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PRODUCTION OF RADIOISOTOPIC GAMMA RADIATION
SOURCES IN JAERI

April 1980

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(Received March 5, 1980)

The present state of production of gamma radiation sources in Japan Atomic Energy Research Institute (JAERI) is described.

Sources of ^{192}Ir , ^{60}Co and ^{170}Tm for industrial and ^{198}Au and ^{192}Ir for medical applications are produced and delivered routinely by JAERI.

Prefabricated assembly targets are irradiated in JRR-2, JRR-3, JRR-4 or JMTR. The irradiated targets are disassembled in a heavy density concrete cave or a lead-shielded cell, depending on the level of radioactivity. The yield of radioactivity in each target is measured with the aid of an ionization chamber. Where necessary, irradiated targets are encapsulated hermetically in capsules of aluminum, stainless steel or other material. The yield of radioactivity is estimated in relation with the burn-up of target nuclide and product nuclide.

Keywords: Gamma Radiation Source, Iridium-192 Source, Cobalt-60 Source, Thulium-170 Source, Gold-198 Source, Radioisotope Production, Production Facility, Neutron Activation, Sealed Source, Radioactivity Yield, Target Assembly, Burn-up, Waste Disposal, Decontamination, JAERI.

原研におけるガンマ放射線源の製造

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(1980年3月5日 受理)

日本原子力研究所におけるガンマ放射線源の製造の現況が報告される。日本原子力研究所では、工業用線源として ^{192}Ir 、 ^{60}Co 、 ^{170}Tm 線源が、医療用線源として ^{198}Au 、 ^{192}Ir 線源が定常的に製造され、頒布されている。

あらかじめ組み立てられたターゲット試料は、JRR-2、-3、-4、JMTRのいずれかの原子炉で照射される。照射済みの試料は、その放射能強度に応じて、重コンクリートしゃへいケーブまたは鉛しゃへいセルにおいて解体される。各試料の生成放射能は電離箱で測定される。照射済みの試料は、必要に応じて、アルミニウム、ステンレスまたは他の材質のカプセル内に密封される。放射能生成量の評価は、ターゲット原子核と生成原子核のバーンアップの関連で述べられる。

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1. Introduction

The demand for radioisotopic gamma radiation sources in Japan has increased steadily over past ten years in accordance with developments of their utilization in the fields of industry and medical science. This tendency will continue still in future.

In Japan radioisotopic gamma radiation sources have been produced since 1967 in Radioisotope Center, JAERI. During the course of the production, practical problems inherent to the production of radioisotopes for radiation sources have been solved and techniques for fabricating them to sources have been developed. Above all were studied especially in detail the problems of choice of shape and dimensions of targets to suppress self-shielding effect, design of target assembly for reactor irradiation to prevent local depression of neutron flux and determination of irradiation conditions to yield prescribed radioactivity without excess or deficiency. Techniques of post-irradiation handling of highly radioactive materials including hermetical encapsulation were also essential items of development for the production of radiation sources. The radioisotopes produced for radiation sources in JAERI are shown in Table 1. The numerical values in Table 1 are taken from "Table of Radioisotopes".⁽¹⁾

Iridium-192 and ^{60}Co are the radioisotopes most widely used for non-destructive inspection of industrial products, e.g. gamma-radiography of the blade of jet engine, and for gaging in various industrial processes, e.g. management of erosion of inside wall of a smelting furnace. Thulium-170 is best applied to radiographic inspection of thin materials of low density such as the body of aircrafts because of its low gamma energy. Gold-198 "gold-grain" is especially well suited as radiation sources for brachytherapy of cancer, because of its physiological fitness to in vivo applications. Iridium-192 also has been applied in recent years to interstitial brachytherapy in accordance with the development of "after-loading" techniques.

These radioisotopes are produced by neutron irradiation of targets in the nuclear reactors of JRR-2, JRR-3, JRR-4 or JMTR and shipped routinely to customers.

2. Nuclear properties⁽¹⁾

Iridium-192 is produced by (n,γ) reaction on iridium in a nuclear

reactor. Iridium of natural isotopic composition is composed of 37.3 % of ^{191}Ir and 62.7 % of ^{193}Ir . Thermal neutron capture cross section for the reaction $^{191}\text{Ir} (n,\gamma) ^{192}\text{Ir}$ is 940 barns (400 barns to $^{192\text{m}}\text{Ir}$ plus 540 barns to ^{192}Ir). Iridium-192 undergoes subsequent neutron capture, of which cross section is 1500 barns (Fig. 1). Major part of ^{192}Ir decays with a half-life of 74.17 days to stable ^{192}Pt emitting beta particles of maximum energy of 0.672 MeV (46 %) and gamma-rays of energies of 0.604, 0.468 and 0.308 MeV (Fig. 2).

Cobalt-60 is produced by irradiation of cobalt metals in a nuclear reactor through (n,γ) reaction on ^{59}Co . Cobalt is composed of 100 % of ^{59}Co . Thermal neutron capture cross section is 37 barns (19 barns to $^{60\text{m}}\text{Co}$ plus 18 barns to ^{60}Co as shown in Fig. 1). Cobalt-60 decays with a half-life of 5.27 years to stable ^{60}Ni emitting beta particles of a maximum energy of 0.318 MeV and gamma-rays of energies of 1.17 and 1.33 MeV as illustrated in Fig. 2.

Thulium-170 is produced by irradiation of thulium (III) oxide in a nuclear reactor through (n,γ) reaction on ^{169}Tm . Thulium-170 decays with a half-life of 128.6 days to stable ^{170}Yb as shown in Fig. 2. This decays is accompanied with emission of beta particles of maximum energies of 0.968 MeV (76 %) and 0.884 MeV (24 %). Decay by the latter emits 0.084 MeV gamma ray. Thermal neutron capture cross section of ^{169}Tm is 98 barns. Thulium-170 undergoes subsequent neutron capture, of which cross section is 92 barns (Fig. 1).

Gold-198 is produced by irradiation of gold metal in a nuclear reactor through (n,γ) reaction on ^{197}Au . Gold-198 decays with a half-life of 2.70 days to stable ^{198}Hg emitting beta particles of maximum energy 0.961 MeV (approximately 99 %) and gamma-rays of energies of 0.412, 0.676 and 1.088 MeV as shown in Fig. 2. Gold is composed of 100 % ^{197}Au . Thermal neutron capture cross section is 98.8 barns. Gold-198 undergoes subsequent neutron capture, of which cross section is 2.58×10^4 barns.⁽²⁾ Yield of ^{199}Au to be obtained by neutron capture of ^{198}Au is described in 5-4-3.

3. Facilities for the production of radiation sources

3.1 Irradiation facilities

Four nuclear reactors, i.e. JRR-2, JRR-3, JRR-4 and JMTR shown in Table 2, are used for production of both non-sealed and sealed radioisotopes production in JAERI.

The JRR-2 reactor is used for production of ^{192}Ir and ^{198}Au . It is operated at a thermal output of 10 MW. The maximum thermal neutron flux density available for irradiation amounts to $1 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$ in the hollow cylindrical fuel core. An operating cycle is 3 weeks, comprising 12 days (approximately 288 hr) of operation at full power and 9 days of shutdown for maintenance and refueling of the reactor and assembling and disassembling of experimental facilities.

The JRR-3 reactor is used for production of ^{198}Au and ^{192}Ir for radiotherapy. It is also operated at an output of 10 MW. The maximum thermal neutron flux density in pneumatic tube, which is used for the production of these two radioisotopes is $3 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$. An operating cycle is the same as that of JRR-2.

The JRR-4 reactor is supplementally used for production of ^{198}Au .

The JMTR reactor is used for the production of ^{192}Ir , ^{60}Co and ^{170}Tm for industrial use. Available thermal neutron flux density is $1 \sim 3 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$. An operating cycle includes 24 days (approximately 570 hr) operation at full power of 50 MW and 30 days shutdown.

3.2 Production facilities

Irradiated targets are transferred from the reactors to one of properly shielded facilities in the Radioisotope Production Laboratory.⁽³⁾ A heavy density concrete cave⁽⁴⁾ (Fig. 3) is used for handling ^{192}Ir , ^{60}Co and ^{170}Tm in Curie level. This cave was designed for a series of "hot works"; i.e. decanning of irradiation capsules, disassembling of target assemblies, measurement of yield of radioactivity and encapsulation of irradiated targets for practical uses. The wall of the cave is made of heavy density concrete 65 cm thick, which enables us to handle ^{60}Co up to 500 Ci. The inner wall of the cave is lined with steel plate coated with epoxy resin paint for ease of decontamination after hot-run. The cave is equipped with a pair of Model-8 master-slave manipulators, a periscope and a viewing window of 60 cm high, 80 cm wide and 65 cm thick. There is a 300 kg electric travelling crane at the ceiling in the cave. In the cave are set various equipments such as an automatic dispensing apparatus⁽⁵⁾ for ^{192}Ir of high specific activity, a jig for fixing a capsule to be sealed by remote operation welding, a decanner for irradiated capsule, a well type ionization chamber for measuring radioactivity, a hot-plate for testing leakage of sealed sources, and storage pits for irradiated

targets. The radiation background in the cave usually ranges from 10 mR h⁻¹ to 100 mR h⁻¹ due to contamination.

Production of ¹⁹⁸Au and small gamma radiation sources (less than 1 curie; e.g. ¹⁹²Ir for radiotherapy) is carried out in a conventional lead shielded cell⁽⁶⁾ (lead shielding of 10 cm thick). The inside of the cell is usually kept clean for radioactive contamination.

4. Calculation of yield

Yield of ¹⁹²Ir to be obtained by neutron activation of iridium target, in which significant burn-up of the product nuclide as well as the target nuclide occurs as shown in Fig. 1, is given by

$$A = \frac{N_0 \sigma \phi}{3.7 \times 10^{10} \{1 + (\sigma' - \sigma) \phi / \lambda\}} (e^{-\sigma \phi t} - e^{-(\lambda + \sigma' \phi) t}) \quad (7)$$

where

- A : specific activity of ¹⁹²Ir in Ci.
- N₀: number of target nuclides per gram.
- σ : thermal neutron capture cross section of ¹⁹¹Ir, 940 barns.
- σ' : thermal neutron capture cross section of ¹⁹²Ir, 1500 barns.
- φ : thermal neutron flux density in cm⁻² s⁻¹.
- t : irradiation time in seconds.
- λ : decay constant of ¹⁹²Ir.

Calculation of yield of ¹⁹²Ir, which is produced through cyclic irradiation, is given by following equation derived from equation (1)

$$A_n = A e^{-\sigma \phi (n-1) t_A - \lambda (t_1 - t_A)} \frac{1 - e^{-n \{ \phi t_A (\sigma' - \sigma) + \lambda t_1 \}}}{1 - e^{-\{ \phi t_A (\sigma' - \sigma) + \lambda t_1 \}}} \quad (7)$$

where

- A_n: specific activity of ¹⁹²Ir produced by irradiation of n cycles in Ci.
- n : number of irradiation cycles in integer.
- t_A: single operation period of reactor in seconds.
- t₁: operation period plus suspension period of a reactor cycle in seconds.

The results of calculation according to equation (1) are shown in Fig. 4, where neutron flux density is taken as a parameter.

Burn-up of targets and product nuclides gives targets maximum specific activities at certain irradiation times, which are dependent on neutron flux density.

Yield of ^{192}Ir is affected remarkably by the quantity and the arrangement of targets in an irradiation capsule because of the local depression of neutron flux density caused by large neutron absorption cross section of iridium (440 barns).

The effect of the neutron perturbation by pellet shaped iridium targets was determined by irradiating them in JMTP-CF (Critical Facility). A target assembly (Fig. 5), which consisted of a piece of aluminum cylinder, three cylindrical graphite blocks inside the cylinder to hold iridium pellets, and three thin copper wires placed at the axis of the graphite blocks and inside and outside of aluminum cylinder. After irradiation radioactivity formed in the copper wires along their length was determined. When 190 pieces of pellet shaped iridium targets were irradiated in the nuclear reactor, this assembly caused a local depression of neutron flux density at the middle of the center axis of 64 % of the value obtained when 1 pellet was irradiated (Fig. 6).

Yield of a radioisotope is reduced in practical activation by the effect of self-shielding more or less depending on the size of cross section. When cross section is large, as is the case of iridium, activation takes place only in the superficial layer of target, leaving the interior of the target unactivated. An autoradiograph of an ^{192}Ir cylindrical source of 2 mm in diameter by 2 mm thick shown in Photo. 1⁽⁸⁾ reveals the effect of self-shielding.

In order to obtain accurate yield of ^{192}Ir under these circumstances, yield calculated according to equations (1) or (2) should be corrected for these effects. As shown in Table 3, the value of a factor F for correcting the calculated value against these effects was 0.2 for an iridium pellet of 2 mm in diameter by 2 mm thick and 0.3 for an iridium wafer of 3 mm in diameter by 0.4 mm thick.

Radioactivity of ^{60}Co to be obtained when cobalt target is irradiated in a nuclear reactor can be estimated from the equation (3) because neither cobalt target nor product nuclide suffer from significant burn-up. Radioactivity of ^{198}Au also can be estimated from the equation (3). Yield of ^{199}Au to be obtained by neutron capture of ^{198}Au is described in 5.4.3.

$$A = F \frac{N_0 \sigma \phi}{3.7 \times 10^{10}} (1 - e^{-\lambda t}) \quad (3)$$

where

A : specific activity of ^{60}Co (or ^{198}Au) in Ci.

N_0 : number of target nuclides per gram.

σ : thermal neutron capture cross section of ^{59}Co , 37 barns
(or ^{197}Au , 98.8 barns).

ϕ : thermal neutron flux density in $\text{cm}^{-2} \text{s}^{-1}$.

t : irradiation time in seconds.

λ : decay constant of ^{60}Co (or ^{198}Au).

The correction factor F determined experimentally for cobalt targets of various sizes ranges from 0.9 to 0.5 as shown in Table 4. The value F of gold target is approximately 0.6.

Yield of ^{170}Tm to be obtained by neutron activation of thulium target, in which significant burn-up of the product nuclide as well as the target nuclide occurs as shown in Fig. 1, can be estimated from equation (1).

5. Production

5.1 Iridium-192

5.1.1 Specifications of target

Five kinds of iridium target are used (Fig. 7). Iridium targets for radiotherapy (hairpin, single-pin and seed) are made of platinum-iridium alloy (Ir 20~25 weight %) and enclosed in pure platinum sheath 0.1 mm thick, of which both ends are electroplated with platinum to a thickness of 0.1 mm. The platinum sheath cuts off less penetrating beta rays so as to minimize unnecessary radiation exposure to patients. Details of specification of the targets are shown in Table 5.

5.1.2 Irradiation

Configuration of various target assemblies are shown in Fig. 8. Pellet and wafer shaped iridium targets are loaded alternately with aluminum spacers in holes arranged along the circumference of an aluminum cores (Fig. 8-C) and enclosed in an aluminum capsule. Hairpin and single-pin shaped iridium targets are assembled in an aluminum frame (Fig. 8-D). Seed shaped iridium targets are loaded in a polyethylene tube with nylon spacers (Fig. 7) and enclosed in a polyimide resin capsule. Capsules used

for reactor irradiation of these target assemblies are shown in Fig. 9 and 10.

Cores and frames containing targets and capsules are usually made of high purity aluminum (e.g. AA-1100, Al>99 %). The use of high purity aluminum minimizes formation of undesirable long-lived radioisotopes from impurity elements. The aluminum capsules are cold-welded or welded by electron-beam (EB welding) and submitted to leak test by bubbling in ethyleneglycol under a reduced pressure or by helium leak detector.

Pellet and water shaped iridium targets for production of ^{192}Ir of high specific activity are irradiated mostly in position I-11 (aluminum reflector region) of JMTR for one reactor cycle (approximately 550 hours at 50 MW) in a thermal neutron flux density of about $2 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$ and occasionally in VT-1 hole of JRR-2 for two or three reactor cycles (approximately 580 or 860 hours at 10 MW) in a thermal neutron flux density of about $1 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$. Radioactivity of ^{192}Ir produced under these irradiation conditions ranges from 12 Ci to 18 Ci per 1 pellet and from 9 Ci to 13 Ci per 1 wafer.

Hairpin and single-pin shaped iridium targets are irradiated in horizontal hole of JRR-2 for 40 minutes and in pneumatic tube of JRR-3 for 90 minutes. Seed shaped iridium targets are irradiated in horizontal hole of JRR-2 for 2.5 hours or in pneumatic tube of JRR-3 for 5 hours. Examples of the results of irradiation are shown in Table 6.

5.1.3 Decay heat of ^{192}Ir

Iridium-192 generates heat due to absorption of their beta and gamma rays in themselves. The heat causes many undesirable phenomena, i.e. cracking of glasses, melting of polyvinyl and rubber sheets, and burning of paper and cloth during post-irradiation handling. The rate of generation of the decay heat from an ^{192}Ir pellet of 2 mm in diameter by 2 mm thick of an activity of 21 Ci per piece (a specific activity of 157 Ci g^{-1}) was calculated to be approximately $8 \times 10^{-2} \text{ W}$ per piece (0.57 W g^{-1}) by the theoretical equation.⁽⁹⁾

Temperature measured with a thermocouple at the surface of an irradiation capsule (Fig. 9-2) containing a total activity of 4310 Ci was about 120 °C, when the capsule was placed on the floor of the cave under a normal ventilation condition, i.e. an average flow rate of air of 30 cm s^{-1} . Temperature at the surface of a ^{192}Ir pellet and distribution of

temperature inside the irradiated capsule were calculated from the decay heat and heat transfer between each of components of the target assembly and the irradiation capsule according to the natural convective heat transfer equation.⁽¹⁰⁾

$$\alpha = N_u \lambda / L$$

where

- α : heat transfer coefficient (kcal/m²h°C)
- λ : heat convection of H₂ or air (kcal/m h°C)
- L : outer diameter of irradiation capsule (m)
- N_u: Nusselt number

$$N_u = 0.55 (G_r \cdot P_r)^{1/4}$$

where

- G_r: Grashof number
- P_r: Prandtl number

Figure 11 shows surface temperature of an ¹⁹²Ir pellet as a function of specific activity. The surface temperature of an ¹⁹²Ir pellet of a specific activity of 157 Ci g⁻¹ is approximately 200 °C. The temperature distribution in the irradiation capsule is shown in Fig. 12.

5.2 Cobalt-60

5.2.1 Specifications of target

As shown in Table 5, three kinds of cobalt target, i.e. needle, wafer and pellet, are used. Wafer shaped cobalt target is prepared from cobalt metal either by clothing with high purity aluminum of approximately 0.6 mm thick or by plating with nickel to a thickness of approximately 0.05 mm. Pellet shaped cobalt target is plated with nickel to a thickness of approximately 0.01 mm.

5.2.2 Irradiation^{(11), (12)}

Pellet and needle shaped cobalt targets are loaded in the reactor grade aluminum core (Fig. 8-A) or in holes arranged along the circumference of graphite cores (Fig. 8-B) of various sizes and enclosed in aluminum

capsules as shown in Fig. 10.

Needle shaped cobalt target for the production of small ^{60}Co sources is irradiated from a period from 12 hours to 58 hours in hydraulic rabbit of JMTR in an average thermal neutron flux density of $6 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$. Intensity of radioactivity of the small ^{60}Co sources is divided into three classes of 1 mCi, 10 mCi and 20 mCi per one piece with a standard deviation of $\pm 10\%$. Irradiation conditions and results are summarized in Table 7.

Wafer shaped cobalt target for the production of large scale ^{60}Co sources is irradiated for one reactor cycle in position E-10 or L-5 (beryllium reflector region) of JMTR in a thermal neutron flux density of about $1.9 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$.

Pellet shaped cobalt target for the production of ^{60}Co sources of high specific activity is irradiated for a period from 3000 hours to 5000 hours in the same position as that for wafer shaped cobalt targets. Composition of capsules for irradiation of cobalt targets is shown in Table 8. The specific activity of ^{60}Co produced in these irradiation conditions is 8.5 Ci g^{-1} for wafer shaped target, and from 50 Ci g^{-1} to 70 Ci g^{-1} for pellet shaped target. The results of the irradiation of wafer and pellet shaped targets are shown in Table 9.

5.3 Thulium-170

5.3.1 Specifications of target

Specifications of thulium target are shown in Table 5. Thulium (III) oxide pellet is prepared from thulium (III) oxide (Tm_2O_3) powder of 99.9 % purity in a following way. A desired amount of the oxide powder is transferred to a steel die and moulded into a pellet having a required thickness by applying a pressure about 5 kg cm^{-2} . The moulded powder is sintered in vacuum at 2100°C . The pellets thus obtained have a density of approximately 87 % of theoretical density. The pellet is covered with high purity aluminum jacket of 0.2 mm thick to prevent possible spread of highly radioactive powder during irradiation and post-irradiation handling.

5.3.2 Irradiation

Thulium target is irradiated for two reactor cycles (approximately 1100 hours at 50 MW) in position G-11 (aluminum reflector region) of JMTR in a thermal neutron flux density of about $2.5 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$. The radioactivity of ^{170}Tm is about 40 Ci at the end of irradiation.

5.4 Gold-198 grain

5.4.1 Specifications of target

Specifications of gold target are shown in Table 5 and Fig. 7. Gold target used for the production of ^{198}Au has a purity higher than 99.9 % to ensure purity of radiation of the products. A tiny gold cylinder is enclosed in a platinum sheath, of which both ends are electroplated with platinum to a thickness of 0.05 mm. The platinum sheath cuts off less penetrating beta rays so as to minimize unnecessary radiation exposure to patients. For neutron irradiation, 14 gold targets are loaded into an aluminum tube (inner diameter 0.9 mm, outer diameter 2.5 mm, length 38 mm).

5.4.2 Irradiation^{(13),(14),(15)}

The gold targets assembled into an aluminum tube are enclosed in a polyimide resin capsule as shown in Fig. 10-4.

Each of ^{198}Au grains should have an average radioactivity of 5 mCi at the time of use. Therefore conditions of irradiation of gold target are adjusted according to the period between the end of irradiation and the use because of its short half-life of 2.7 days. Gold target for the production of the ^{198}Au grains is irradiated mostly in pneumatic tube of JRR-3 for a period from 2.5 hours to 17 hours in a thermal neutron flux density of about $3.3 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$ and occasionally in pneumatic tube of JRR-2 for a period from 1 hour to 24 hours in a thermal neutron flux density of about $5 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$ or in T-pipe of JRR-4. Example of irradiation conditions for ^{198}Au grains are shown in Table 10.

5.4.3 Amount of ^{199}Au in ^{198}Au

Having a very large thermal neutron capture cross section (2.58×10^4 barns), ^{198}Au transforms itself into ^{199}Au by successive (n, γ) reaction during irradiation of gold in a thermal neutron flux. The content of ^{199}Au in ^{198}Au after irradiation is calculated by following equation⁽¹⁶⁾

$$A_9/A_8 = \frac{\phi\sigma_8\{(1-e^{-\lambda_9 t}) - \frac{\lambda_9}{\lambda_9 - (\lambda_8 + \phi\sigma_8)} (e^{-(\lambda_8 + \phi\sigma_8)t} - e^{-\lambda_9 t})\}}{\lambda_8\{1 - e^{-(\lambda_8 + \phi\sigma_8)t}\}} \quad (4)$$

where

A_9 and A_8 : radioactivities of ^{199}Au and ^{198}Au respectively
 ϕ : thermal neutron flux density in $\text{cm}^{-2} \text{ s}^{-1}$

λ_9 and λ_8 : decay constants of ^{199}Au and ^{198}Au (2.55×10^{-6} ,
 $2.97 \times 10^{-6} \text{ s}^{-1}$) respectively

σ_8 : neutron capture cross section of ^{198}Au

t : irradiation time in seconds

Calculation by equation (4) shows that yield of ^{199}Au in ^{198}Au under the prescribed conditions of irradiation is less than 0.4 %, which is quite negligible for the use (Table 11).

5.5 Post-irradiation procedures

Handling of ^{192}Ir (except ^{192}Ir for radiotherapy), ^{60}Co and ^{170}Tm after irradiation is carried out in the heavy density concrete cave described before (see 3-2). Irradiation capsules are decanned with a capsule opener, after which the target assembly inside is disassembled, care being taken not to lose tiny pieces of radiation sources during remote handling with the manipulators.

Handling of ^{198}Au grain and ^{192}Ir for radiotherapy are carried out in a cell with lead shielding 10 cm thick. After decanning, irradiation capsules are transferred to another lead cell 10 cm thick to prevent ^{198}Au grains and ^{192}Ir from possible radioactive contamination with radioisotopes induced on the surface of the irradiation capsules.

5.5.1 The automatic apparatus for remote handling of pellet shaped ^{192}Ir

This apparatus was developed to improve efficiency of post-irradiation procedures and accuracy of measurement. Outline of the mechanism (Fig. 13) and the functions (Fig. 14) of this apparatus are as follows.

- 1) Selection of ^{192}Ir pellets and aluminum spacers: approximately 300 pieces of ^{192}Ir pellets and 200 pieces of aluminum spacers are separated automatically from a mixture by the vibrating parts-feeder.
- 2) Supply of ^{192}Ir : iridium-192 pellets are supplied one by one to the transfer arm by the escapement mechanism.
- 3) Measurement and classification of ^{192}Ir : radioactivity of each of ^{192}Ir pellets is measured and the pellets are grouped into three classes according to intensity of their radioactivity.

The apparatus is supplied with power and controlled automatically with the data processor connected to the apparatus. The data processor is set in front of the cave near the viewing window to permit easy operation. The data processor is also connected with an electrometer for

radioactivity measurement. The data processor prints out not only the total number and intensity of radioactivity of ^{192}Ir pellets, average activity and standard deviation, but also these values of each group.

6. Radioactivity measurement

Intensity of radioactivity of a radiation source is generally in the order of curies. Routine radioactivity measurements of ^{192}Ir (except ^{192}Ir for radiotherapy), ^{60}Co and ^{170}Tm are carried out with a well-type ionization chamber placed in lead shielding 10 cm thick in the cave. This measuring system is shown in Fig. 15. Radioactivities of ^{198}Au grains and ^{192}Ir sources for radiotherapy are usually estimated with a well-type ionization chamber in a cell with lead shielding 10 cm thick. These measuring systems are calibrated at times with an ionization chamber, of which efficiency was determined accurately. Error tolerated in most routine measurements of radioactivity is $\pm 10\%$. During past few years of use, these instruments have shown little tendency to drift or deviate, although periodic calibration has been made as a matter of routine.

7. Production of radioactive sealed sources^{(11),(12)}

A large number of the radiation sources such as ^{192}Ir , ^{60}Co and ^{170}Tm have been made into sealed source by encapsulation at JAERI. The encapsulation is performed in the cave by means of remotely operated welding equipment.

7.1 Assembling of sealed sources

In sealed sources, radioisotopes are usually enclosed in metal capsules of design dependent on the operational requirements of source application. The majority of capsules of radiation sources today are made of stainless steel of suitable grades, because this alloy is resistive against heat and corrosion, and can be argon arc welded easily.

Iridium-192 is enclosed in a 304 stainless steel capsule as shown in Fig. 16, and this is loaded in the holder for radiography as shown in Fig. 17.

Sealed sources of ^{60}Co are produced by loading a number of wafer or

pellet shaped sources in a cylindrical capsules of 316-L stainless steel as shown in Fig. 18. Pellet shaped ^{60}Co in a specific activity ranging from 50 to 70 Ci g^{-1} are encapsulated double by using aluminum inner capsule (Fig. 18-Middle). Double sealing ensures higher safety, because it is unlikely that the sealing of both inner and outer capsules will fail. After loading of the ^{60}Co into the capsule, an aluminum spacer is inserted to fill up any free space to prevent ^{60}Co from moving in the capsule. Table 12 shows the detail of examples of the ^{60}Co sealed sources.

Thulium-170 source is enclosed in a titanium capsule, which does not attenuate so much soft gamma rays emitted from ^{170}Tm . Structure of ^{170}Tm sealed source is shown in Photo. 2. Each capsule is sealed by welding a plug to an end of the capsule body.

7.2 Remote weld for sealing capsules

Sealing of the radiation sources must be reliable to prevent leakage of radioactive materials from the capsule.

In JAERI capsules are sealed by tungsten inert gas (TIG) welding. The remote welding equipment consists of a power supply unit for controlling a direct current within a range from 4 to 100 A, a control panel for supplying argon gas to prevent oxidation during welding.

The remote welding device is designed to occupy a minimum of space inside the cave. A capsule to be sealed is vertically held in the chuck of the device and rotated around the vertical center axis of the capsule so that the weld area is always vertical to a fixed tungsten electrode. Rotation of the capsule is given with a remotely operated variable speed drive, and the speed of rotation is adjusted before welding starts. Position of the tungsten electrode can be adjusted by moving its holder vertically or horizontally with the manipulators. The thoriated tungsten electrode is 3.2 mm in diameter and has a sharp needle point. For fixing of capsules into the chuck and removing them from it, operation with the manipulators rather than automatic equipment are used because of the wide diversity in the design of capsules. The power supply and the control panel are placed in front of the cave near the viewing window to permit easy operation. The example of weld conditions of stainless steel capsules are shown in Table 13.

7.3 Tests for sealed sources

Sealed radiation sources often are left in use for a long period without inspection. Failure in sealing may cause radioactive contamination of surroundings as well as sealed sources themselves. Therefore soundness of sealing should be guaranteed to some extent by manufacturers of sealed sources. The tests for sealing are carried out according to following procedures.

- 1) Capsules are rigorously inspected by visual examination before assembling.
- 2) After welding, sealed sources are subjected to wipe test for surface contamination of capsules.
- 3) They are tested for leakage of radioactivity from inside of the capsules.

After welding, the sealed sources are washed by fresh water, and whole surface of sealed sources are wiped with moistened gauze and the activity on the gauze is measured. When the capsules are judged uncontaminated, the sealed sources are immersed in water, which is then heated to boil. After 20 minutes radioactivity in the water shows the presence of blow hole which may be caused by expansion of air inside the capsule. These testing procedures and the limits of detection are shown in Table 14. After passing these tests, the sealed sources are ready for shipment.

8. Waste disposal and decontamination procedure

All radioactive wastes arising in the production of radiation sources are solid such as aluminum capsule, aluminum spacers, vials and polyvinyl chloride sheets. Gamma dose rate of these wastes amounts at maximum to 20 R h^{-1} on their surface. These highly radioactive wastes are packed into a cask surrounded with lead shielding 6 cm thick by remote handling. The cask is checked for leak radiation level and surface contamination after withdrawal from the cave. Gamma ray leak dose rate at the surface of the cask is usually from 100 mR h^{-1} to 150 mR h^{-1} . Low level wastes are disposed of in a carton box periodically. Gamma ray leak dose rate at the surface of the box is below 50 mR h^{-1} .

Contamination of the facilities for the production of gamma radiation sources is loose contamination with hot spots as shown in Photo. 3. The

inside wall of the production facilities are coated with epoxy resin paint, which facilitates decontamination. Rough decontamination is carried out by scrubbing wall and floor with wet paper or cloth at intervals with the aid of manipulators. Thorough decontamination is carried out in the case of prolonged entry into the cave for repair, modification or installation of major equipments. In this case, operating personnels enter the cave after rough decontamination under the protection of an air-line mask and a suit against contamination. These decontamination procedures reduce background of gamma ray dose rate in the cave from approximately 200 mR h^{-1} before decontamination to approximately 10 mR h^{-1} , and the surface contamination on floor is below $10^{-4} \mu\text{Ci per } 100 \text{ cm}^2$.

9. Conditions of shipment

Conditions of transportation of radiation sources are governed by laws dealing with transportation of radioactive substances and radioisotopes legislated according to the IAEA regulations in 1973. Iridium 192 exceeding 20 Ci and ^{60}Co exceeding 7 Ci must be transported as approved type "B M" packages. A container shown in Fig. 19⁽¹⁷⁾ has been designed to this purpose. When ^{192}Ir of about 4.5 kCi are loaded in it, the lead dose rate is a few milli-Roentgen per hour at the surface of the lead cask and a background level at the surface of the "B M" package.

10. Conclusion

Production of radiation sources has been continued in JAERI for past 13 years, during which technical development for new items of sealed sources have been continued steadily. At present 3 kinds of sealed sources are produced, among which ^{192}Ir radiographic source and ^{198}Au grains for brachytherapy cover all domestic demands. Development of ^{192}Ir sealed sources in several types are under progress for brachytherapy.

Besides ^{192}Ir and ^{198}Au , a number of radioisotopes are examined in the field of radiation therapy for possibilities of application to brachytherapy. Above all, ^{137}Cs , ^{125}I and ^{182}Ta as well as ^{192}Ir are thought to be promissive. Techniques for production of these radioisotopes will be developed in the near future.

Acknowledgement

The authors gratefully acknowledge the invaluable assistance of Mr. Katsuhiko Tachikawa. The authors also wish to appreciate the staffs of the Inspection Section for continued assistance at radioactivity measurement.

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Table 1 Kinds of radiation sources produced in JAERI

Nuclide	Half - life	Energy of radiation in MeV	
		β energies	γ energies
^{192}Ir	74.17 d	0.240 (8 %) 0.536 (41 %) 0.672 (46 %)	0.296, 0.308, 0.317 0.468, 0.589, 0.604 0.612
^{60}Co	5.27 y	0.315 (99.7 %) 0.670 (0.18 %) 1.492 (0.08 %)	1.332 1.173
^{170}Tm	128.6 d	0.884 (24 %) 0.968 (76 %)	0.084
^{198}Au	2.7 d	0.290 (1 %) 0.961 (99 %) 1.371 (0.025 %)	0.412, 0.676, 1.088

Table 2 Nuclear reactor in JAERI and their characteristics of interest to the production of radioisotopes

Reactor	JRR-2	JRR-3	JRR-4	JMTR
Fuel	93% enriched U	1.5% enriched UO_2	93% enriched U-A2	93% enriched U
Moderator	Heavy water	Heavy water	Light water	Light water
Cooling	Heavy water	Heavy water	Light water	Light water
Maximum thermal output	10 MW	10 MW	3.5 MW	50 MW
Irradiation hole (thermal neutron flux)	VT-1 ($1 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$) HR ($5 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$)	Hydraulic rabbit ($4 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$)	T-pipe ($8 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$) S-pipe ($5 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$)	I-11 ($1 \sim 3 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$) Hydraulic tube ($3 \sim 6 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$)
Operation periods	288 hr / 1 cycle 13 cycles / 1 year	288 hr / 1 cycle 13 cycles / 1 year	6 hr / 1 day 4 days / 1 week 43 cycles / 1 year	570 hr / 1 cycle 6 cycles / 1 year
Radioisotopes produced in JAERI	^{192}Ir , ^{198}Au , ^{42}K ^{197}Hg , ^{51}Cr , ^{32}P etc.	^{198}Au , ^{60}Co , ^{42}K ^{64}Cu , ^{192}Ir etc.	^{198}Au , ^{42}K etc.	^{192}Ir , ^{60}Co , ^{32}P ^{35}S , ^{51}Cr , ^{170}Tm etc

Table 3 Correction factor for iridium targets

Target size	Factor
2 mm ^φ x 0.2 mm [†]	0.68
2 mm ^φ x 0.5 mm [†]	0.42
2 mm ^φ x 2.0 mm [†]	0.20
1.6mm ^φ x 0.8 mm [†]	0.36
3 mm ^φ x 0.4 mm [†]	0.30

Table 4 Correction factor for cobalt targets

Target size	Factor
0.46mm ^φ x 10mm ^ℓ	0.92
0.91 mm ^φ x 15mm ^ℓ	0.78
1.0 mm ^φ x 1.0mm [†]	0.73
8.0 mm ^φ x 2.3mm [†]	0.52

Table 5 Specifications of targets

Target	Shape	Dimensions*	Weight*	Coating thickness
Iridium	Pellet	2.0 mm ^φ x 2.0mm [†]	140 mg	Bare
	Wafer	3.0 mm ^φ x 0.4mm [†]	60 mg	"
	Hairpin	0.45mm ^φ x 93mm ^ℓ	306 mg	Platinum sheath : 0.1mm
	Singlepin	0.45 mm ^φ x 47.6mm ^ℓ	156 mg	"
	Seed	0.45 mm ^φ x 3.0mm [†]	9.9 mg	"
Cobalt	Needle	0.91 mm ^φ x 15 mm ^ℓ	85 mg	Bare
	"	0.46 mm ^φ x 10 mm ^ℓ	15 mg	"
	Pellet	1.0 mm ^φ x 1.0mm [†]	6.9mg	Nickel plating : 0.01 mm
	Wafer	8.0 mm ^φ x 2.3mm [†]	1.1 g	Aluminum cladding: 0.6mm
	"	8.0 mm ^φ x 2.3mm [†]	1.1 g	Nickel plating : 0.05mm
Thulium	Pellet	2.8 mm ^φ x 2.8mm [†]	100 mg	Aluminum jacket : 0.2 mm
Gold	Grain	0.5 mm ^φ x 2.5mm [†]	8.7 mg	Platinum sheath : 0.15mm

* Numerical values excluding coating material

Table 6 Irradiation conditions and produced radioactivities of ^{192}Ir

Shape of target	Reactor	Irradiation time (hr)	Thermal neutron flux ($\text{cm}^{-2} \text{s}^{-1}$)	* Average of activity (Ci)	Specific activity (Ci g^{-1})
Pellet	JMTR	509	2×10^{14}	19	135
"	JRR-2	767	1×10^{14}	16	114
Wafer	JMTR	509	2×10^{14}	13	217
"	JRR-2	767	1×10^{14}	10	167
Hairpin	JRR-2	40min	6×10^{13}	20mCi	—
"	JRR-3	90min	3×10^{13}	20mCi	—
Singlepin	JRR-3	90min	"	10mCi	—
Seed	JRR-3	120~300min	"	1.0mCi	—

* Activity : at the end of irradiation

Table 7 Irradiation conditions of needle shaped cobalt targets in JMTR and produced radioactivities of ^{60}Co

Dimension of target	Thermal neutron flux ($\text{cm}^{-2} \text{s}^{-1}$)	Irradiation time (hr)	Average of activity (mCi/piece)	Standard deviation (%)
$0.46 \text{ mm}^{\phi} \times 10 \text{ mm}^{\ell}$	6×10^{13}	12	1.2	5.9
$0.91 \text{ mm}^{\phi} \times 15 \text{ mm}^{\ell}$	5×10^{13}	28	11.4	5.7
$0.91 \text{ mm}^{\phi} \times 15 \text{ mm}^{\ell}$	3×10^{13}	58	22.4	8.0

Table 8 Details of irradiation of wafer and pellet shaped cobalt targets in JMTR

Sample No.	Target	Contents (pieces/capsule)	Weight (g/capsule)
1	Ni - plating wafer	23	25.3
2	Al - cladding wafer	3	3.3
3	Ni - plating pellet	~550	3.8
4	Ni - plating pellet	~290	2.0

Table 9 Irradiation conditions of wafer and pellet shaped cobalt targets in JMTR and produced radioactivities of ^{60}Co

Sample No.	Irradiation time (hr)	Thermal neutron flux ($\text{cm}^{-2} \text{s}^{-1}$)	Average of activity (Ci/piece)	Specific activity (Ci/g)	Self-shielding factor
1	393(at32MW)	1.5×10^{14}	5.4	4.9	0.54
2	535(at50MW)	1.9×10^{14}	9.3	8.5	0.57
3	3393(")	1.4×10^{14}	0.33	47.8	0.69
4	5573(")	1.1×10^{14}	0.45	65.2	0.69

Table 10 Irradiation conditions and radioactivities of ^{198}Au grain

Reactor	Irradiation hole	Irradiation time	Thermal neutron flux ($\text{cm}^{-2} \text{s}^{-1}$)	Activity after irradiation (mCi/grain)	Activity at the use (mCi/grain)
JRR-2	HR-1	1h03m	5.4×10^{13}	29.8	4.9
JRR-2	HR-1	1h40m	"	55.0	4.9
JRR-2	HR-1	24h	6.4×10^{13}	384	4.6
JRR-3	HP-1	2h30m	3.3×10^{13}	39.0	5.0
JRR-3	HP-1	17h	"	230	4.9
JRR-4	T-pipe	1h20m	6.0×10^{13}	36.5	5.1

Table 11 Content of ^{199}Au in ^{198}Au after irradiation of gold

Reactor	Irradiation time	Thermal neutron flux ($\text{cm}^{-2} \text{s}^{-1}$)	$^{199}\text{Au} / ^{198}\text{Au}$
JRR-2	1h20m	6.5×10^{13}	5.7×10^{-5}
JRR-3	264h37m	2.8×10^{12}	4.0×10^{-3}
JRR-4	2h30m	4.1×10^{13}	6.0×10^{-5}

Table 12 Examples of ^{60}Co sealed sources

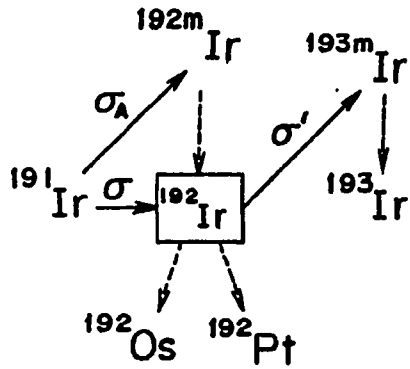
Contents	Weight of cobalt (g)	Total activity (Ci)	Radiation output (R/min at 100cm)	Immersion test(dpm)	Wipe test (dpm)
Al-cladding wafer	17.6	74	1.5	1.3×10^3	$< 10^2$
Ni-plating wafer	26.4	110	2.3	2.7×10^2	$< 10^2$
Ni-plating pellet	18.8	419	9.5	0.8×10^4	$< 10^2$
Ni-plating pellet	9.4	390	> 10	0.1×10^4	$< 10^2$

Table 13 Weld conditions of stainless steel capsules

Conditions	Circumference weld	Spot weld
Weld current (DCA)	20	30
Weld time (sec)	6	2
Rotation of jig (rpm)	10	30
Argon flow rate (ℓ/min)	10	10
Argon flow time (sec)	15	5
Electrode - capsule gap (mm)	3	3

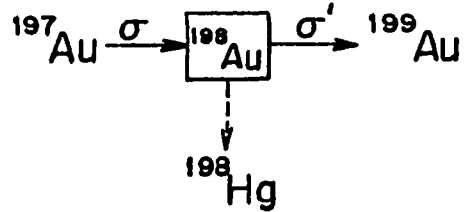
Table 14 Immersion and wipe tests for sealed ^{60}Co sources

Testing	Method	Limit
Wipe test	A sealed ^{60}Co source is wiped with gauze moistened with ethanol, and the activity removed is measured.	$0.005 \mu\text{Ci}$ $1.1 \times 10^4 \text{ dpm}$
Immersion test	A sealed ^{60}Co source is immersed in water at 100°C for 20 min and the activity in the water is measured.	$0.02 \mu\text{Ci}$ $4.4 \times 10^4 \text{ dpm}$



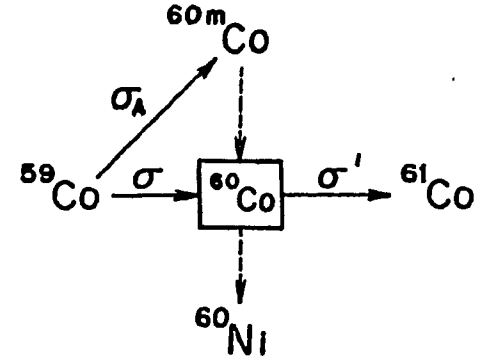
Activation cross section

$\sigma = 540$ barns
 $\sigma_A = 400$ barns
 $\sigma' = 1500$ barns



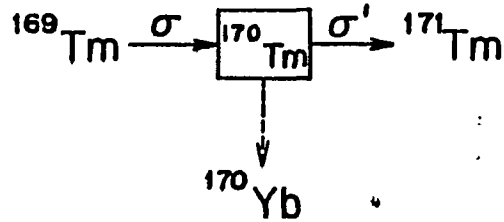
Activation cross section

$\sigma = 98.8$ barns
 $\sigma' = 2.58 \times 10^4$ barns



Activation cross section

$\sigma = 18$ barns
 $\sigma_A = 19$ barns
 $\sigma' = 2$ barns



Activation cross section

$\sigma = 98$ barns
 $\sigma' = 92$ barns

Fig.1 Production processes

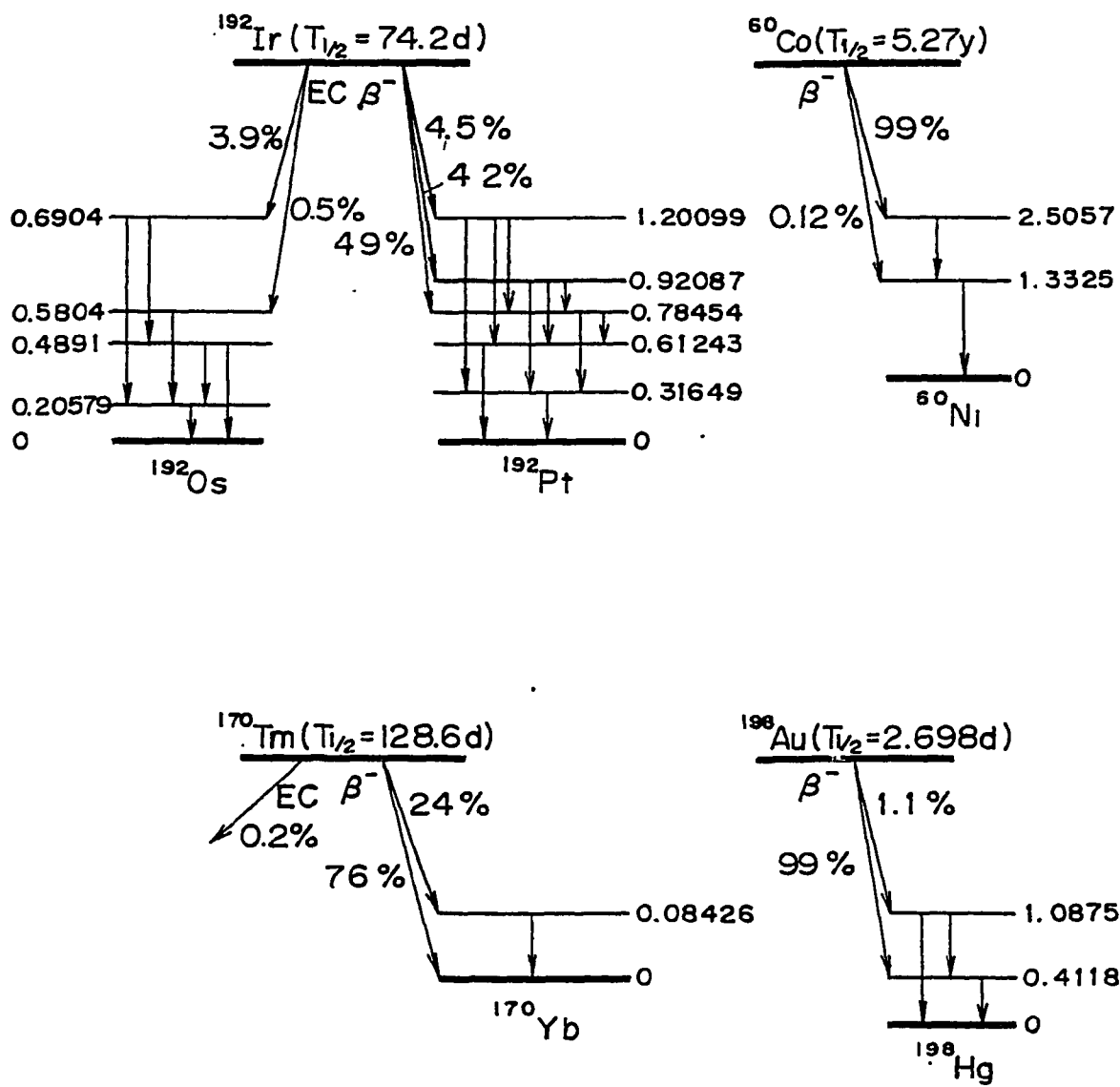


Fig. 2 Decay schema

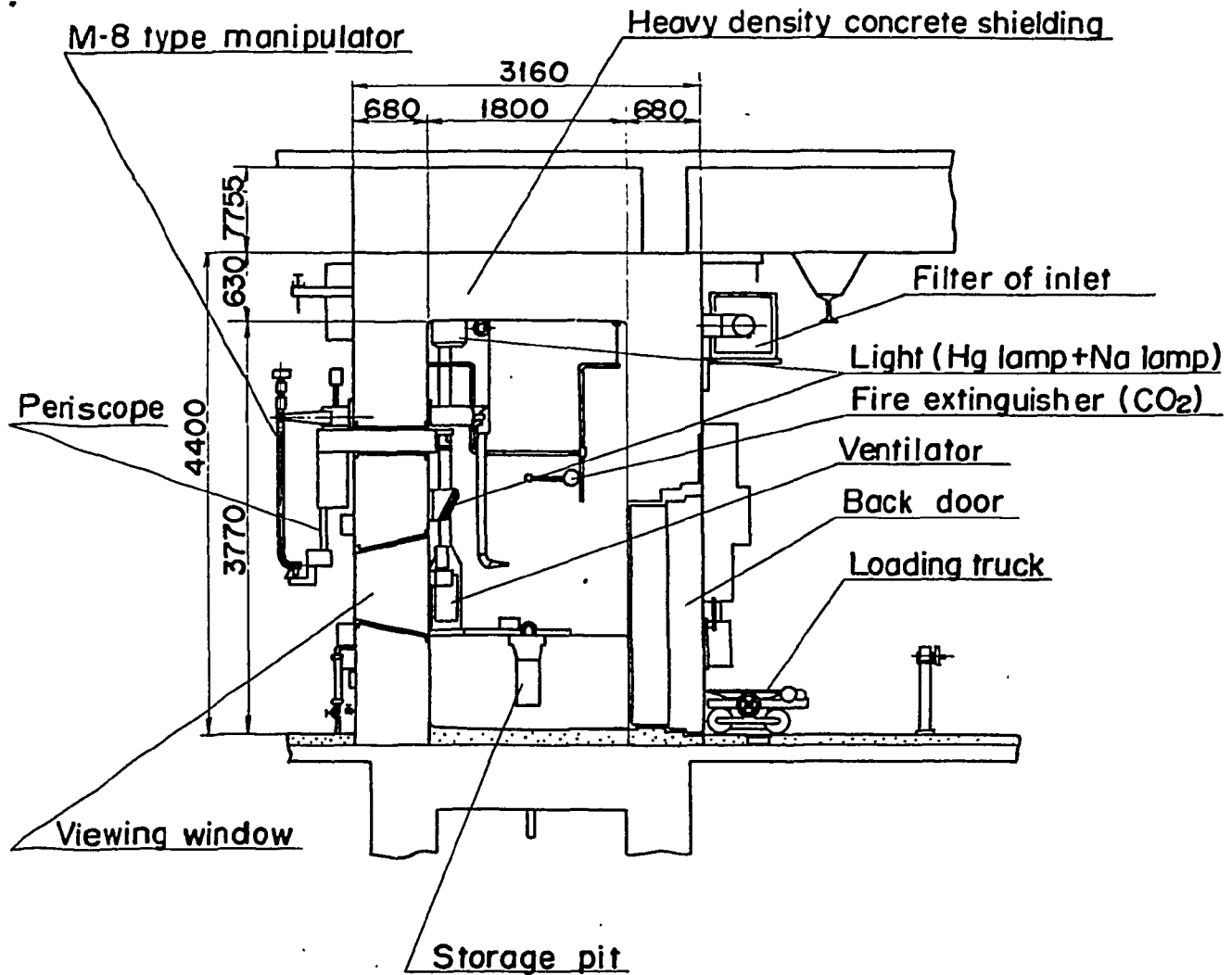


Fig. 3 Cross - section of the cave for the production of radiation sources.

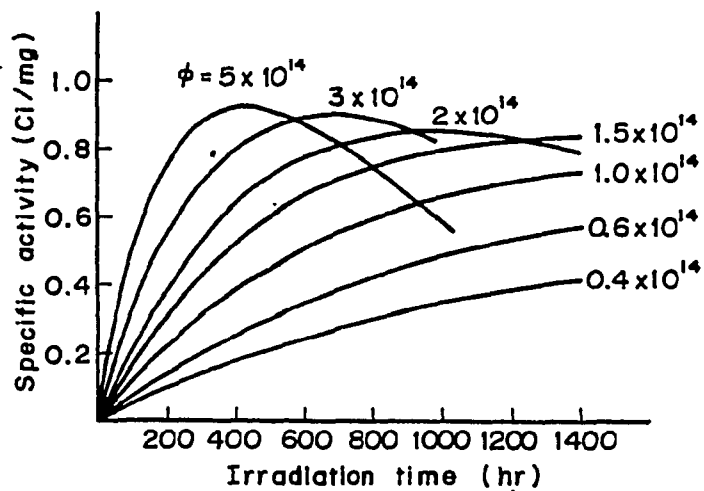
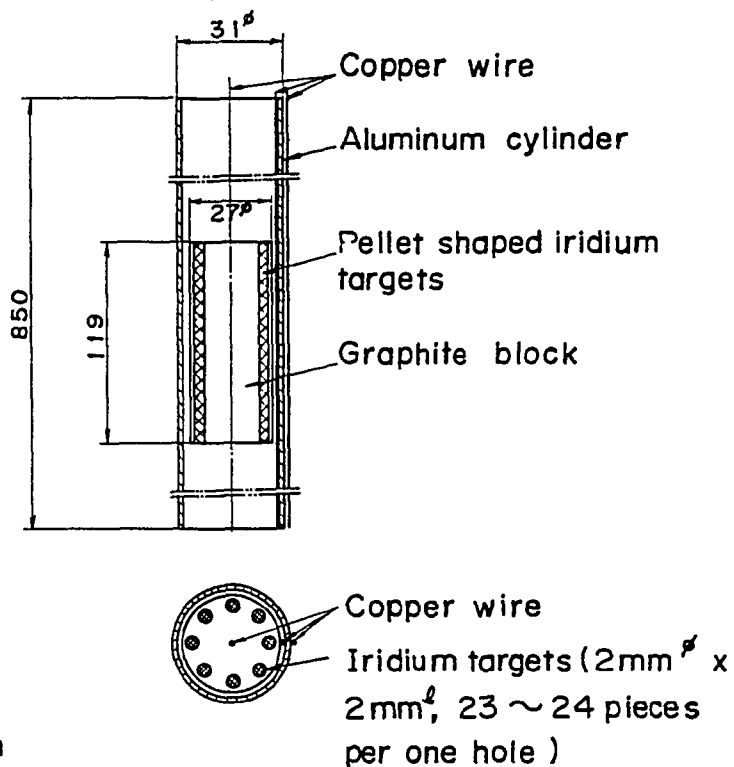


Fig.4 Specific activity of ^{192}Ir produced as a function of the irradiation time for various values of the thermal neutron fluxes (ϕ)



Unit : mm

Fig. 5 Target assembly for measuring neutron perturbation by iridium targets

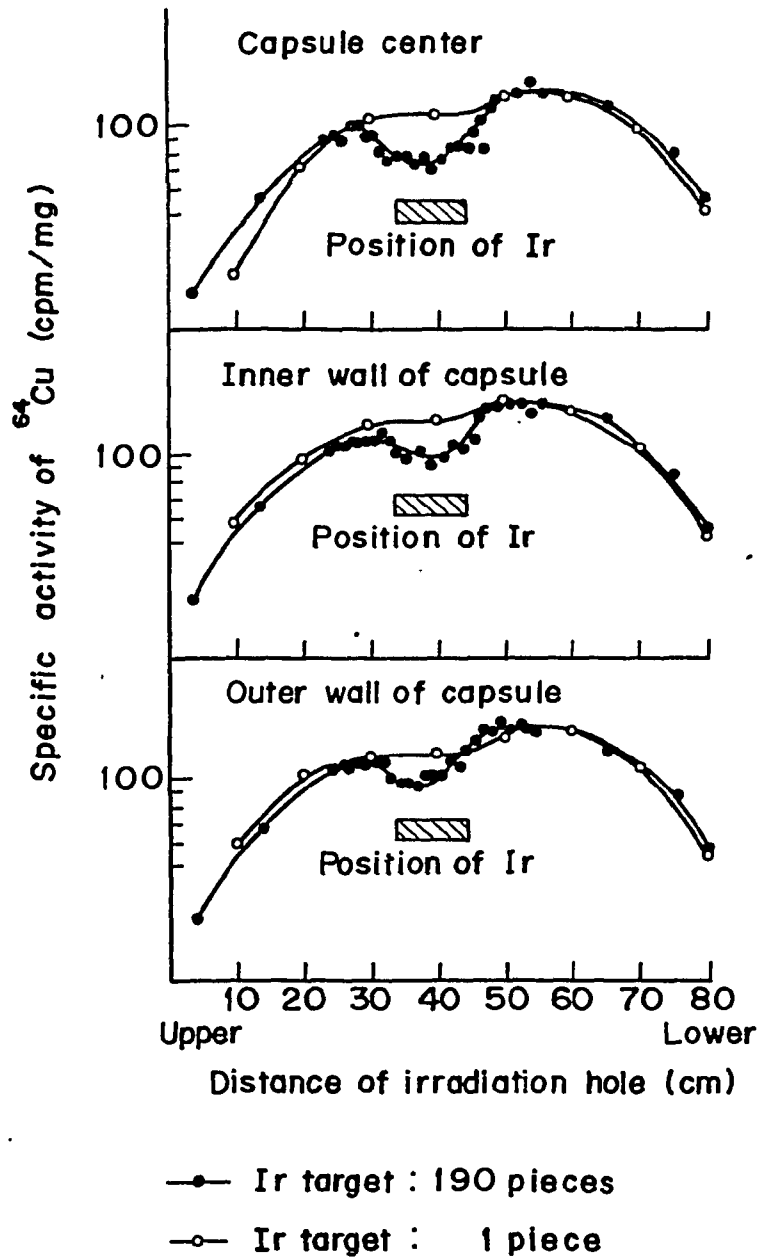


Fig. 6 Vertical distribution of thermal neutron flux by neutron perturbation of iridium targets

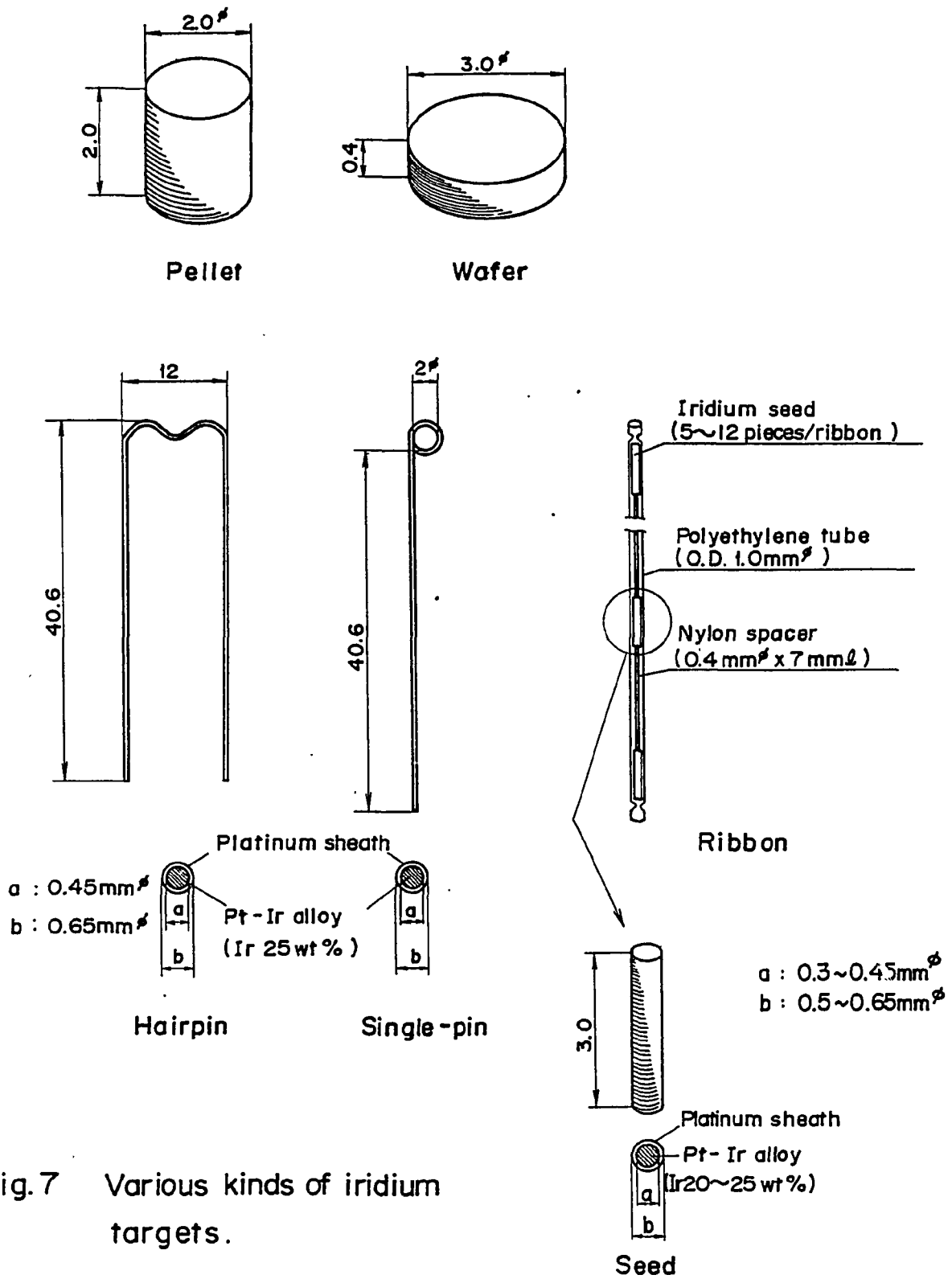


Fig. 7 Various kinds of iridium targets.

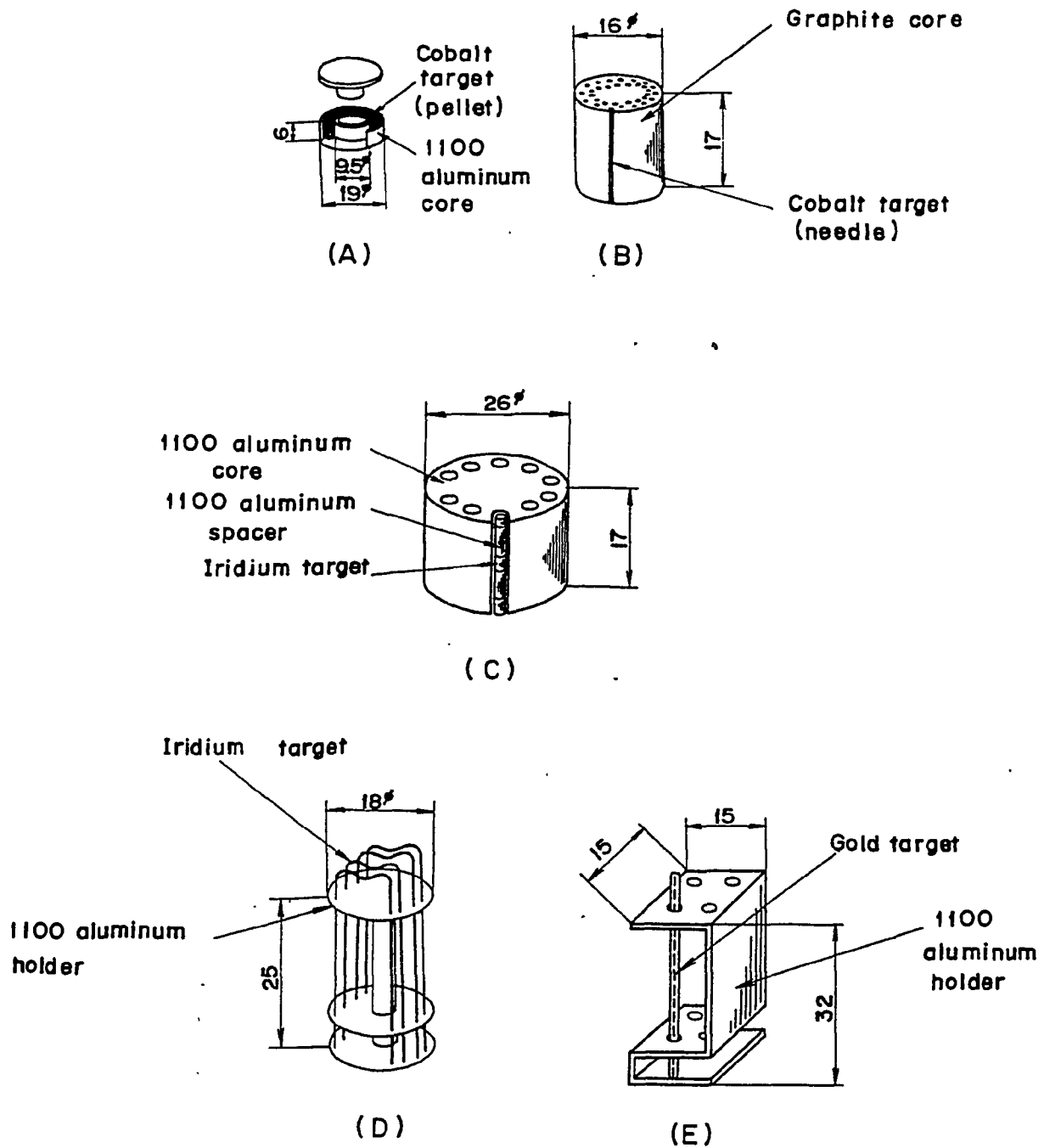
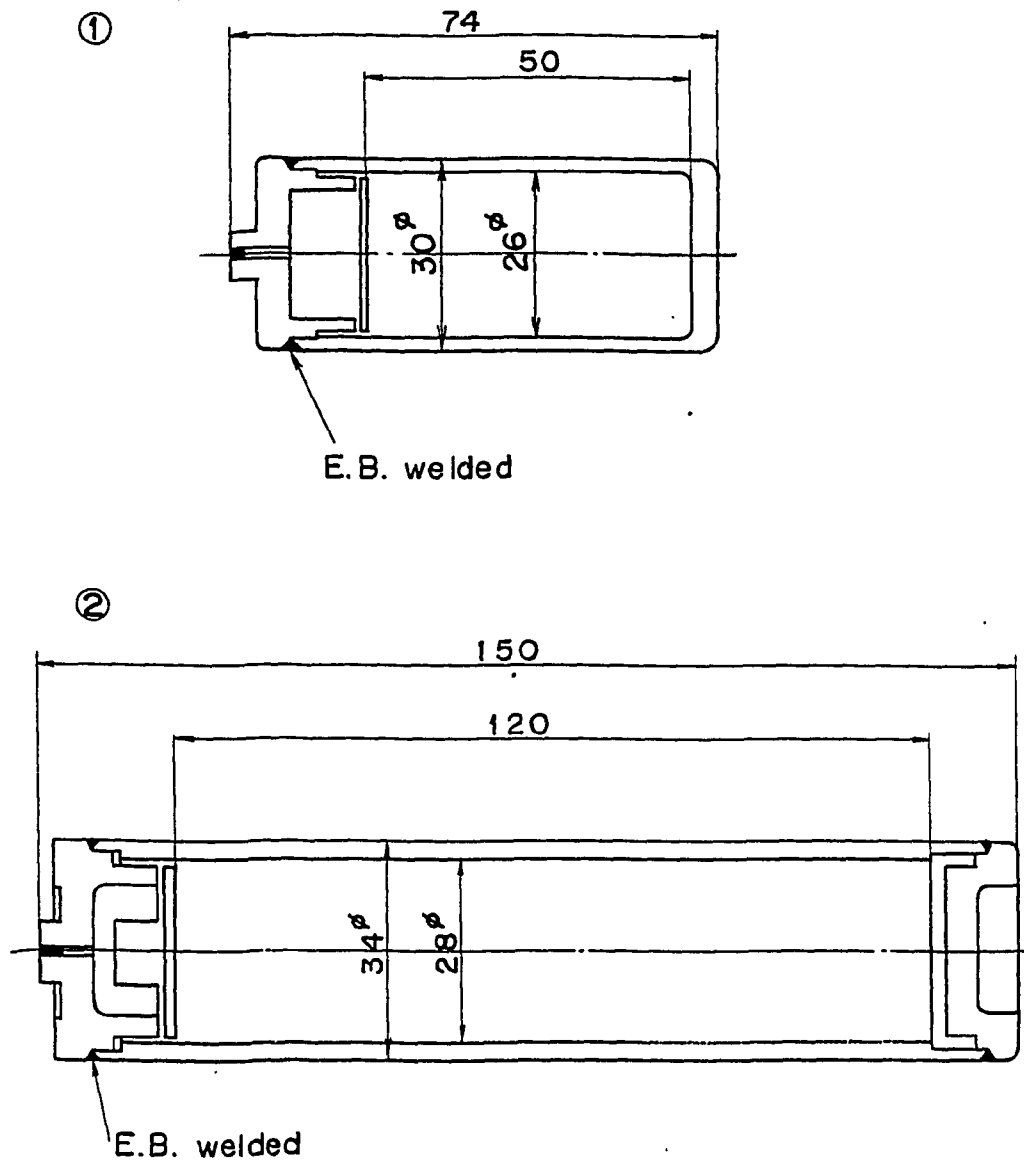


Fig.8 Target designs for neutron irradiation



All material : 1100 aluminum

Fig.9 Irradiation capsule

- ① For iridium targets at JRR-2(Aw-3011type)
- ② For iridium and thulium targets at JMTR

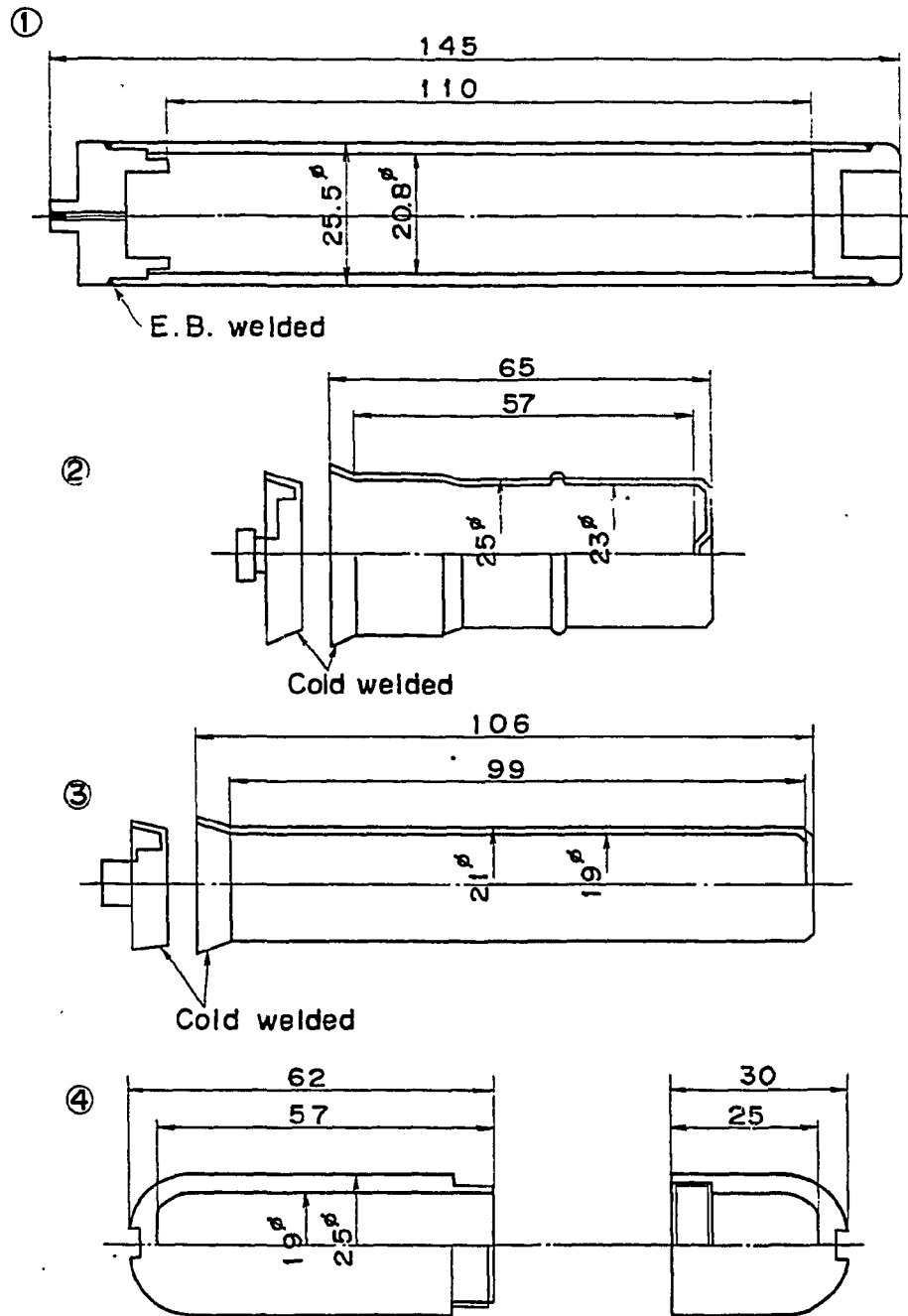


Fig.10 Irradiation capsule

- ① For pellet and wafer shaped cobalt targets at JMTR (1100 aluminum)
- ② For gold targets at JRR-2 or JRR-4 (1100 aluminum)
- ③ For needle shaped cobalt target at JMTR (1100 aluminum)
- ④ For gold and iridium (radiotherapy) targets at JRR-3 (Polyimide resins)

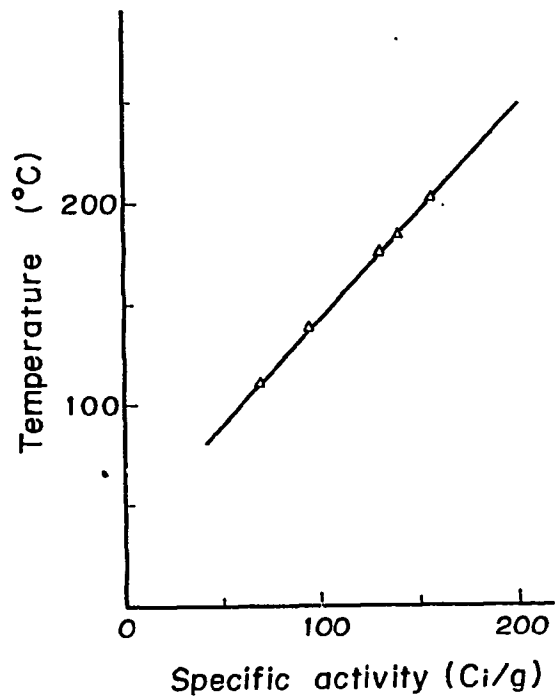
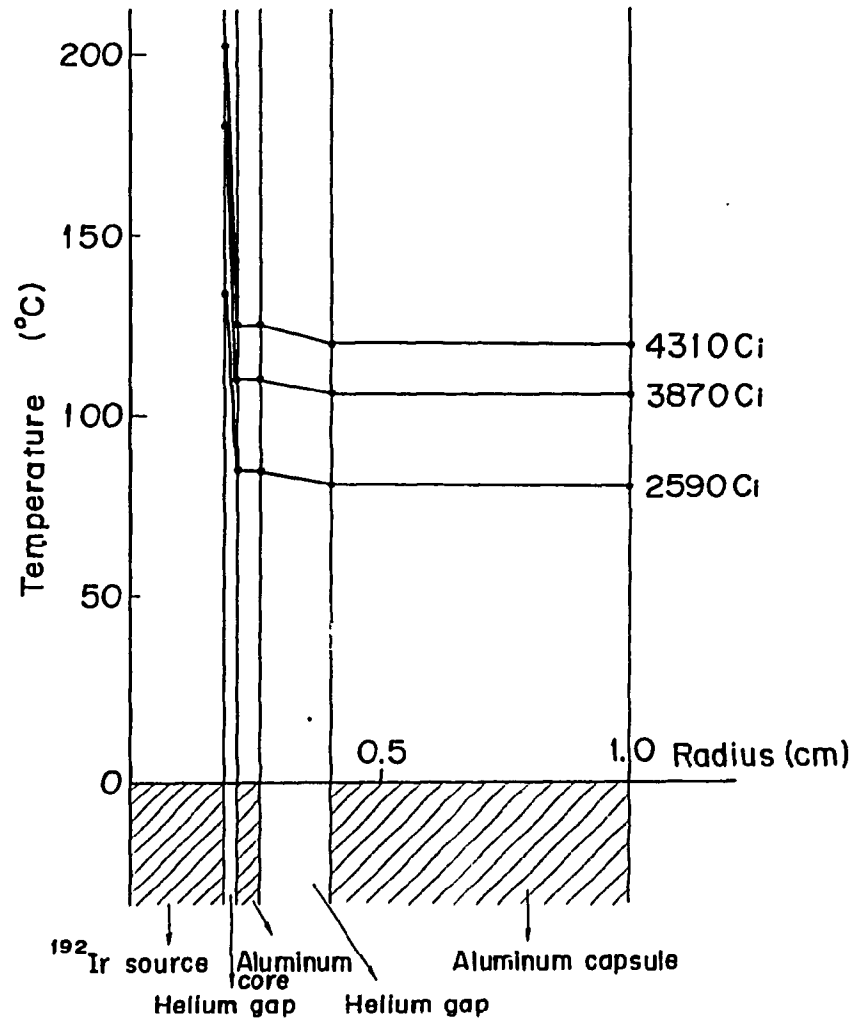


Fig. 11 Surface temperature of a ^{192}Ir source as a function of specific activity



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Fig. 12 Temperature distribution in the irradiation capsule

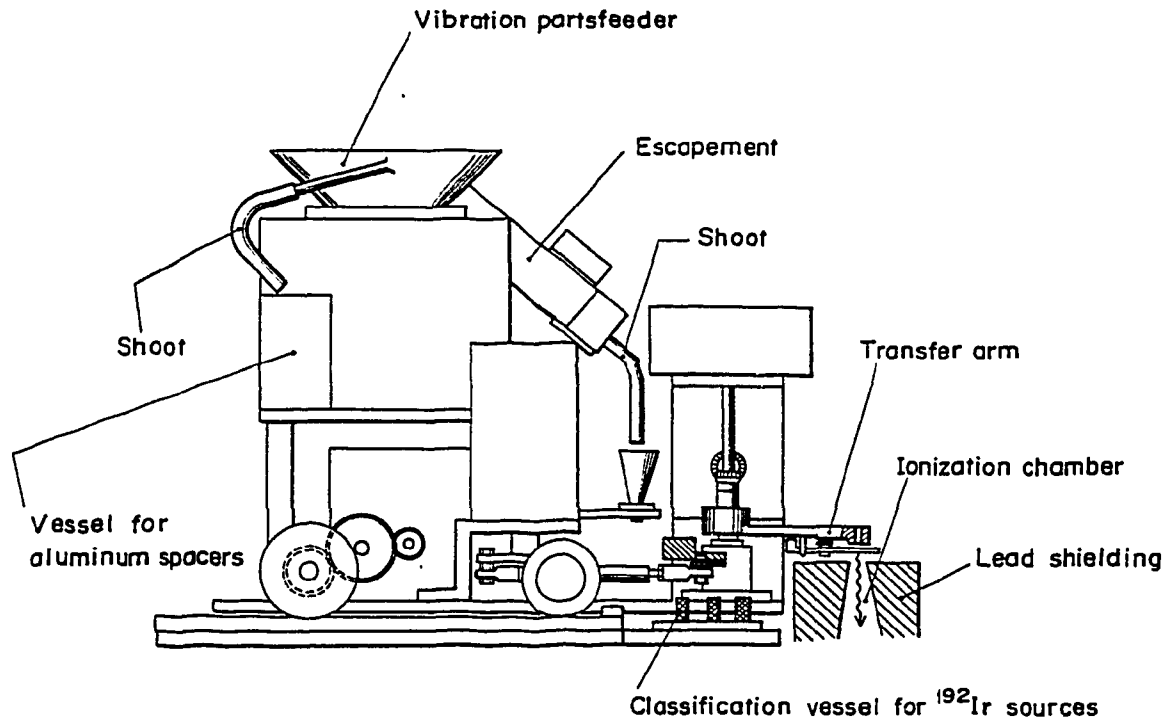


Fig.13 Conceptional design of the automatic apparatus

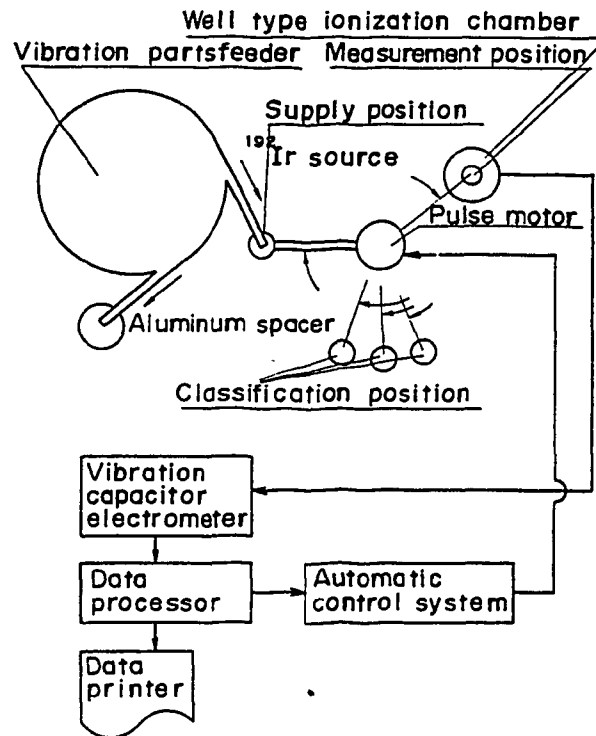


Fig. 14 Fundamentals of the remote handling system

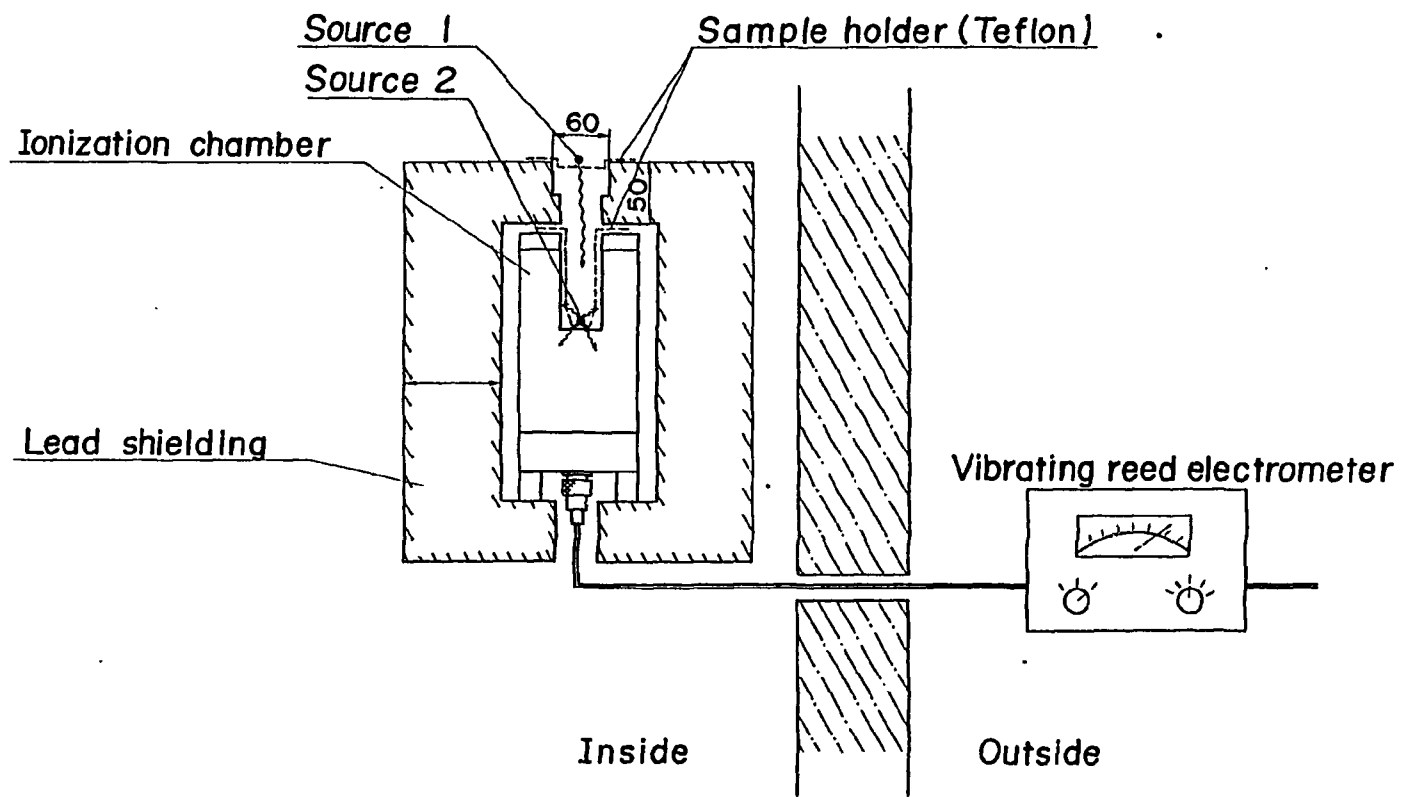


Fig. 15 Measuring system for ^{192}Ir and ^{60}Co

Source 1 : Measurement position of ^{192}Ir

Source 2 : Measurement position of needle shaped ^{60}Co

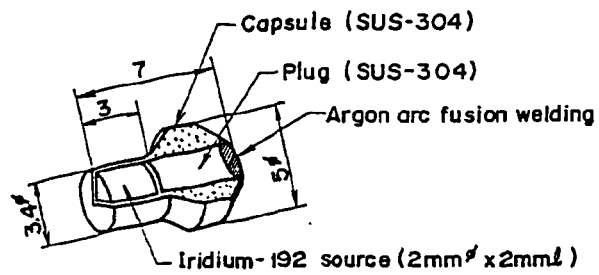
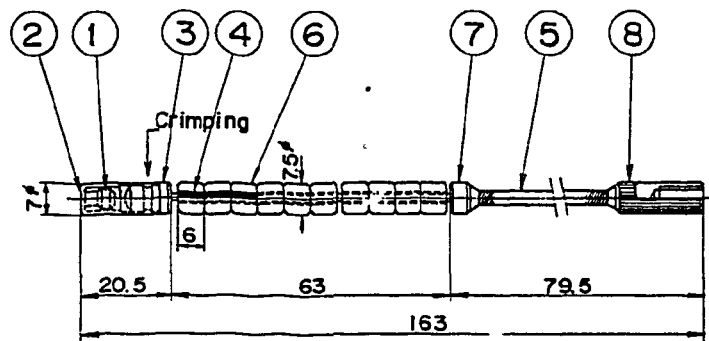


Fig.16 JAERI-Type ^{192}Ir sealed capsule



- 1 Encapsulated ^{192}Ir source
2. Cap for enclosing source. (SUS-304)
3. Fitting (SUS-304)
4. Flexible cable.
5. Flexible cable.
6. Shielding (tungsten alloy)
7. Stopper (SUS-304)
8. Screw joint (SUS-304)

Fig.17 Source holder for radiography irradiator

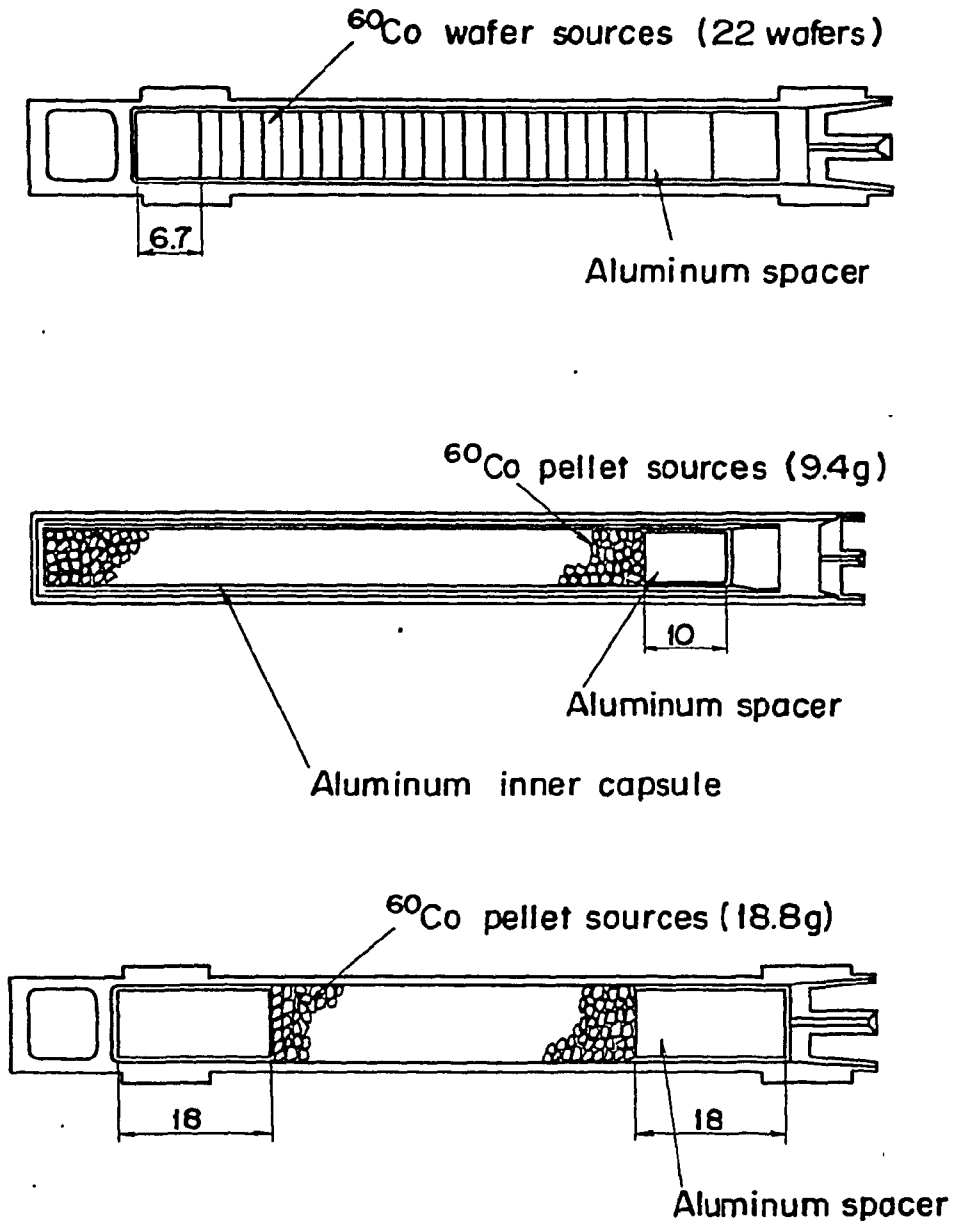


Fig.18 Examples of ^{60}Co sealed sources
(Outer capsule material : SUS-316L)

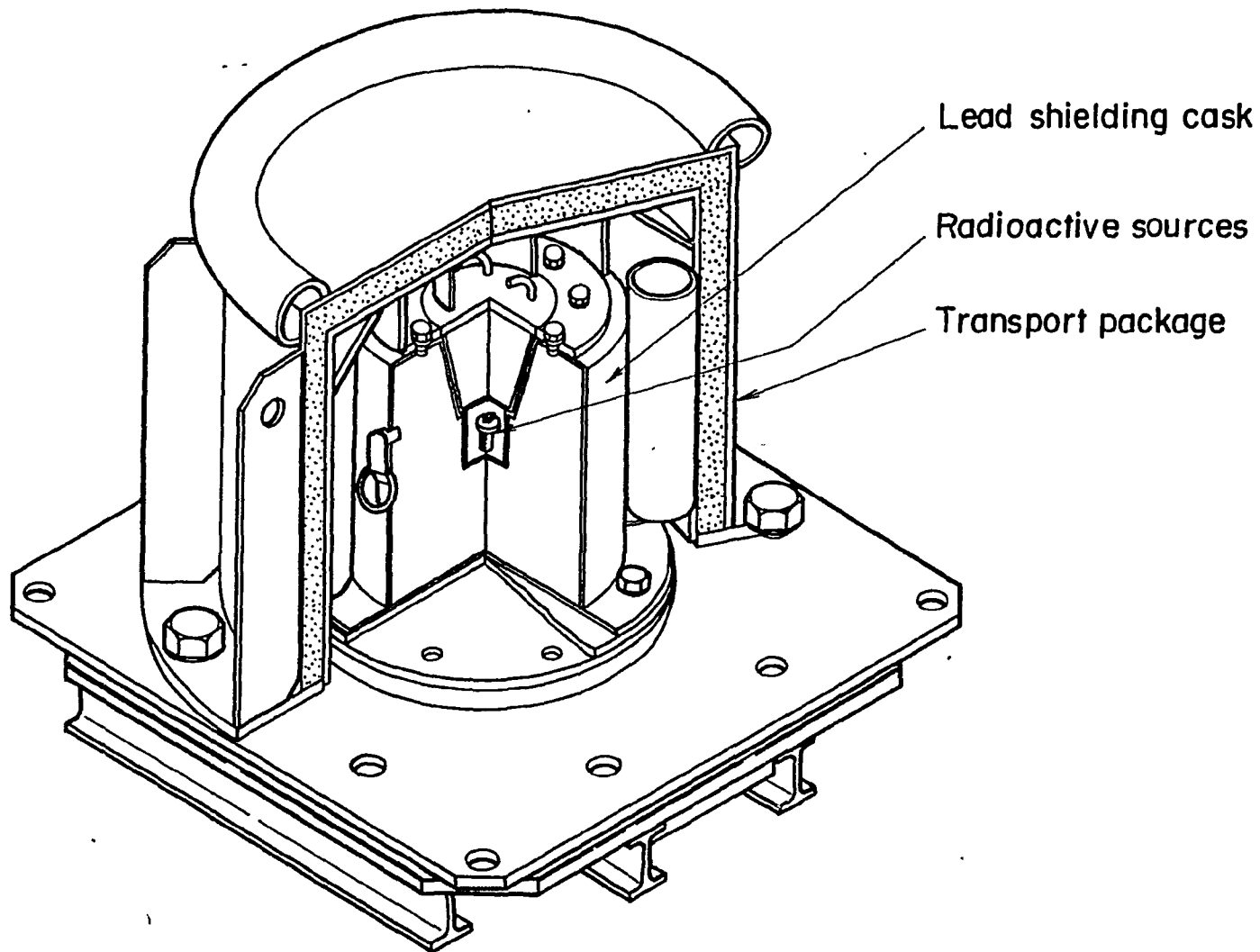


Fig.19 Transport container for large radioactive sources

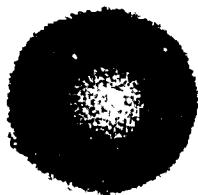
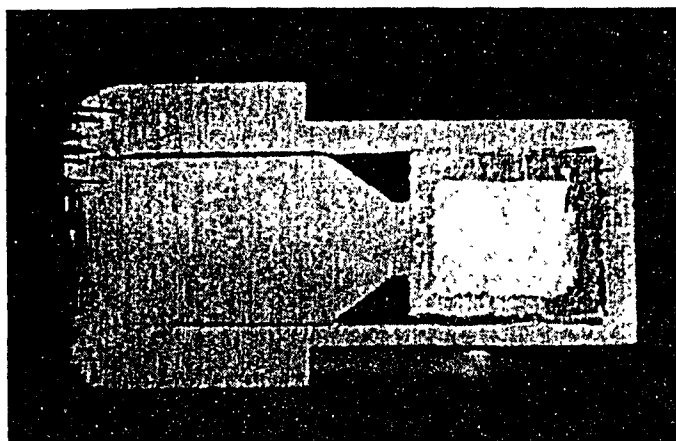


Photo. 1 Autoradiograph of pellet
shaped ($2\text{mm}^{\phi} \times 2\text{mm}$) ^{192}Ir



1 mm

Photo. 2 ^{170}Tm sealed source

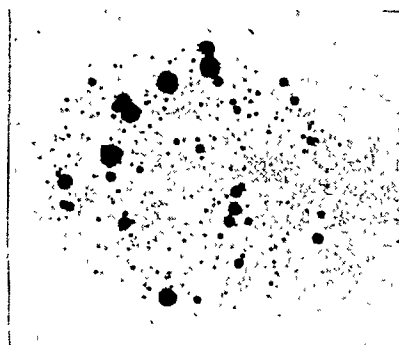


Photo. 3 Radiograph of contamination
spots excoriated from the
surface of ^{192}Ir sources