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TARGETS FOR PRODUCTION OF I-123 AT HIGH  
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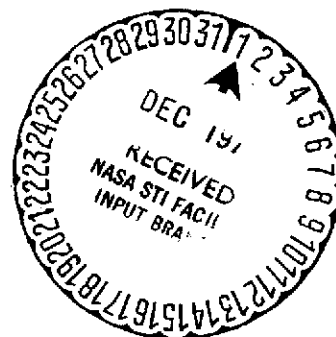
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SELF-VAPOR COOLED TARGETS FOR PRODUCTION  
OF  $^{123}\text{I}$  AT HIGH CURRENT ACCELERATORS

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# SELF-VAPOR COOLED TARGETS FOR PRODUCTION OF $^{123}\text{I}$ AT HIGH CURRENT ACCELERATORS

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## Introduction

The Bureau of Radiological Health is soon likely to take a position in which they discourage the use of  $^{131}\text{I}$  for thyroid studies because of an unacceptable risk that the radiation dose to the thyroid in such procedures may induce cancer.  $^{99\text{m}}\text{Tc}$ -pertechnetate can be substituted for  $^{131}\text{I}$  if the clinician is willing to forego the physiologic behavior of iodine. There is a radioiodine,  $^{123}\text{I}$ , that has long been recognized (Ref. 1 ) as an acceptable agent.

The first production problem with  $^{123}\text{I}$  was the purity, because the early cyclotron product contained significant amounts of other radioiodines. A means of production of  $^{123}\text{I}$  was found (Ref. 2 ) that gave a pure product, but it was clear that a sufficient quantity could not be made with conventional cyclotrons to provide for routine thyroid studies. The hope of increased quantities of  $^{123}\text{I}$  came when spallation reactions using the high current accelerators were planned (Refs. 3 , 4 ). These spallation targets use the high purity method (Ref. 2 ) in which  $^{123}\text{Xe}$  is the isotope produced and separated from radioactive contaminants and then allowed to decay into  $^{123}\text{I}$ . Los Alamos Scientific Laboratory and Brookhaven National Laboratory are both developing the spallation reaction to produce  $^{123}\text{I}$  at their facilities designated LAMPF and BLIP, respectively.

$^{123}\text{I}$  must also be produced with low unit cost if it is to replace  $^{131}\text{I}$  or be used instead of  $^{99\text{m}}\text{TcO}_4^-$ . This requires that a target must be capable of long hours of operation and that the  $^{123}\text{Xe}$  must be available

on a continuous basis with a minimum of human intervention. The spallation target used at BLIP requires hot cell chemical techniques to remove the  $^{123}\text{Xe}$ . The LAMPF target uses a heating cycle to remove the  $^{123}\text{Xe}$  which has been produced and stored in the target. This method has been shown (Ref. 5 ) to reduce the yield by 20 to 50 percent, depending on the cycle timing, due to the decay of  $^{123}\text{Xe}$  to  $^{123}\text{I}$  in the target prior to removal.

A new concept in targetry is herein proposed which will provide  $^{123}\text{Xe}$  continuously as it is produced and will also provide a means for removal of the beam heating. The novelty of the method lies in the use of the target material as a working fluid in a heat transfer system. The nuclei of the working fluid undergo a spallation reaction to produce  $^{123}\text{Xe}$  and the target material is transformed from the liquid to the vapor phase by the beam heating. The vapor travels from the target volume to a condenser section and then the fluid returns to the target volume. The  $^{123}\text{Xe}$  produced in the target volume is non-condensable and is swept from the target where it is collected and decayed into  $^{123}\text{I}$ .

### Vapor Cooled Target System

Figure 1 shows the basic elements of a target system. The high energy protons are incident upon a liquid in the target volume ( 1 ) and cause nuclear reactions of which  $^{123}\text{Xe}$  is one of the products. A means ( 2 ) may be provided to prevent surging of the liquid up the vapor tube ( 3 ) to a region ( 4 ) in which condensation takes place. A heater ( 5 ) may be required to initially heat the vapor tube so that the vapor will not freeze in a cool region and thereby empty the target volume. A means ( 6 ) of injecting helium at a low rate, controlled to maintain some set pressure at the top of the column, is required. The helium transports the  $^{123}\text{Xe}$  through a low temperature trap at  $-80^{\circ}\text{C}$  ( 7 ) to remove radioiodine impurities and to a collection trap ( 8 ) operated at liquid nitrogen temperature. This gas handling and purification system is the same as that used with the conventional cyclotron generator (Ref. 5 ). An equally efficient removal of radioiodines from the gas stream has been achieved (Ref. 6) with hot silver. The gas passing through the liquid nitrogen trap

should be free of radioactivity. Therefore, a radioactive monitor (9) will provide an indication of system malfunction. A pump (10) will be required if the helium pressure is maintained below atmospheric pressure. A bubbler (11) is useful as an indicator of the flow in the system.

The transport of heat by means of vaporization and subsequent condensation is a technique that has been used for hundreds of years. In 1963 Groover et al (Ref. 7) reported on novel devices in which the condenser end was physically located below the evaporation region so that gravity could not be used to return the liquid. The liquid was returned by a wicking action in a screen mesh wall. To these devices, he gave the name heat pipe.

The proposed high current targets are a liquid target, vapor cooled device that could be operated as a heat pipe or as a conventional condenser. It is necessary to operate the system with two components, one, the working fluid that exists in both liquid and vapor phases and the other, a non-condensable gas which can be used to flush the column of the  $^{123}\text{Xe}$  produced in the nuclear reaction.

Two component heat pipes have not been as extensively studied as the single component type. Barsch (Ref. 8) has given a summary of the literature of two component devices. From his report it is clear that the theory has not progressed to the point of solving the eight coupled, nonlinear, second-order partial differential equations which describe the general system. For the design of the vapor cooled targets, a simplified model was developed in which energy balance, mass flow conservation, and an assumption of thermal equilibrium are used.

The following calculation of a two component device is given to point out the important features of the operation. Figure 2 shows the model and the governing equations. The specific assumptions are:

1. The liquid is assumed to be at a uniform temperature due to a beam heating rate =  $I\Delta E$  where  $I$  is the beam current and  $\Delta E$  the proton energy loss.

2. The vapor pressure immediately above the fluid is the equilibrium vapor pressure.
3. The partial pressure of helium immediately above the fluid is zero.
4. The velocity of the vapor escaping the liquid has a constant value,  $U$ , in both radial and axial dimensions.
5. Heat is lost by radiation from the wall of the tube.

The calculation starts by assuming a liquid temperature  $T_0$ . For an increment of tube height, immediately above the liquid, the radiated power is calculated. This power loss corresponds to a vapor loss  $\Delta m$  which is related to a vapor density decrease  $\Delta \rho$ . The vapor density increment is then related, by means of the vapor pressure vs temperature relationship, to a temperature lapse  $\Delta T$ . The temperature of the next increment of tube height  $T_0$  is  $(T_0 - \Delta T)$  and this temperature is used to calculate the radiated power from that element. Following this procedure stepwise up the tube gives the vapor density, vapor pressure and temperature as a function of height. The density distribution is then used to calculate the potential energy which balances the dynamic pressure, i. e.,

$$g \int_0^H Z d\rho = \rho_0 \frac{U^2}{2}$$

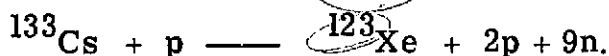
Trial and error selection of  $T_0$  to find the density distribution which satisfies the equality completes the calculation.

Figure 3 shows the distributions calculated for a 100 watt input to a 1.90 cm diameter, cesium filled, vapor tube radiating heat as a black body;  $P^0_{He}$  was taken to be 215 torr. The temperature is seen to decrease rapidly at a height of 9.0 cm. This behavior is characteristic of two component columns when one of the components is noncondensable.

The stability of the operation of a column is apparent, since an increase in the input power need only cause a small temperature increase of the liquid in order to cause a significant change in the height of the hot zone and therefore the amount of radiated power.

## Target Fluids

The choice of the target fluid is dependent upon the specific nuclear reaction chosen to produce  $^{123}\text{Xe}$ . The nuclear reaction is dictated by the type and energy of the bombarding particle. High current accelerators at present accelerate protons and the specific reactions considered for the vapor cooled targets were:



*Xe-133*  
*or I-123-125*

The reaction using  $^{127}\text{I}$  was studied by Fusco et al (Ref. 9) and was shown to have a significant yield for bombarding energies from 47 to 63 MeV. The reaction with  $^{133}\text{Cs}$  to produce  $^{123}\text{Xe}$  has not been previously reported and was therefore included in this investigation.

A 12.06 gm sample of  $\text{CsCO}_3$  was compacted into 33 mm diameter container and covered (gas tight) with a 0.025 mm aluminum foil. The sample presented a uniform thickness of  $1.15 \pm 0.1 \text{ gm/cm}^2$  of Cs to the beam of 575 MeV protons from the NASA-SREL cyclotron. The beam was focused so that all of the beam was incident upon the sample. Aluminum monitor foils in front of and behind the target were counted to determine the amount of  $^{24}\text{Na}$  produced in the irradiation. Using a cross section of 10.7 mb for this reaction, the number of protons striking the target was determined to be  $1.35 \times 10^{14}$ . The amount of  $^{123}\text{Xe}$  produced in the irradiation was determined by counting the intensity of the 149 keV and 178 keV gamma rays of  $^{123}\text{Xe}$  and correcting with a 2.10 hr. half-life back to the bombarding time. The gamma ray yields per disintegration were taken (Ref. 10) to be 49.9% and 16.0% respectively. The production cross for  $^{123}\text{Xe}$  bombarded with 575 MeV protons was determined to be  $14.3 \text{ mb} \pm 2.4 \text{ mb}$ .

Liquid cesium has been extensively studied as the working fluid of heat transfer devices. Consequently the viscosity, thermal conductivity, and electrical conductivity have been measured. Iodine has not been so extensively studied. Therefore, a column was constructed of a pyrex

tube of 15 mm. ID. The column was filled with 50 gms. of iodine and mounted in a vertical orientation with the electrically heated evaporator below the condenser. The air was evacuated and then the tube filled to a pressure of 0.5 atmospheres with helium. At an input power of 100 watts, the iodine melted and boiled with some "bumping." An adiabatic section, 15 cm. in height above the evaporator, was made by insulating the wall of the tube. This adiabatic section provided a region in which the liquid slugs generated by bumping could surge. Consequently, the condenser section, above the adiabatic region, saw a smooth vapor flow. The condenser was cooled by the natural convection of air rising over the outside wall of the tube.

The vapor was observed to condense to a liquid to a height about 15 cm. above the adiabatic section. Above this height the iodine vapor collected as a solid. The sharp line of liquid and solid was clearly visible. The iodine was so dark that it was not possible to see how much iodine was present at any height. Therefore, a radioactive source of  $^{241}\text{Am}$ , with 60 keV gamma rays, was collimated to a 3 mm. beam and the attenuation of this beam in passing through the column was measured as a function of height.

Figure 4 shows the gamma ray counting rate as a function of vertical position for three times after the application of power to the evaporator. The runs at 1 1/2 and 2 3/4 hours both show a region of strong attenuation (low counting rate) for about a 1 cm. region just above the liquid-solid interface. The conclusion drawn from this measurement is that the amount of solid reaches an equilibrium at long times. Thus, if the target volume has sufficient iodine at the beginning of the "beam on" period, the target volume will not become exhausted due to vapor collecting as a solid in a cool region. As a practical matter however, the entire column can be operated at a temperature above the melting point and then the problem of exhausting the target volume disappears.

### Target Designs - TRIUMF

The 500 MeV accelerator, built by a consortium of western Canadian universities is expected to produce beam currents of 100 microamperes. A number of beam lines are to be developed but in the early period of operation there will be two lines. One is a beam of protons for meson production. This is accomplished when 10 microamperes of protons strikes a beryllium target. The beryllium target is located in a shield assembly designated T-2 and shown in Figure 5. Behind the beryllium target a water cooled copper block is positioned to stop the protons.

For  $^{123}\text{I}$  production at TRIUMF, it has been decided to use cesium as the target material. The cesium target will be placed immediately in front of the copper block. The vapor tube could have come horizontally and to the rear or vertically through the steel plug shield. The latter arrangement was chosen because of possible interference with meson beam magnets at the rear of T-2.

The target details are shown in Figure 6. The liquid cesium is held in a 5.0 cm diameter by 5.0 cm long cylindrical chamber from which a 2.0 cm diameter vapor tube rises. The vapor tube rises at an angle to the vertical in order to clear the meson beam and to reach a vertical hole already present in the shield plug. The vapor tube is covered on the outside with a low emissivity surface so that the heat radiated to the outer tube is small. The target chamber and vapor tube is contained within a second tube so that a target failure would be contained. The cesium vapor is condensed by water cooling the vapor tube above the top plate of the shield assembly. The mechanical design is such as to allow the target and vapor tube to be installed from below, through an existing 3.5 cm diameter hole in the top plate. Ten microamperes of beam loses 50 MeV in the liquid cesium gives a heating of the target at a rate of about 500 watts. Collecting all of the  $^{123}\text{Xe}$  produced and allowing it to decay into  $^{123}\text{I}$  will give a yield of 200 mCi per hour. The temperature of the liquid cesium will rise to  $600^{\circ}\text{C}$ . The vapor will also be about  $600^{\circ}\text{C}$  all the way to the condenser since very little power (about 50 watts) is radiated from the vapor tube. A small flow ( $1\text{ cm}^3$  per min) of clean dry helium is injected



in the top of the vapor tube to provide a means of sweeping the  $^{123}\text{Xe}$  from the target. The purification of the  $^{123}\text{Xe}$  by a trap at  $-80^{\circ}\text{C}$  and its collection at liquid nitrogen temperature is similar to that shown in Figure 1. The vacuum space surrounding the vapor tube will be pumped by a liquid nitrogen absorption pump. In this way the system is contained and the containment can be monitored by checking the radioactivity in the sorption pump.

#### Target Design-BLIP (Figure 7)

A target using liquid iodine as the target fluid has been preliminarily designed for the Brookhaven Linac Isotope Producer (BLIP). The design takes advantage of the fact that the irradiations at BLIP are done in a deep pool of water. The liquid iodine is contained in a flat circular container of stainless steel with a thickness of iodine of 1.0 cm. 65 MeV protons lose about 20 MeV of energy in the iodine before entering a water cooled, stainless clad, copper block. The high current beam pulses from the linac come every 0.1 sec and will produce a flash boiling and the liquid will surge upwards. Before the next pulse, more fluid will fill the beam space. The vapor produced will travel up the tube, condense on the walls as a liquid, and return to the target area. The walls of the vapor tube will be kept above the melting point, with an electrical heater, so that iodine cannot freeze in a cool region. Helium gas fills the volume between the vapor tube and the outer containment tube and serves to conduct the heat from the vapor tube to the outer wall. This surface is cooled by the water in the shielding tank. The heat flux is low, but the conducting surface area is large because of the long length of the vapor tube. Fifty microamperes of 65 MeV protons should yield 200 mCi per hour of  $^{123}\text{I}$ .

#### Conclusion

The preliminary design of two self-vapor cooled, liquid targets has been given. One uses cesium and is appropriate for accelerators with a proton beam of 400 to 500 MeV. This target is planned for TRIUMF and would be appropriate for SIN and LAMPF. The other, uses elemental iodine and is appropriate for accelerators where the beam energy is 60 to 70 MeV. Both targets produce a continuous yield of  $^{123}\text{Xe}$  which is collected,

remote from the irradiation area, by freezing at liquid nitrogen temperature.  $^{123}\text{I}$ , produced by the decay of  $^{123}\text{Xe}$  is free of radiocontaminants except  $^{125}\text{I}$ , the daughter of  $^{125}\text{Xe}$ , and this contaminant can be kept to the level of a few percent if the time allowed for  $^{123}\text{Xe}$  to decay is made short.  $^{125}\text{I}$  does not degrade the quality of the scans because the emitted gamma ray energy is low energy. An advantage of  $^{123}\text{I}$  produced from  $^{123}\text{Xe}$ , in addition to the purity, is the possibility of hot atom labeling of radiopharmaceuticals by chemically reacting the nascent iodine atom which is formed when the  $^{123}\text{Xe}$  atom decays.

Other possible applications of the self-cooled, liquid target can be envisioned. An example would be the production of  $^{127}\text{Xe}$  from a cesium target with protons of about 70 MeV.

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### VAPOR COOLED TARGET SYSTEM

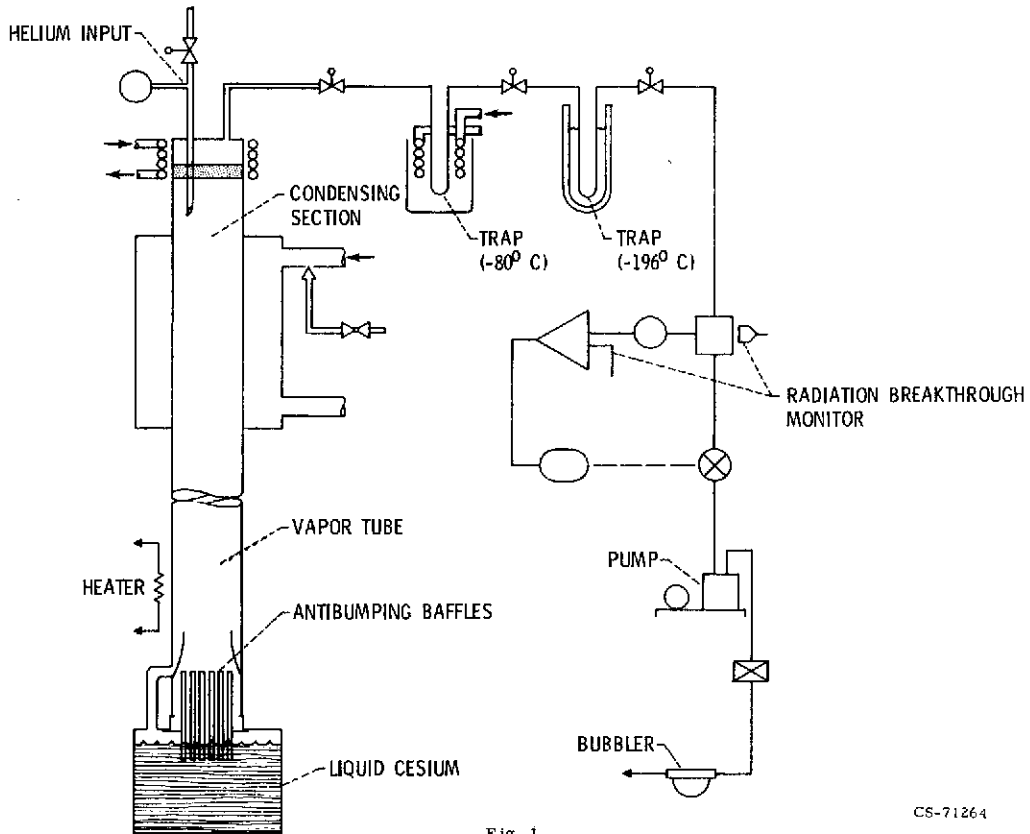
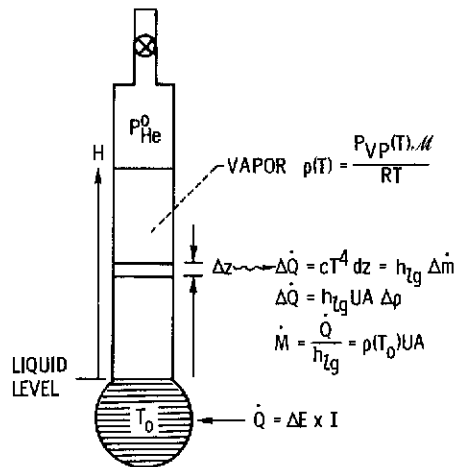


Fig. 1

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### SIMPLIFIED TWO COMPONENT COLUMN



$T_0$	LIQUID TEMP	$U$	VAPOR VELOCITY
$\Delta E$	PROTON ENERGY LOSS IN LIQUID	$g$	GRAVITATIONAL CONSTANT
$I$	PROTON CURRENT	$c$	A CONSTANT
$\rho(T)$	VAPOR DENSITY AT T	$P_{He}^0$	HELIUM PRESSURE AT TOP OF COLUMN
$R$	GAS CONSTANT	$H$	MAX HEIGHT OF VAPOR
$M$	MOLECULAR WEIGHT OF VAPOR	$A$	AREA OF COLUMN
$h_{tg}$	HEAT OF VAPORIZATION		

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Fig. 2

CALCULATED TEMPERATURE AND DENSITY DISTRIBUTIONS

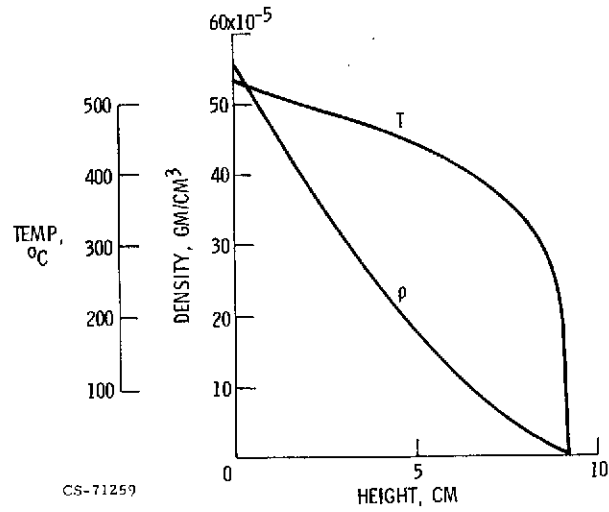


Fig. 3

CURVES SHOWING 60 keV GAMMA ATTENUATION IN IODINE COLUMN

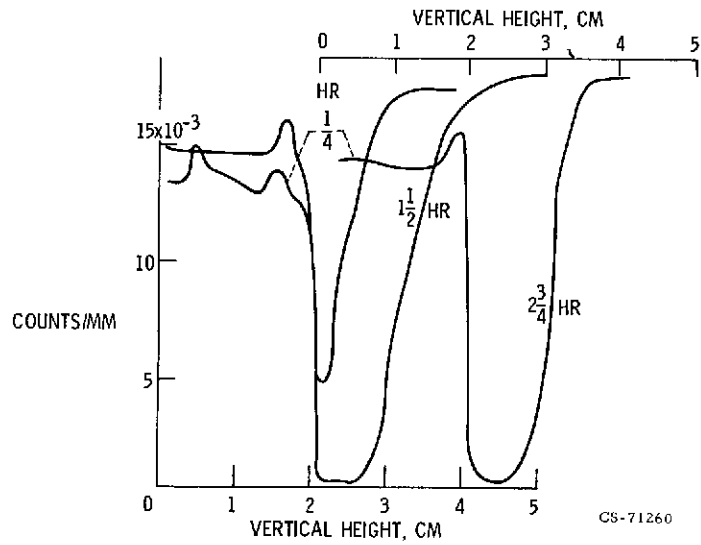


Fig. 4

### TRIUMF MESON TARGET AREA T-2

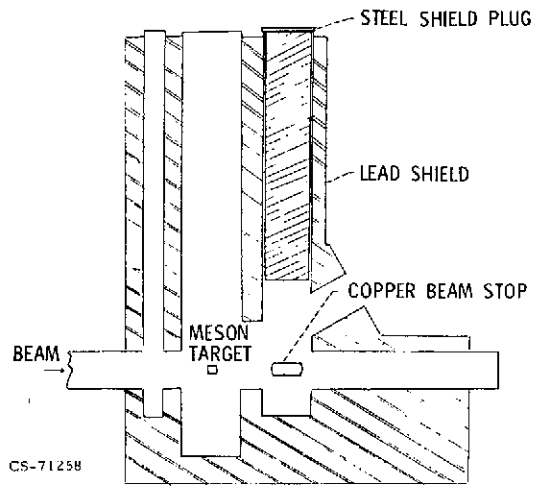


Fig. 5

### TRIUMF TARGET

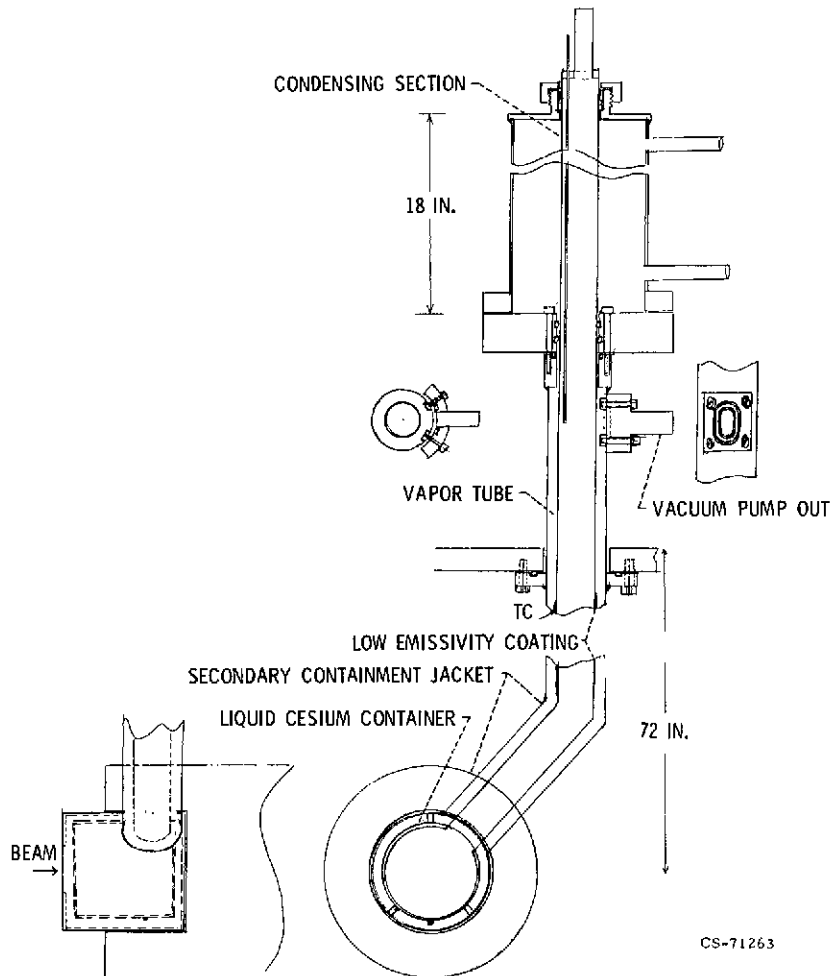


Fig. 6

# BLIP TARGET

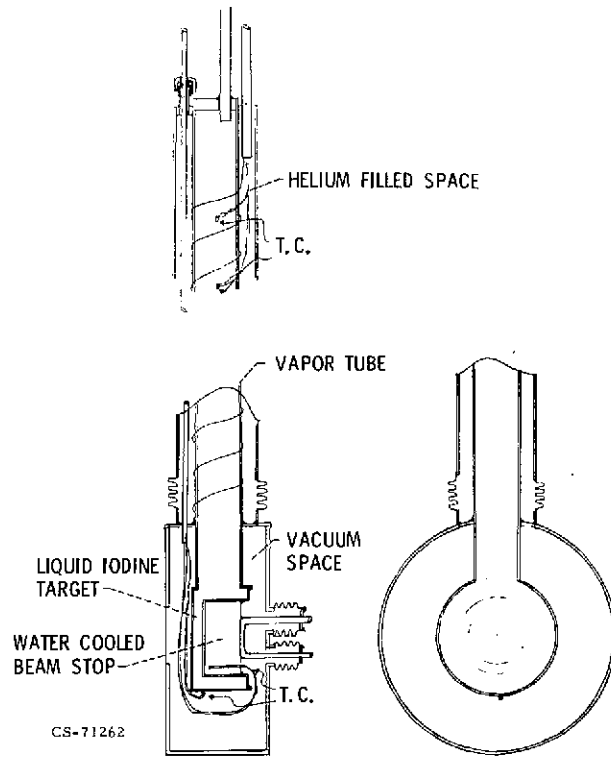


Fig. 7