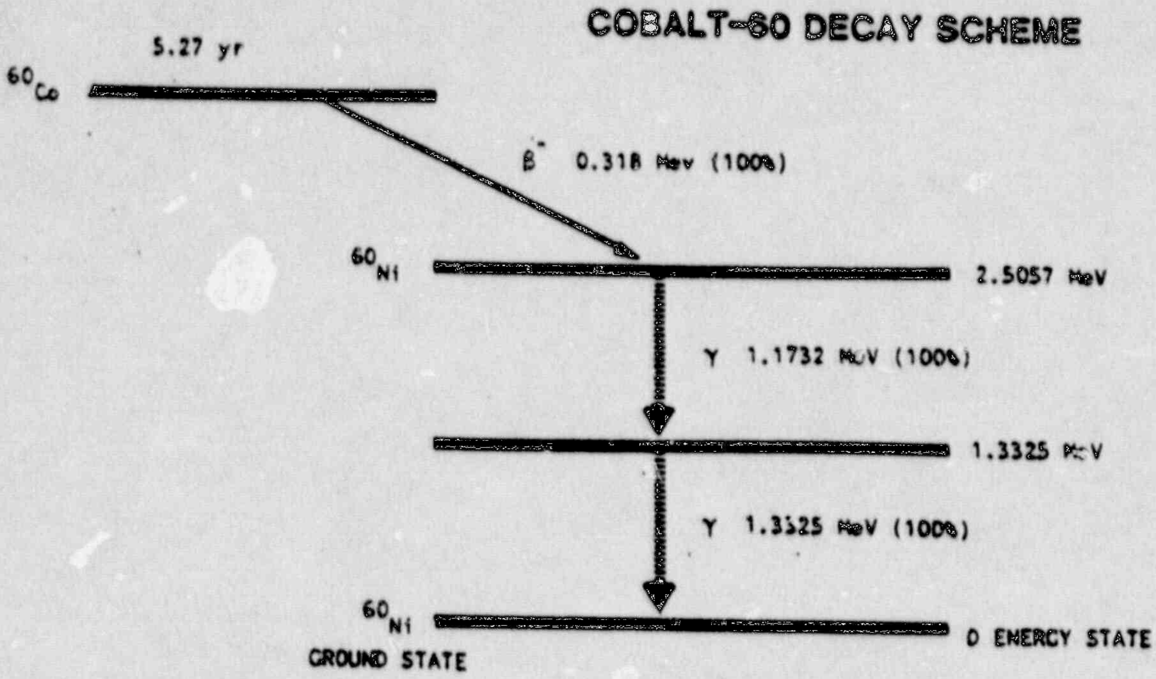
[2] Half-life: the time during which the activity or decay rate falls by a factor of two.

[3] Nucleons = protons and neutrons in a nucleus.

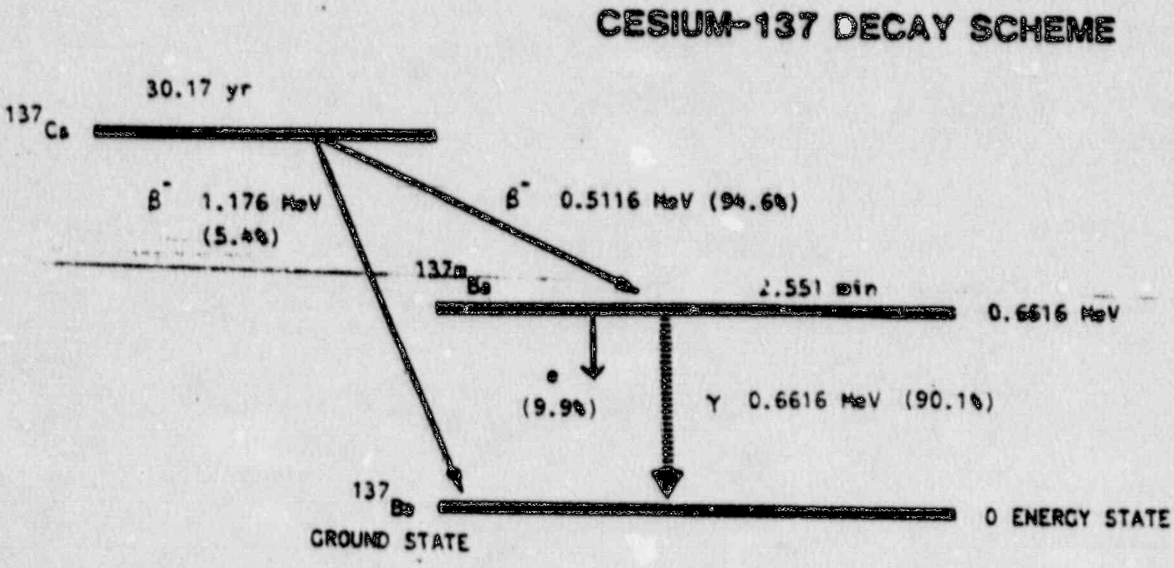
[4] A massless particle called an antineutrino is also emitted during beta decay. It interacts very, very weakly with nuclei and does not produce ionization.

[5] An isomeric state refers to when a nuclide has the same mass number and atomic number but possesses different radioactive properties in different long-lived energy states.



COBALT-60 DECAY SCHEME

Figure 1. Cobalt-60 decay scheme



CESIUM-137 DECAY SCHEME

Figure 2. Cesium-137 decay scheme

one leads to the metastable state of barium, Ba-137m. This last nuclide has its own characteristic half-life (2.551 minutes) and decays by two distinct modes: (1) internal conversion[6], denoted by the symbol (e), and (2) gamma (γ) emission. The ratio of the frequency of these two events, (e/γ), is 0.11. The complete decay scheme of Cs-137 is presented in Figure 2.

Table 1 below lists some of the salient features of both Co-60 and Cs-137.

Table 1
ISOTOPE COMPARISON

Quantity	Co-60	Cs-137	Ratio of Co-60 to Cs-137
Gammas per decay	2.0	0.85	2.35
Energy (MeV) per gamma	1.33,1.17	0.662	1.89
Energy (MeV) per decay	2.5	0.56	4.45
Half-life (years)	5.27	30.17	0.17

Note that while Co-60 has both higher energy gammas and more gammas per decay than Cs-137, it also has a much shorter half-life.

The energy levels of the gammas emitted from Co-60 and Cs-137 are substantially below the energy threshold required for target activation. Therefore, it is not possible for the gammas to generate "radioactive" materials, regardless of the total amount of radiation energy absorbed.

Material sensitivity to a radiation environment (e.g., discoloration or breakdown of a target material) is dependent on (1) the total quantity of radiation energy absorbed, (2) the rate that the energy was deposited, and (3) the various mechanisms and interaction processes by which the radiation deposits its energy in the target. Usually materials exhibit little sensitivity to items (2) and (3) above. It is the total quantity of radiation energy absorbed by the material that is the dominant factor. Items (1) and (2) are easily controlled by varying the amount of source material and/or the product throughput in a facility. As for item (3), the energy deposition mechanisms do differ according to the type of radiation (e.g., neutron, charged-particle, or gamma-ray) and the energy spectrum of the radiation. However, in comparing just those gamma-rays with

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[6] An internal conversion process involves the direct transfer of energy from the nucleus to one of the orbital electrons and the electron is then ejected from the atom.

energies between 1 and 3 MeV, the manner in which the gamma energy is [illegible] is very similar. Thus, for a particular facility with equivalent amounts of either Cs-137 or Co-60 such that items (1) and (2) are satisfied, the amount of radiation enhancement or damage to a product will be the same regardless of which isotope is used.

ISOTOPE PRODUCTION AND SUPPLY

Isotope production often entails long and arduous manufacturing processes, each of which must be covered by extensive quality control measures to ensure that the final product meets all of the engineering and safety specifications. Cobalt and cesium sources are no exception.

Cobalt-60

The procurement and manufacturing process for the cobalt source pins, as followed by the Atomic Energy of Canada, Ltd. (AECL), is listed below:

1. Cobalt-rich ores are mined, principally, in Zaire, Canada, and the Soviet Union.
2. The ore is refined and purified.
3. The cobalt powder is formed into slug- or pellet-form and covered with a layer of nickel to prevent oxidation.
4. The Co-59 is welded into zircalloy capsules, the capsules assembled into bundles, and the bundles formed into long rods for placement into a reactor.
5. After a suitable period of neutron activation (12 to 18 months), the target rods are removed from the reactor and dismantled.
6. The cobalt capsules are placed in hot cells for double encapsulation. The first layer is usually zircalloy, and the second layer is stainless steel. Each encapsulation is performed separately with its own set of stringent weld examinations and tests.
7. These source pins are then wipe-tested, measured for curie content, and transferred to a water pool for temporary storage prior to shipping. The total production time required for the source pins, from activation of the Co-59 through the encapsulation and shipment, is 15 to 24 months.

The source pin dimensions may vary greatly depending on their specific application but normally measure greater than 40 cm in length and approximately 1 cm in diameter. The source pins are kept thin to minimize heat buildup and maximize efficiency due to the high self-absorption of the cobalt: self-absorption factors (including the encapsulating material) range from 5 to 9 percent for a 1 cm diameter source pin. The specific activity of the cobalt is a strong function of thermal neutron flux and irradiation time in the reactor but usually varies from 70 to 100 curies per gram for industrial applications.

The 1983 installed commercial base of Co-60 in the U.S. is approximately 32 MCi. The current U.S. supply of Co-60, monitored by DOE and produced at the Advanced Test Reactor facility in Idaho, accounts for only 25 percent of the annual source replenishment requirement, or about 1 MCi per year.

Most of the world's demand for Co-60 is being met through production at a nuclear reactor complex near Pickering, Ontario, Canada. This facility has the capacity to produce approximately 13 MCi/yr. Coupling the current capacity with the additional proposed capacity from seven new reactors scheduled to be in operation by 1985, and four more scheduled to be in operation by 1990, the Canadian facilities have a potential annual Co-60 production capacity of 63 MCi. If worldwide demand expands to a point where this rate of production is inadequate, three other alternatives could be expected to contribute to the supply of Co-60: (1) the Canadian facilities could increase production an additional 30 percent by operating at less than optimum conditions (this would require increased fuel burnup and associated higher prices), (2) Canadian research reactors and other sources could increment the supply another 3 to 7 MCi[7], and (3) other reactors in the U.S. and worldwide could initiate Co-60 production.

Cesium-137

Rockwell International currently encapsulates radioactive cesium at the Waste Encapsulation and Storage Facility (WESF) at Hanford, Washington. The steps in the production of the WESF capsule are as follows:

1. The high-level wastes containing cesium are removed from the underground storage tanks and purified by ion exchange.

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[7] Fraser, F., 1983, "Cobalt-60 Supply, Demand and Price," AECL Third Gamma Processing Seminar, Technical Paper GPS314.

2. Elution of cesium from the ion exchange bed generates Cs_2CO_3. This is later converted to CsCl via an exothermic reaction with HCl.

3. The purified CsCl solution is boiled down to a solid salt. The salt is dried and then brought to a molten state by induction heating.

4. The CsCl is melt-cast into a stainless steel cylinder. After cooling, a sintered disk helium source is added, a lid is welded on, and the capsule integrity is checked using a helium leak test and a bubble test.

5. After decontamination of the capsule, it is placed into another stainless steel cylinder for double encapsulation and the welds are ultrasonically inspected.

6. The source capsules are measured for curie content and placed in storage.

Figure 3 illustrates the physical dimensions and characteristics of the WESF gamma source pin. The source strength for the WESF capsule is approximately 25 curies per gram, or approximately 70 thousand curies per capsule. A source plaque containing Cs-137 WESF capsules would be four to five times as large in area, yet contain half as many capsules as a Co-60 source plaque generating an equivalent flux field. The size of the plaques and the number of capsules they must hold are merely facility criteria that are accounted for in preliminary design.

The WESF capsules were originally designed for pool storage. Therefore, the efficiency of the capsules is relatively low (less than 70 percent) and the thermal heat load is greater than for the slender Co-60 source pin. The outer surface temperature of the WESF capsule (in air) has experimentally been determined to be on the order of 100°C[8]. In anticipation of their use as gamma radiation sources, various participating national laboratories subjected the source pins to a battery of tests, including free drop, percussion, heating, and immersion[9]. This evaluation of the capsule's performance and integrity showed that the welds and thick

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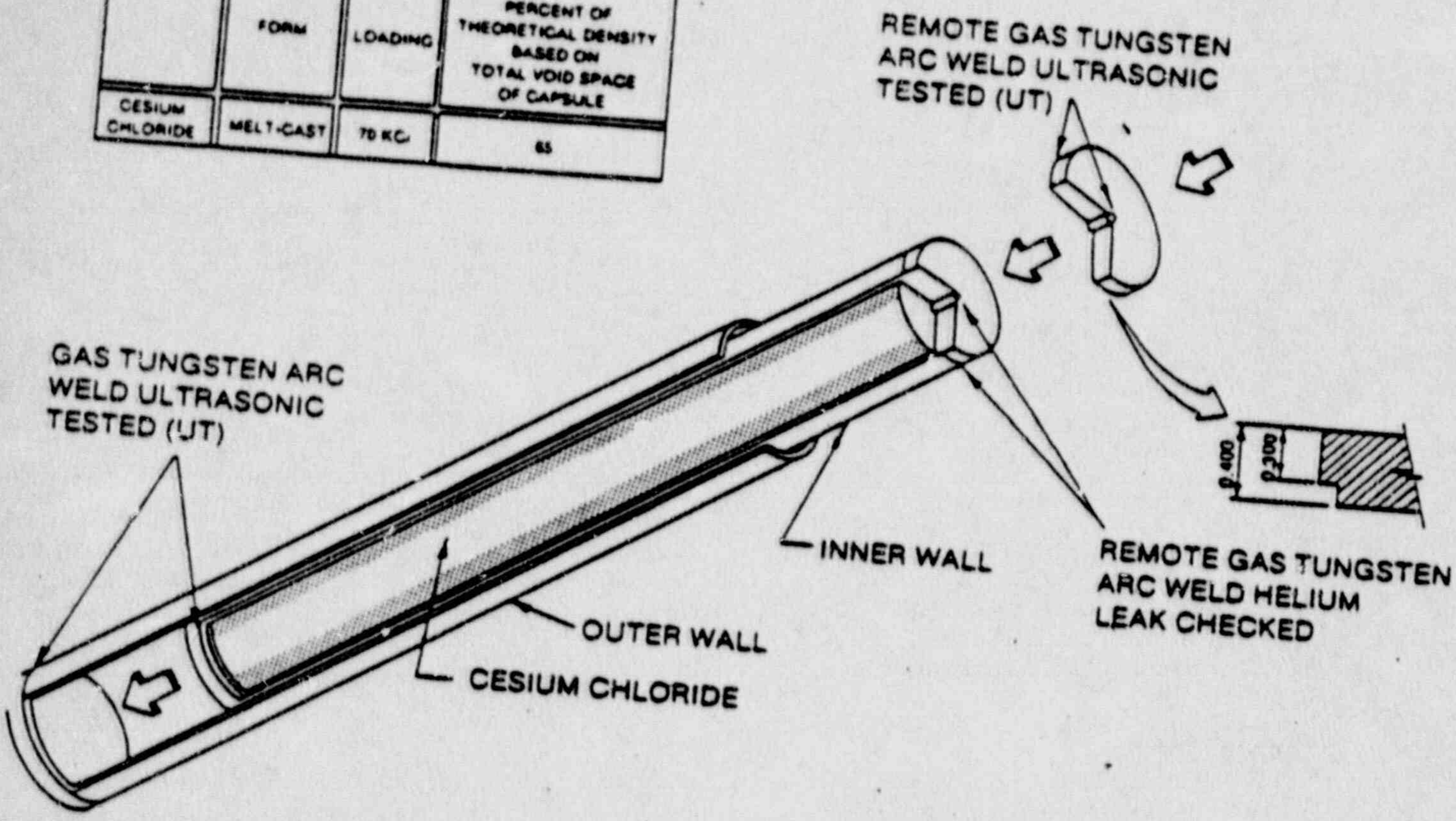
[8] Kenna, B.T., and Schultz, F.J., "Characterization of an Aged WESF Capsule", SAND83-0928. See also: Sandia National Laboratories, Byproduct Utilization Program, Bimonthly Report, March/April 1983.

[9] Kenna, B.T., "WESF ^{137}Cs Gamma Ray Sources", SAND82-1492. See also: Niemeyer, R.G., "Leak Testing Encapsulated Radioactive Sources", ORNL-4529.

	FORM	LOADING	PERCENT OF THEORETICAL DENSITY BASED ON TOTAL VOID SPACE OF CAPSULE
CESIUM CHLORIDE	MELT-CAST	70 KCi	65



	CAPSULE									
	INNER					OUTER				
	MATERIAL	WALL THICKNESS	OUTSIDE DIAMETER	TOTAL LENGTH	TOTAL CAP THICKNESS	MATERIAL	WALL THICKNESS	OUTSIDE DIAMETER	TOTAL LENGTH	TOTAL CAP THICKNESS
CESIUM CHLORIDE	316L STAINLESS STEEL (UT)	0.095 (UT)	2.250	19.725	0.400	316L STAINLESS STEEL (UT)	0.109 (UT)	2.625	20.775	0.400

NOTE. ALL DIMENSIONS ARE IN INCHES

Figure 3
WASTE ENCAPSULATION AND STORAGE FACILITY (WESF) CAPSULE

double steel walls provided substantial structural strength. Corrosion was also shown to be minimal.

The source material in the WESF capsule, CsCl, is soluble in water (162.3 gm/100 gm water at 0.7°C and 290.0 gm/100 gm water at 119.5°C). This does not mean that when in contact with water, the CsCl reacts vigorously or effervesces. In the unlikely event of a capsule leak, the dissolved or leached cesium could be removed relatively easily by passing the solution through ion-exchange columns.

There are approximately 193 MCi of Cs-137 available from high-level nuclear wastes located at Hanford and the Savannah River Plant (SRP). Approximately 90 MCi of this Cs-137 has been encapsulated and is ready for immediate use. Table 2 shows the estimated Cs-137 inventories from various sources[10].

The recovery, but not packaging, of Cs-137 from spent nuclear fuels is currently being planned at SRP for waste management purposes. Assuming that packaging facilities are utilized at SRP for product form conversion and encapsulation, then the potential cumulative amount of Cs-137 available by 1990 from both Hanford and SRP will be approximately 250 MCi. This quantity of Cs-137 could be greatly increased if the cesium from commercial spent fuel was also scheduled for recovery. Figure 4 shows the cumulative potential inventory of Cs-137 available from high-level wastes up to the year 2000 (the graph is taken from the reference in footnote 10).

Two transportation casks are being fabricated by the Department of Energy for use in loading Cs-137 irradiators with the WESF capsules. Each cask will contain 16 capsules. The cask bodies have been forged, machined, and surface treated. Manufacture of the lids will be contracted shortly. The half-scale model tests leading to Nuclear Regulatory Commission, Department of Transportation, and International Atomic Energy Agency Type B licensing will start in December 1983. The estimated completion date for the full-scale casks, including licensing, is July 1984.

SOURCE UTILIZATION AND DOSE UNIFORMITY

Calculations have been performed to determine the relative source utilization of Co-60 and Cs-137. The source utilization indicates how efficiently the gamma radiation produces

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[10] Wheelwright, E.J., 1983, "Byproduct Inventories", Waste Management '83, Vol. 2, p. 123-127.

Table 2
ESTIMATED INVENTORIES OF CESIUM-137 FROM VARIOUS SOURCES

Source	Effective Inventory Date	MCi
Hanford		
- Cesium chloride capsules through 6/82	1/83	63.6
- Cesium chloride capsules through 10/83[a]	1/83	27.7
- Stored wastes[b]	1/83	26
- Future wastes	1/83 to 1/91	38.7
Savannah River Plant		
- Current wastes	1/83	102
- Future wastes	1/83 to 1/2001	109
Commercial Spent Fuel		
- Accum through 1981	1/83	510
- Accum. through 2020	1/2021	11,000

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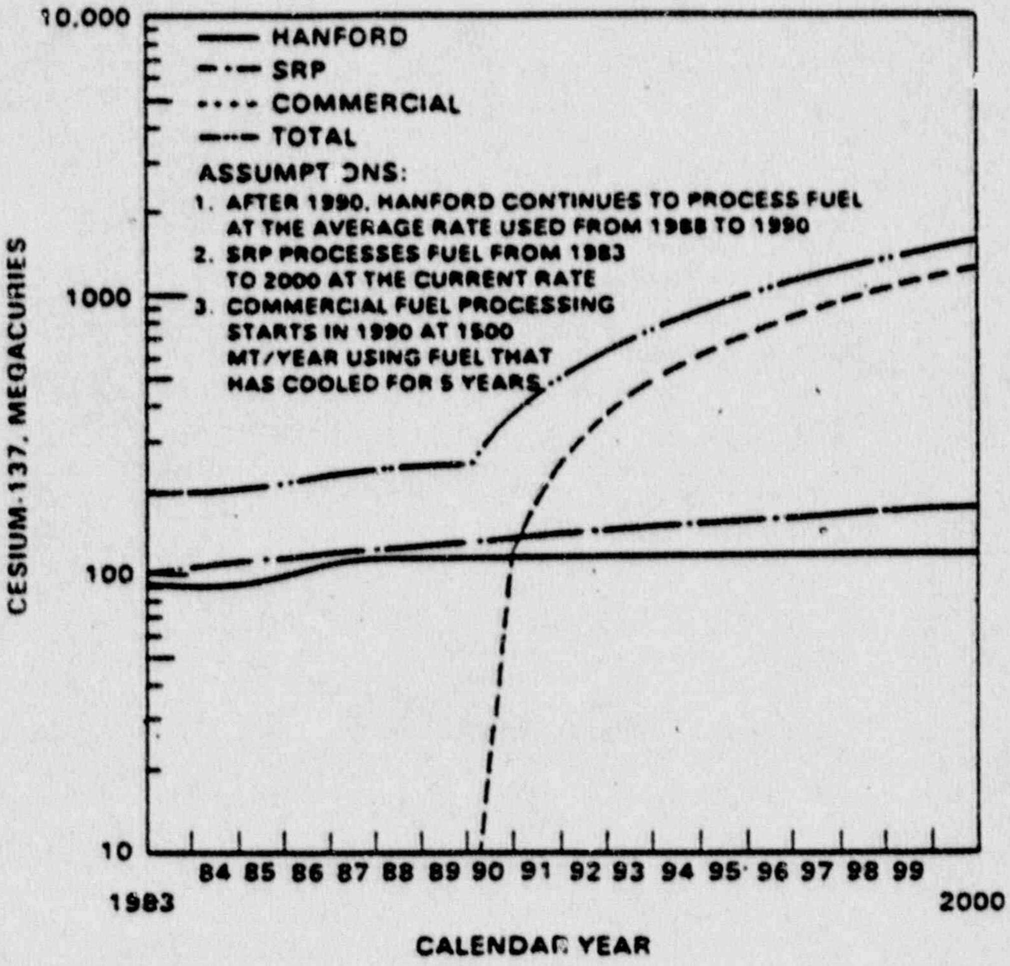
[a] Completion date established through personal communication with I.E. Reep of Rockwell-Hanford.

[b] Cesium contained in current waste but not considered recoverable.



ASSUMPTIONS:

1. AFTER 1990, HANFORD CONTINUES TO PROCESS FUEL AT THE AVERAGE RATE USED FROM 1988 TO 1990
2. SRP PROCESSES FUEL FROM 1983 TO 2000 AT THE CURRENT RATE
3. COMMERCIAL FUEL PROCESSING STARTS IN 1990 AT 1500 MT/YEAR USING FUEL THAT HAS COOLED FOR 5 YEARS

Figure 4. Cumulative inventory of Cs-137 available from high-level wastes

a desired effect. It is usually given in terms of dose[11] per unit source (Mrad per MCi). A high value for the source utilization would indicate a relatively high absorbed dose for a unit quantity of isotope.

The calculations were performed using a Monte Carlo transport code (MORSE-SGC) for three different case studies:

Case 1: Co-60 versus Cs-137 (each assumed to be 100 percent efficient[12]; no self-absorption).

Case 2: Co-60 source pins (95 percent efficient) versus the CsCl WESF capsule (68.61 percent efficient).

Case 3: Co-60 source pins (95 percent efficient) versus a 1-inch diameter CsCl capsule (86.34 percent efficient).

The 95 percent efficiency used for the Co-60 source pins in Cases 2 and 3 was employed to approximate the efficiency of a standard AECL source pin. Case 3 was studied to determine the incremental benefit achieved by optimizing the design parameters of the WESF capsule for commercial irradiation rather than pool storage.

The ratio of the source utilization of cobalt to the source utilization of cesium reaches its maximum value (largest difference in the benefits from the two isotopes) when all of the energy available from the nuclides is absorbed in the target material (requires essentially an infinite target mass). These maximum values are shown below in Table 3 for Cases 1, 2, and 3.

Table 3
MAXIMUM SOURCE UTILIZATION RATIOS
(Co-60/Cs-137)

	Case 1	Case 2	Case 3
$\frac{\text{Mrad/MCi of Co-60}}{\text{Mrad/Mci of Cs-137}}$ =	4.45	7.41	5.53

As might be expected, Case 2 has a higher ratio than either Case 1 or 3 because of the relatively low WESF capsule efficiency. If the diameter of the WESF capsule (2.625 inches) was decreased, then self absorption would decrease, the

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[11] Dose: energy deposited per unit mass; all calculations were referenced to dose in water.

[12] Efficiency is defined as the number of gammas which escape from the source geometry per source gamma.

gamma energy spectrum would harden, and the ratio for Case 2 would decrease to that given for Case 3 (used a 1-inch diameter CsCl capsule) and eventually approach that given for Case 1 (point source). Two interpretations of the values in Table 3, assuming very large targets and using Case 2 as an example, are:

(1) a Co-60 irradiator with some specified number of curies has a plant capacity (or provides an absorbed dose) 7.41 times larger than that for a WESF capsule irradiator using the same number of curies of Cs-137.

(2) a WESF capsule facility requires 7.41 times more source (curies of Cs-137) to provide the same total absorbed dose to a target as a Co-60 irradiator.

For finite-sized targets, the source utilization becomes dependent not only on source capsule efficiencies and source gamma energies but also on how that energy is deposited along the gamma's track through the target. Figure 5 shows a graph of source utilization ratios (dose per unit source) for the same three case studies, for target depths of 0 to 75 cm, and for specific gravities (sp gr) of 1 and 0.1. Note that the utilization ratio curves decrease initially from some characteristic value at zero thickness (Case 1 = 4.04, Case 2 = 6.73, Case 3 = 5.02) to a minimum, and then swing back up to approach the maximum values shown in Table 3 as the target thickness becomes very large. The location of this minimum value or dip in the curve will move to the right (toward larger values of target thickness) as the density decreases. The reason for the initial decrease in the utilization ratio curve is twofold:

1. The physical observation that gammas interact more strongly with target atoms as a function of decreasing energy and increasing density creates a positive feedback process for the energy deposition. Because the source energy spectrum from Cs-137 lies at a lower energy than Co-60, the Cs-137 gammas will travel on the average a slightly shorter distance or path length in the target before they have an interaction. With each interaction, they lose energy; this increases their probability of interaction and decreases their path length correspondingly. Thus, the gamma energy spectrum from Cs-137 broadens out more quickly than the spectrum for Co-60 and the energy deposition becomes slightly more efficient.

2. At energies near the source energies of the two isotopes, backscattering (reflection) of photons

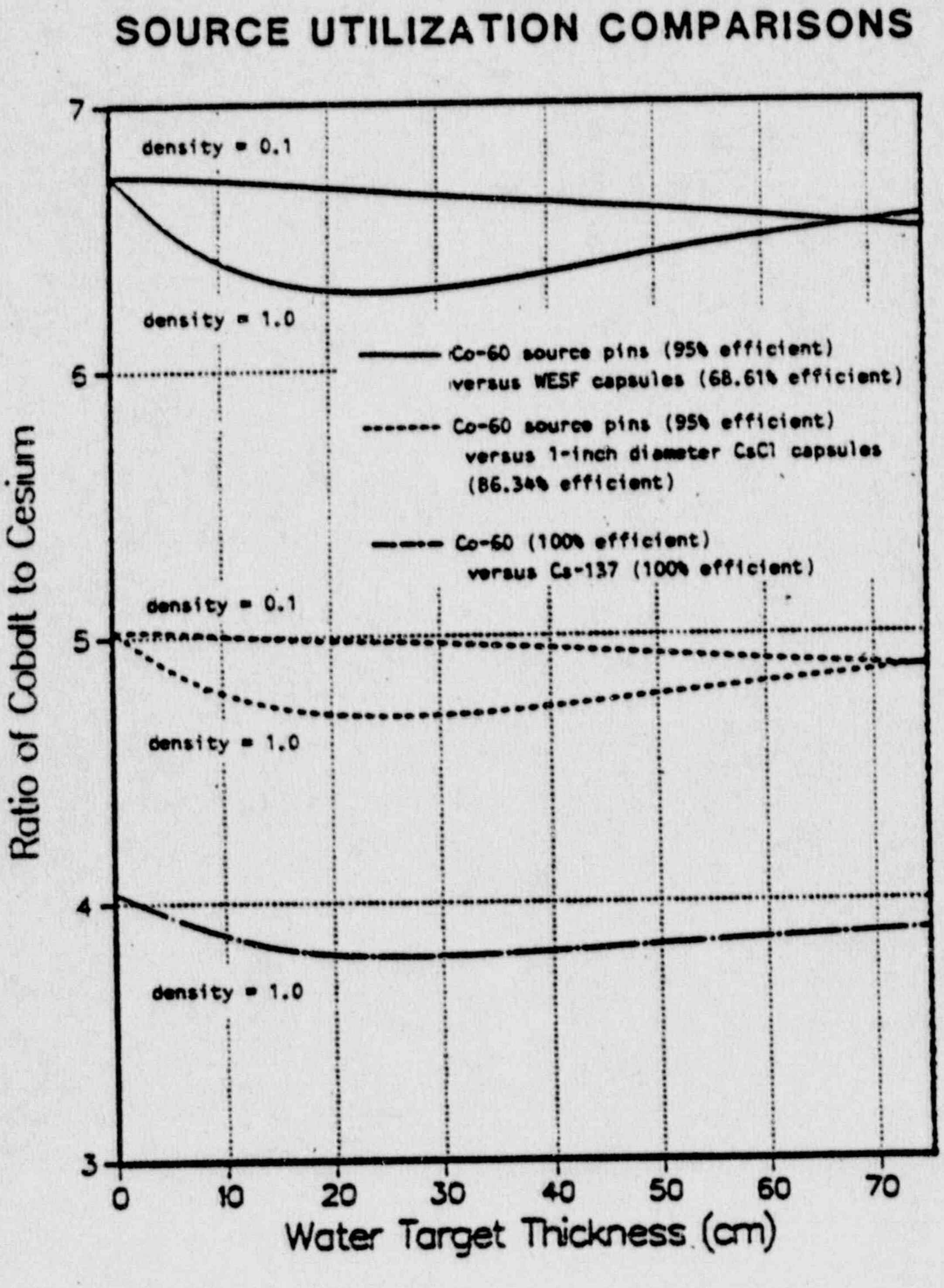

Figure 5. Source utilization comparison between Co-60 and Cs-137 (dose per unit source)

> is greater for Cs-137 than for Co-60. This holds true until the energies of the gammas drop low enough that the dominant interaction is absorption (photoelectric absorption) rather than scattering (Compton scattering). Since backscattering causes more energy to emerge into the backward hemisphere, the energy deposition from the cesium gammas becomes more efficient.

Thus, Figure 5 shows that for many targets of finite size, (1) the plant capacity for a Co-60 irradiator ranges from 6.29 to 6.73 times larger than the plant capacity for a Cs-137 irradiator using WESF capsules, assuming equal source strength (same number of curies), or (2) the source requirement (number of curies needed) for a Cs-137 irradiator using WESF capsules ranges from 6.29 to 6.73 times larger than the source requirement for a Co-60 irradiator, assuming the same product dose.

In addition to source utilization, another basis for isotope comparison is the dose distribution as a function of target depth. Since the energy of the Cs-137 gamma is less than the energy of the Co-60 gammas, and since the probability of interaction with the target nuclei generally increases as a function of decreasing gamma energy, it would seem likely that the Cs-137 radiation would be attenuated or absorbed much more quickly than the Co-60 radiation in passing through a target material. However, a gamma does not necessarily dissappear everytime it interacts with matter. Sometimes it just scatters. If such was not the case, it would be much more difficult to obtain a relatively flat or uniform distribution with any isotope. Actually, it has been noted that in "materials of low atomic number, the diffusion phenomena are extremely important, and the dose received is due more to the photons that have been subjected to diffusions, rather than to the photons coming directly from the source"[13].

For target irradiation from one side only, Figure 6 shows graphically the necessary target conditions (specifically thickness and specific gravity) for one isotope to provide a flatter or more uniform dose distribution than the other[14].

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[13] Eymery, R., 1974, "The Prospects of Using Cesium for Radiosterilization", Sterilization by Ionizing Radiation, Gaughran, E. R. L., and Goudie, A. J., eds., p. 173-184.

[14] The data base for the graph comes from a Brookhaven National Laboratory report (BNL 11194, p. 50) by F. X. Rizzo, G. E. Cunningham, L. Galanta, and J. H. Cusack. The graph is intended to provide a rough comparison only and should not be used for precise interpretation. See also: Josephson, E.S. and Peterson, M.S., eds., 1982, Preservation of Food by Ionizing Radiation, Vol. 1, p. 151-153.

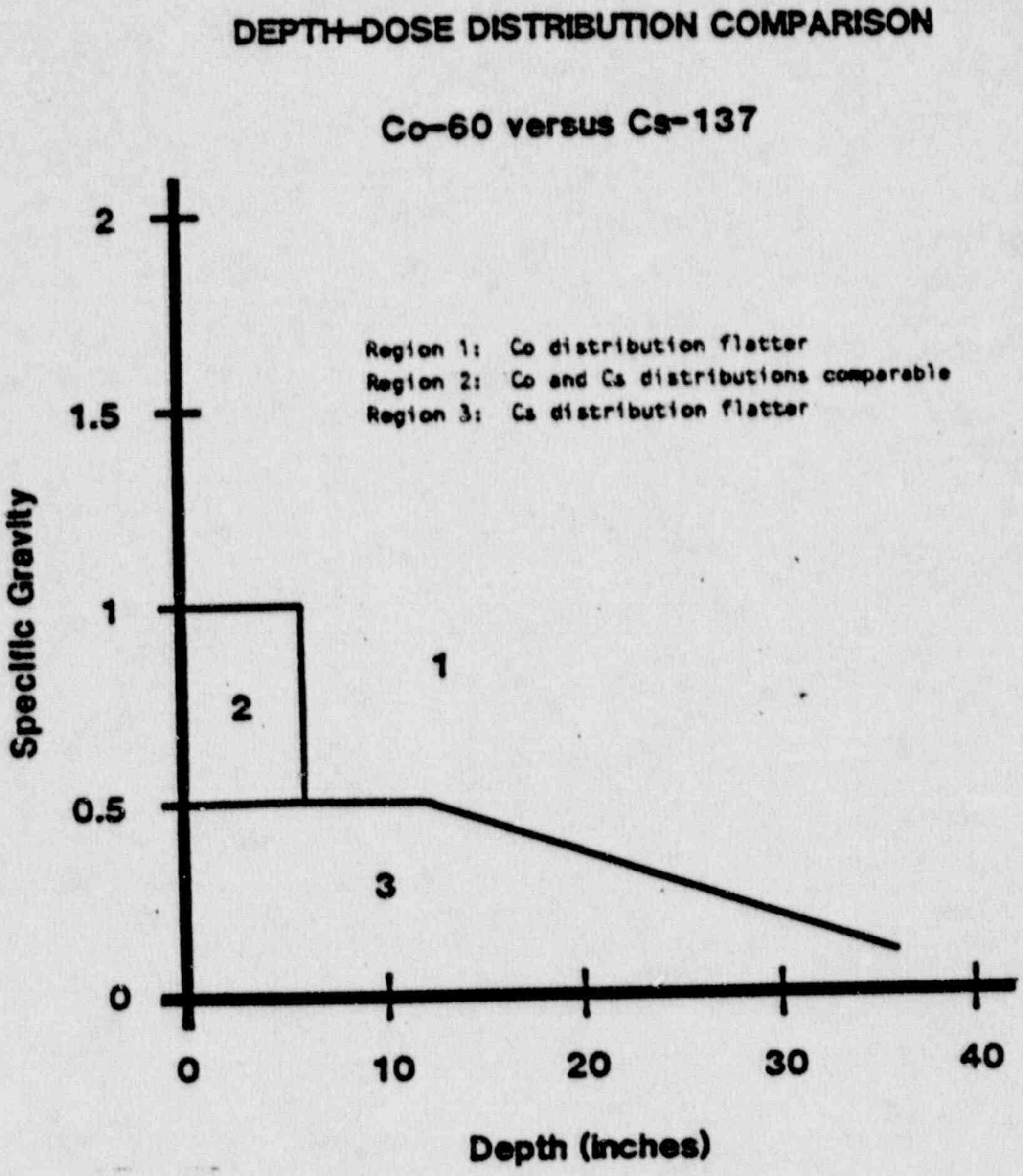

Figure 6. Depth-dose distribution comparison between Co-60 and Cs-137 for one-sided irradiation.

From the standpoint of dose uniformity, Cs-137 appears to be slightly more favorable at low densities and small penetrations, and Co-60 appears to be more favorable at high densities and large penetrations. For two-sided irradiation, however, the differences in depth-dose "are minimized and both isotopes provide a similar uniformity over a wide range of target thicknesses and densities" (see footnote 14 for reference).

ISOTOPE DEMAND

The current isotope demand in this country consists of material for new irradiators and replenishment material for existing irradiators to offset isotope loss through radioactive decay. With no new irradiators, the demand for replacement material is approximately 4 MCi/yr of Co-60 plus 0.04 MCi/yr of Cs-137. Any growth in this industry will produce an immediate isotope demand for the irradiation facilities to be brought on-line, plus an associated increase in the long-term demands for source replenishment. The actual future demand will be primarily dependent on the amount of new irradiation capacity installed.

The predicted worldwide cumulative Co-60 demand over the next 10 years will be on the order of 400 MCi (see footnote 7 for reference) to 500 MCi. This assumes that food preservation and waste treatment applications will grow to the extent of requiring about 88 MCi (or 20 percent of the total) over the next 10 years. Primary uncertainties in predicting the supply-demand relationship are (1) the rate of growth of the medical products and consumer products market, (2) the rate of conversion of medical product sterilization facilities from using ethylene oxide (ETO) to using gamma irradiation (ETO is considered a health hazard by OSHA), and (3) time delays in the construction or administration of facilities for the production and/or reprocessing of the required isotopes.

The potential supply of Co-60 over the next 10 years, approximately 435 MCi, will be adequate to meet only the less optimistic of the two demand predictions. If WESF capsules containing CsCl were to be used to meet the worldwide demand, at least 2700 MCi of Cs-137 would need to be made available through reprocessing over the next 10 years. The previously mentioned potential supply of 250 MCi of Cs-137 satisfies only 9 percent of this requirement. Encapsulating the CsCl into more efficient capsules (e.g., a one-inch diameter capsule) would provide an increase in the amount of usable WESF curies and satisfy an additional 3 percent of the demand. Reprocessing of commercial spent fuel in concert with efficient capsule design could conceivably increase the total supply of Cs-137 to 650 MCi by 1993 (see Figure 4) and satisfy 25 percent of the 10-year demand.

ISOTOPE COSTS

It is questionable whether the potential production capacity of Co-60 will be sufficient to meet the projected demand. The use of Cs-137 could help to offset any shortfalls in supply which may occur. If the recovery and encapsulation of Cs-137 increases, then to a great extent, the relative demand for the two isotopes will depend on cost.

The mid-1984 price for Co-60 in the United States is approximately $1.00 per curie. For comparison, the price set by DOE for Cs-137 is $0.10 per curie. Taking into account the lower source utilization, Cs-137 is still less expensive on an equivalent basis.

Cesium-137 also decays at a slower rate than Co-60. As shown in Table 1, the half lives of Cs-137 and Co-60 are 30.17 years and 5.27 years, respectively. To maintain the desired level of activity, the gamma-ray emitting source must have material added on a periodic basis. For Co-60, 12.3 percent of the initial charge of isotope must be added on a yearly basis. Due to its longer half life, Cs-137 only requires a 12.3 percent addition every 5.7 years.

Looking at total source costs, assuming a discount rate of 15 percent, a fixed Co-60 price of $1.00/Ci, and neglecting shipping and handling costs, 1 curie of Co-60 replenished annually to maintain the same source strength over a 20-year period, would have a present value of $1.76. An equivalent number of curies of Cs-137 (WESF capsule), with a fixed price of $0.10/Ci, would have a present value of only $0.73. Assuming a 40-MCi demand of Co-60, the use of Cs-137 as a replacement would generate a 20-year present-value equivalent savings of $41 million. The actual first-year savings would be $13 million. Figure 7 shows graphically the 20-year present value for the Co-60 source pin, the WESF capsule, and the 1-inch diameter CsCl capsule.

Figure 8 generalizes the comparison in Figure 7 for different source utilization ratios and different initial isotope costs. Assuming an initial cost for either Cs-137 or Co-60, and using a calculated value for the source utilization ratio (Co-60/Cs-137), one can graphically determine not only the life-cycle costs for that isotope in terms of present worth, but also the cost that must be charged for the alternative isotope to obtain the same present worth. This is illustrated in Figure 8 for a given source utilization ratio of 6.73 (relating the Co-60 source pin and the WESF capsule):

a. An initial cost of $0.10 per curie for Cs-137 gives a 20-year present value of $0.73 per curie (Co-60 curie equivalent). The price for Co-60

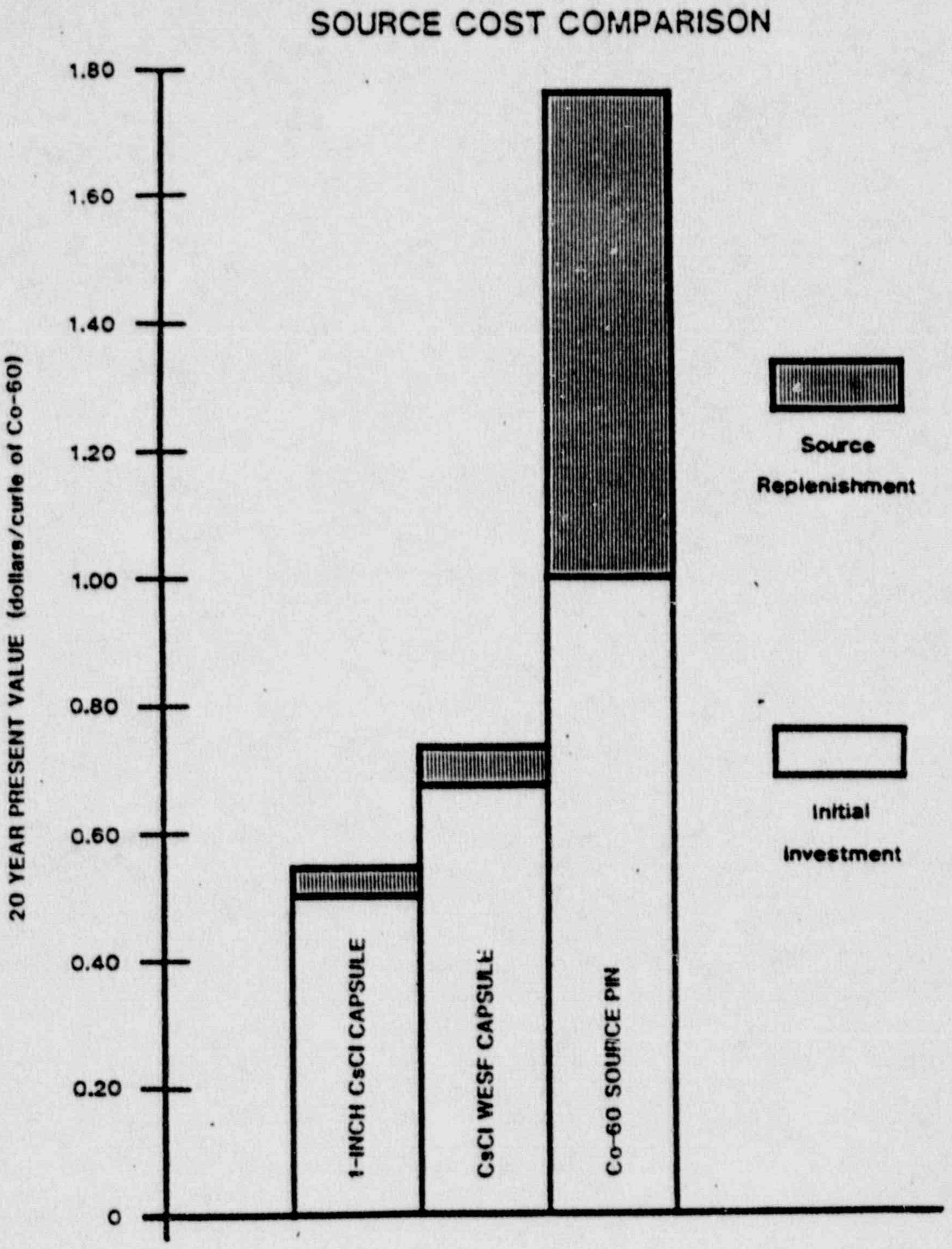

Figure 7. Source Cost Comparison

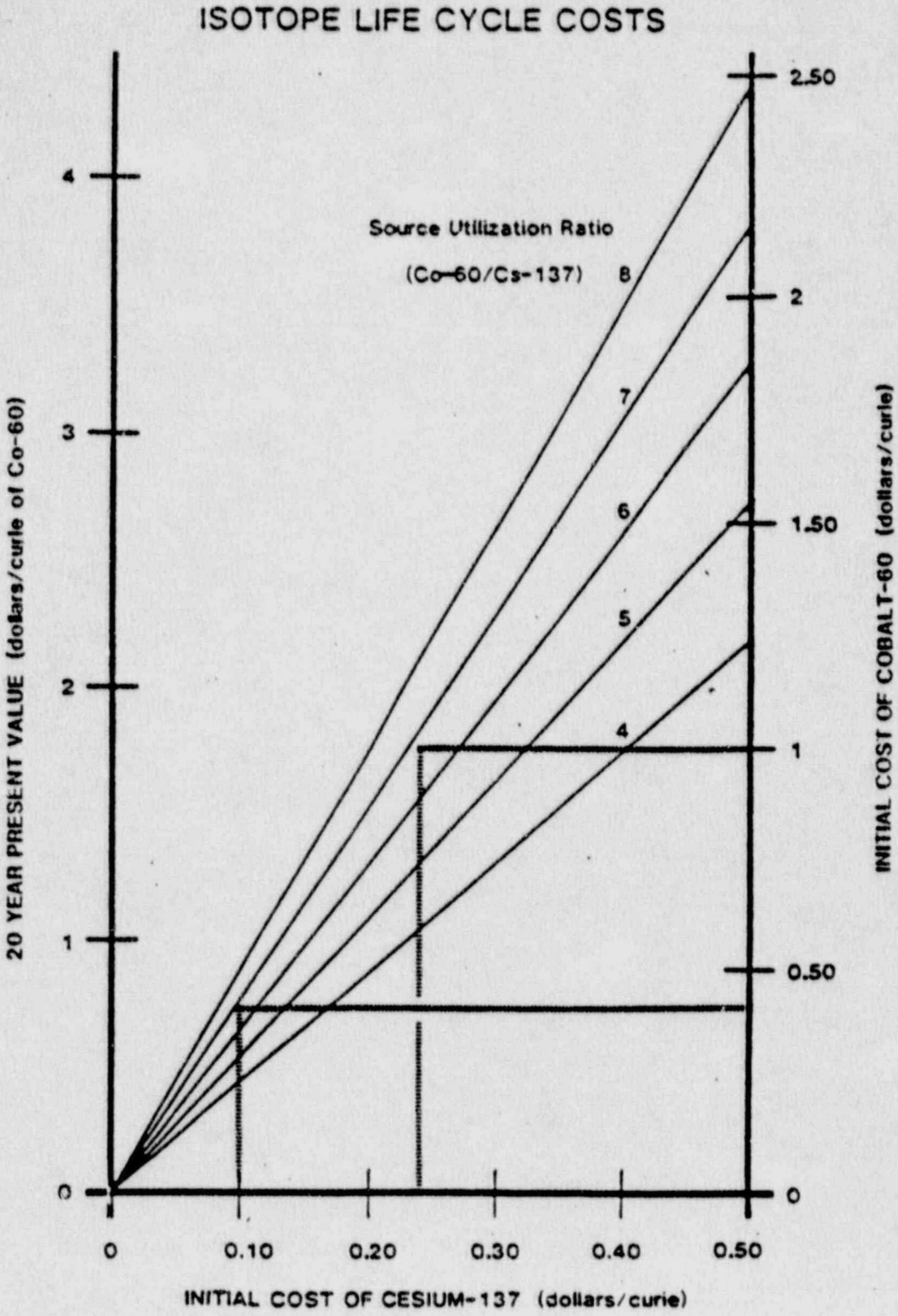

Figure 8. Isotope Life Cycle Costs

need only be $0.42 per curie for the two isotopes to be equal on a present-worth basis.

b. An initial cost of $1.00 per curie for Co-60 gives a 20-year present value of $1.76 per curie. The price per curie of Cs-137 may be raised to $0.24 before the costs of the two isotopes are equal on a present-worth basis.

Transportation/shipping costs were not included in the above analysis. It is estimated that the transportation cost contributes one to two cents per curie to the base price of each of the isotopes, with Co-60 being slightly more expensive than Cs-137 on that basis. However, accounting for the greater curie load of Cs-137 required to render an equivalent dose to Co-60, and acknowledging the differences in source replenishment requirements for the two isotopes, the total transportation costs for Cs-137 are on the order of one and a half times the transportation costs of Co-60. It is important to note that these costs are still only a fraction of the total source costs.

On the other hand, the costs of shutting down the facility, opening it up for replenishment, replenishment labor and time, closing the facility and restarting production are non-trivial costs that must be incurred on an annual basis for Co-60 versus the 5.7 year cycle for Cs-137.

With a significantly lower first cost and life-cycle cost, the use of Cs-137 in new irradiation plants appears financially feasible.

OTHER CONSIDERATIONS

From a purely economic viewpoint, it is more cost effective to use Cs-137 as a gamma-ray source. However, there are other considerations that must be included in a comparison between Co-60 and Cs-137. These include:

- Principal cobalt suppliers are and will probably continue to be outside the United States. The cesium supply is domestic and is regulated by the U.S. government.
- Removing Cs-137 from high-level nuclear wastes and spent nuclear fuel has the added benefit of reducing waste disposal costs.

CONCLUSIONS

The projected growth in the irradiation industry will likely require contributions to the supply by both Cs-137 and Co-60. The widespread use of Cs-137 in the irradiation market will supplement the Co-60 supply as well as reduce cost, eliminate reliance on foreign suppliers, and help reduce radioactive waste disposal problems.

GOVERNOR'S STATEMENT

On Monday morning, Radiation Sterilizers, Inc. (RSI) notified the State that they had shut down their sterilization operation due to higher than normal radiation levels. RSI uses radiation from sealed containers to sterilize medical devices and other items.

Staff of Georgia's Department of Human Resources were immediately dispatched to the facility. They determined that while the radiation levels were relatively high above the storage pool, this area was completely contained and did not pose a danger to the public.

Wednesday evening, the Department of Natural Resources and officials from the Nuclear Regulatory Commission visited the facility to determine whether or not any risk existed outside the building. The levels found outside the RSI facility were essentially at natural background levels and pose no public health problem. I have asked both agencies to work actively with the Nuclear Regulatory Commission to continue to assure there is no public danger, and that every effort is being made to expeditiously correct the deficiencies discovered on Monday morning.

Since any potential exposure to the public would only occur if any of the radioisotopes leak from the building into the environment, the Department of Natural Resources will be the lead State agency in monitoring and coordinating these investigations. For additional information, the Department of Natural Resources may be contacted and Mr. Jim Setser at 656-4713 will provide a response.

6/9/88

To: Ken Clark

A/104

BRIEFING SHEET

Scheduled: September 26, 1988, Monday
10:00 a.m. - 6-B-11 - White Flint

Subject: Leaking Cesium-137 Capsule Incident in Georgia

Purpose of Briefing: To discuss with the NMSS Director and Deputy Director the DOE and Georgia investigations concerning the Georgia Incident.

Items of Discussion:

1. Current status of DOE and State of Georgia Investigations
2. Should NRC be taking any additional actions as follow-up to the incidents?

Handouts: None

Attendees:
H. Thompson
R. Bernero
G. Sjoblom
J. Hickey
M. Lamastra
S. Baggett
P. Vacca
T. Rich
J. Lubenau, GPA
K. Black, AEOD

Additional Distribution:
R. Burnett
M. Knapp
R. Browning
J. Austin

A1105

CHARACTERISTICS OF RADIOISOTOPIC HEAT SOURCES

	^{60}Co	^{90}Sr	^{106}Ru	^{137}Cs	^{144}Ce	^{147}Pm	^{170}Tm	^{210}Po	^{238}Pu	^{242}Cm	^{244}Cm
1. Watts/Gram (100% Basis)	17.8	0.933	33.1	0.416	25.6	0.333	13.6	144	0.56	120	2.84
2. Half-Life Years	5.24	28.6	1.0	30	0.78	2.62	0.35	0.38	87.8	0.45	18.1
3. Curies/Gram (100% Basis)	1141	139	3394	87	3180	928	6048	4500	17	3320	81
4. Curies/Watt	64.2	149	102	209	124	2786	445	31	30	28	29
5. Estimated Isotopic Purity %	10[a]	61[b]	3.6[b]	45[b]	5.5[b]	95[b]	10[a]	95	80	90	95
6. Compound Form	Metal	$SrTiO_3$	Metal	CsCl	Ce_2O_3	Pm_2O_3	Tm_2O_3	Metal	PuO_2	Cm_2O_3	Cm_2O_3
7. Melting Point of Compound °C	1495	2040	2310	645	2190	2320	2375	254	2232	2230	2230
8. Active Isotope in Compound[c]	10	27.0	3.6	34.5	4.23	82	8.8	95	70	75	83
9. Watts/Gram Compound	1.78	0.252	1.19	0.143	1.08	0.273	1.2	137	0.39	90	2.35
10. Density of Compound, g/cm³, actual or 90% TD	8.8	5.03	12.4	3.8	6.9	6.6	8.0	9.2	10.3	11.7	11.7
11. Power Density, W/cm³ Compound	15.7	1.27	14.8	0.545	7.47	1.8	9.6	1260[d]	4.0[d]	1053[d]	27.5[d]
12. Dimension of capsule for 50 W, cm[e]	2.2	4.3	2.2	5.5	2.6	3.9	2.5	1.7	3.1	1.7	1.9
13. Availability	Avail.	Avail.	Poten. Avail.	Avail.	Avail.	Avail.	Avail.	Avail.	Avail.	Poten. Avail.	Avail.
14. Major Types of Radiation*	$\gamma\ \beta$	$\beta\ \chi\ \gamma$	$\gamma\ \beta\ \chi$	$\beta\ \gamma\ \chi$	$\gamma\ \beta\ \chi$	β	$\beta\ \chi$	$\alpha\ \chi$	α	$\alpha\ \eta$	$\alpha\ \eta$
15. Spontaneous Fission Half-Life, Yr.	—	—	—	—	—	—	—	—	4.9×10^{10}	7.2×10^{6}	1.4×10^{7}
16. Shielding Required[f] (inches of lead) [MeV of significant β or γ]	Heavy (9.5) [1.33γ]	Heavy (6) [2.26β]	Heavy (9) [3.35β]	Heavy (4.6) [1.17β 0.67γ]	Heavy (10.2) [2.98β 2.18γ]	Minor (1) [0.23β]	Moderate (2.5) [0.97β]	Minor (1) [0.8γ]	Minor (0.1) [0.04γ]	Minor (0.4) [0.04γ]	Moderate (2) [0.04γ]
17. Bio. Haz., MPC, Total Body, Air, Sol. Continuous Exposure Ci/m³ W/m³ g/m³	10^{-7} 1.6×10^{-9} 8.8×10^{-11}	3×10^{-10} 2×10^{-12} 2.2×10^{-12}	3×10^{-7} 3×10^{-9} 8.8×10^{-11}	2×10^{-8} 9.6×10^{-11} 2.3×10^{-10}	10^{-8} 8×10^{-11} 3.2×10^{-12}	2×10^{-9} 7.2×10^{-11} 2.2×10^{-12}	7×10^{-8} 1.6×10^{-10} 1.2×10^{-11}	2×10^{-9} 6.5×10^{-1} 4.5×10^{-11}	5×10^{-12} 1.7×10^{-14} 2.9×10^{-13}	2×10^{-10} 7.1×10^{-12} 6.0×10^{-14}	9×10^{-12} 3.1×10^{-13} 1.1×10^{-13}
18. Estimated Future Price, $/g(Pure), [Present Price]	285 [456]	20[c] [14]	120[g]	10[g] [8.70]	50[g]	75[g] [186]	136	2800[h]	300	2000[i]	185
19. Estimated Future Price $/W	16	22	5	24	2	220	10	20	540	17	65
20. Total kWh, released/initial gram over a mission period of (yr).	582 (5)	73 (10)	209 (1)	33 (10)	150 (1)	6 (3)	33 (0.4)	350 (0.4)	47 (10)	310 (0.4)	209 (10)
21. Minimum Cost, $/kWh,[i] for (yr) mission	15	6.5	23	7	9.5	320	165	235	133	240	22
22. Grams per ton (at 25000 MWD/t) Recoverable from Reactor Fuels.	—	160	53	835	89	60	—	—	(345Np)	—	9.6(204 for Pu recycle)
23. Production in Power Reactors kg/1000 MWY,		18	2.1	42	5.3	2.9	—	—	(18Np)	—	0.5 (10 for Pu recycle)
24. Est. Availability in 1980, kW,[m]	MWs	1100	6000	1100	10,000	54	MWs	MWs	232[k]	—	119 185[l]

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NEED SUMMARY OF POSITION ON NESF CAPSULES:

DRY STORAGE

1. REFERENCES
2. SUMMARY OF PERFORMANCE
3. NRC POSITON - A. OK or NOT OKAY
   B. LICENSE RESTRICTIONS

WET STORAGE

1. REFERENCES
2. DATA ALREADY OBTAINED
3. DATA STILL NEEDED
4. NRC POSITION - DEMONSTRATION ONLY?
5. WHAT CONDITIONS TO PLACE ON LICENSE?

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