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ABSTRACT

A pulsed neutron source has been developed whose principal element is a sealed-off vacuum tube in which a discharge between titanium tritide surfaces produces tritons, which are then accelerated to a deuterium-loaded target. Described is construction of a tube producing approximately 10^7 neutrons in microsecond pulses and having a life of several thousand pulses.

Introduction

High-intensity pulsed neutron sources have many applications in nuclear physics, in the study of reactors, and in geophysical exploration. For the calibration of cloud and bubble chambers as well as nuclear emulsions, pulsed sources provide neutrons of known energy. In reactor design (References 1 through 6), neutron diffusion and multiplication parameters can be determined from the decay of neutrons from a pulsed source. Important constants of a reactor in a subcritical state may also be determined. In some cases a considerably smaller amount of material is required than if conventional critical experimental techniques are used. For geophysical problems (Reference 7), such as oil-well logging, a compact source is desirable when both neutron activation and neutron moderation techniques are used. Because of its general utility, therefore, the development of a compact and self-contained pulsed neutron source was undertaken.

Among the nuclear reactions which produce neutrons, those involving the bombardment of deuterium with deuterons or tritons seemed most suitable to produce a high yield of neutrons:

$$_1H^2 + {}_1H^2 \rightarrow {}_2He^3 + {}_0n^1$$

$$_1H^3 + {}_1H^2 \rightarrow {}_2He^4 + {}_0n^1.$$

Use of these reactions in a sealed-off vacuum tube seemed to involve a pulsed ion gun, a tube construction able to hold off high voltage, and a pulsed target in which the proper hydrogen isotope had been absorbed in a material such as titanium or zirconium. This system, initially developed at Berkeley by Gow, soon produced 0.1 to 1.0 microsecond pulses of approximately 10^6 neutrons, using experimental ion sources of several types. Early in the development, collaboration with the General Electric Research Laboratory was established.

Development of the Ion Source

A pulsed release of ions was first attempted by discharging a capacitor through a fine filament of titanium loaded with deuterium, but the current preferentially short-circuited through the released gas surrounding the filament. Although deuterons were abundantly produced, the lifetime of the filament was extremely limited because of hydrogen embrittlement, and this approach was abandoned.

Next it was found that ions could be produced from a vacuum arc struck between two electrodes of hydrogenated titanium; but the vacuum arc tended to strike erratically and at increasingly higher threshold voltages. These variations were reduced by placing an insulating bead in the gap between electrodes.

Since the use of tritium in the ion source promised a gain in neutron output of 10^2, a source with minimum hydrogen-absorbing material was now sought which could be safely loaded with a radioactive gas. Although with the d-t reaction more neutrons are produced by bombarding tritium with deuterons accelerated to given voltage (say 100 kilovolts) than are produced by bombarding deuterium with 100-kilovolt tritons, the latter allocation of

isotopes was preferred because the amount of tritium required in a target created an unnecessary safety hazard.

A consequent change in the geometry of the source reduced its size and improved performance. Instead of using solid electrodes spaced by an insulator, titanium was evaporated onto the surface of a quartz tube, 12 mm long and 1.5 mm in diameter. A suitable evaporated film of titanium contained about 0.5 milligram of metal per cm^2 and absorbed about 0.1 cc of tritium in the source, about 1/100th the gas occluded in the target. The leads from the source firing capacitor were clamped to the ends of the quartz tube. When the source capacitor (approximately 0.03 microfarad charged to 10 kilovolts) was discharged across the source with the aid of a thyratron switch, arcing occurred instantly across the one or more circumferential gaps inscribed in the titanium film on the quartz.

Tritium is evolved, particularly at the cathode spots, and is ionized as arcing persists while current flows in the source circuit. The firing of the source is regular, and ion production repeats well on successive shots. Alumina tubing was found to be better than quartz, because the adherence of titanium tritide to alumina is superior during the expansion which accompanies its formation, and because grooves can be more easily inscribed in titanium deposited on alumina. Other methods of producing arcs on the metalized surface of the insulator have been found satisfactory; for example, if the titanium is deposited in the form of a helix on alumina, the current short-circuits various turns when the voltage pulse is applied to the ends. But the inscription of one or more circumferential

gaps in uniformly deposited titanium seems best for pulses longer than 0.5 microseconds.

Operation of the Tube

A design of a sealed-off tube is shown in Figure 1. Both ion source and target are mounted within cylindrical stainless steel shields, preventing electrical breakdown of the glass vacuum-tube wall caused by electron bombardment while the high-voltage accelerating pulse is applied to the target. The ion source shield, which may be regarded as a partial Faraday cage, is especially essential.

The target in which the accelerated tritons are stopped may also be a thin evaporated titanium film (1 mg/cm^2). It is saturated with deuterium and should be free of any impurities or surface coating of oxide. Tritons of 200-kilovolt energy have a short range (about 1.8 microns in $TiD_{1.7}$); in stopping them it is desirable that they should react with deuterium for maximum neutron production, and be slowed as little as possible with heavier materials of high stopping power. The efficiency of a typical titanium target loaded with deuterium was found to be 1.5×10^6 neutrons per microcoulomb, or 2.4×10^{-7} neutrons per deuteron, for 230-kilovolt monatomic D^+ ions from a van de Graaff generator.

The usual methods of vacuum tube technology cannot be applied to the seal-off of this type of tube. When the tube shell contains a deuterated target and a tritiated source, a normal bake-out procedure is impossible because the isotopes begin to evolve from the titanium at temperatures in excess of 200°C. The tube shown in Figure 1 was fitted with a small bulb of activated charcoal serving as an auxiliary pump to maintain a high

vacuum. An electronic pump, as shown in figure 3, (Reference 8) has the great advantage that it can remove the usual gases and also the He^3 which is not absorbed by chemical getters.

The most versatile operating arrangement involves powering the source and target from separate circuits. Pulse lengths achieved range from 0.1 to 10 microseconds. For the shorter pulses, a capacitor is discharged directly through the source, peak current of about 120 amperes being desirable. A target pulse in the 100 to 300 kilovolt range is obtained by discharging a capacitor through the primary of a pulse transformer. It is necessary, of course, to insulate the outside of the tube with oil or a pressurized gas like sulphur hexafluoride. The source shield is common to both of the circuits and is usually maintained at ground potential. The neutron output, as a function of a target pulse delay with respect to the source pulse, peaks at about 0.4 microsecond. For longer pulses a pulse-forming network is discharged through the series combination of the termination resistor and the source (Figure 2). In this case it is desirable to limit the current to lower values, i.e., to about 30 amperes. For reliability of operation, the voltage initially impressed across the source should be at least twice the arc-breakdown threshold. Longer target pulses are obtained similarly by the discharge of an LC pulse-forming network through the primary of a pulse transformer which should be designed to supply a tube current of about 10 amperes. The source pulse should be terminated promptly, before any reversal of the high-voltage target pulse occurs, so as to avoid late breakdowns which gas up the tube unnecessarily and reduce source life.

The neutron yield is a linear function of source current up to the point where tube breakdowns become frequent. As a function of target voltage, the yield exhibits the characteristic shape of the predicted thick target yield curve (Reference 9). From the tubes shown in Figures 1 and 3, a representative yield of 10^7 neutrons/pulse is attainable for either short or long pulses. Later work (Reference 10) has shown that the ion current is composed in large measure of titanium ions as well as of tritium ions from the expanding plasma cloud at the source. A rough calculation shows that the neutron yield is consistent with measured tube current at various voltages. For example, at 150 kilovolts, 0.1 microcoulomb of tritons is needed at the deuterided target for 10^7 neutrons. With microsecond pulses, peak currents of 0.1 ampere of tritons involve 0.2 ampere of Ti^+, Ti^{++}, and other useless ions, and about 1 ampere of secondary electrons returning from the target to the ion source shield and header. The capacity current of particular tube envelopes and transformers must be considered. Measured tube currents are indeed of the order of 3 amperes. Pon (Reference 11) has shown that the secondary emission ratio of $TiD_{1.7}$ is from 2.8 to 3.4 for D^+ energies of the order of 100 kilovolts, and that voltage dependence of the ratio is not large.

It is practical to alter the performance and increase the efficiency of the tube by the use of grids across the ion source shield and across the target. By means of a target grid, negative with respect to the target, or with respect to which the target is self-biasing, the secondary electron current may be partially suppressed and the current efficiency

of the tube improved; however, grids may tend to lower the voltage breakdown limit of the tube geometry.

Because of heat dissipation in the tube, the pulse repetition rate is limited to about 10 pulses per minute. The possibility of producing a series of short neutron pulses, very closely spaced in time, was explored with tubes having two or more independent sources mounted side by side in a single shield and opposite one target. For 0.2-microsecond pulses the output of a second source was essentially unaffected by the firing of the first source until the interval between firings was less than 0.5 microsecond. The peak output was not doubled as the two sources were fired in coincidence, but with a slight delay the efficiency of the second one was normal. The use of several sources in one envelope has permitted source designs to be compared quickly for identical processing and vacuum conditions. The lifetime of the neutron tube is determined by arc-induced damage to the evaporated titanium source. Yields become progressively erratic after 4000 to 6000 shots and intolerably so after perhaps 10,000 shots.

The electronic pump and an active barium getter are used to remove those gases which evolve in the neutron tube during pulsing. Initially the tube is processed so as to eliminate any contamination of its parts. During its normal usage the tube pressure between shots is below 0.01 micron. However, some experiments, in which higher pressures of various gases have been deliberately introduced, show that with some gases, particularly helium, rather high pressures can be tolerated. The following table shows approximate "1/2 output" and "output below 1/10th" pressures for a typical tube of the Figure 3 design.

Gas	One-Half Output Pressure - Microns	Below 1/10 Output Pressure - Microns
He	25	65
H_2	5	12
CO	1	3
CH_3	1	3
SF_6	0.2-0.5	1

Charge exchange and scattering of the ions crossing from the source to the target, rather than high-voltage breakdown, set a maximum pressure limit.

When approximately 5 microns of tritium were added to an evacuated tube having no getter, there was actually a small increase of output. Presumably, formation of tritons in the vicinity of the source-gap was now more efficient than when all the gas near the source was evolved from the cathode spot and included metal vapor as well as tritium. A tube was tested in which the metalized source was of evaporated molybdenum containing no tritium. All the tritium in this tube was introduced as a gas. The neutron output was about normal until the gaseous tritium became depleted as the tube hardened following a series of shots. The limitations set by the rapidity of gas exchange at the target and the difficulty of pressure stabilization seem to make such gas-filled tubes less versatile than the well-evacuated neutron tube.

Fabrication of the Neutron Tube

To make a sealed-off neutron tube involves special vacuum procedures and the safe handling of a radioactive gas, even minute quantities of which

present a health hazard (Reference 12). Because of the weak nature of the beta radiation (maximum energy of 18 kev), glass apparatus provides complete radiation protection as long as the integrity of the system is maintained. But, despite the low energy of the radiation, the 12.4 year half-life makes tritium an intense source (2.63 curies/cc, STP) and careful measures must be taken to prevent contamination of personnel. Since the recommended maximum concentration of HTO in air is 7×10^{-5} microcuries/milliliter (Isotopics, January 1956), an adequately ventilated hood and sensitive monitoring are essential.

Clean fabrication of the tube parts, particularly of the source and the target, is important. Fabrication steps which were followed are outlined below.

The ion source is the crux of the tube. The material used as the source insulator is a high-fired alumina tubing such as GERL 2548 or Coors AI 200, in the class of approximately 95 percent Al_2O_3. The fired tubing is cut to length, washed in alcohol, and heated to a dull red in a hydrogen flame. The hollow rod is threaded onto 0.020-inch tungsten wire which is then spot-welded to leads so that the rod is concentric with a spiral-shaped evaporator filament (Figure 4). Under a bell-jar, when the vacuum is of the order of 10^{-6} mm of Hg, the ceramic is outgassed by passing current through the tungsten wire until the ceramic is brought to a dull red. In a number of cycles, titanium is evaporated from the spiral evaporator onto the warm ceramic until the evaporator bulb has become properly opaque. At this point, it seems to facilitate the eventual loading of tritium into the source if the titanium is first loaded with normal hydrogen or deuterium. Titanium loading is accomplished by admitting the gas to a

few centimeters of pressure and then heating the source momentarily to 500 to 600°C. After the source has cooled, the residual gas is pumped, air is admitted, and the source removed. At the center of the coating a shallow groove is cut with a clean diamond tool.

The evaporator filament is prepared in a separate vacuum operation. First a narrow titanium ribbon is wound tightly around a tantalum wire. Then the combination is shaped into a spiral and inserted into the vacuum system. Current heating is used to degas the spiral and to raise it to a temperature where the titanium fuses onto the tantalum, from which it subsequently will be evaporated.

To mount the grooved source to the header posts clean nickel bands or tapered tubular sleeves are used, which can be forced over the ends of the alumina. These parts have been hydrogen-fired and vacuum-fired to a temperature just short of that at which melting occurs. The firing and handling procedures should be rigorous, because any contamination of the ion cloud at the source will lead to the acceleration of unwanted ions to the target and to a loss of tube efficiency. A nichrome heater coil (with a resistance of approximately 60 ohms) of 0.002-inch diameter wire, wound on a 0.010-inch mandrel, is inserted through the nickel sleeves and the 0.020-inch axial hole in the alumina tube. Both sleeves and nichrome leads are spot-welded to the Kovar wires from the header. The nichrome coil provides a means of heating the source during tritium loading and yet has sufficient resistance not to shunt the gap when the source is fired during normal tube operation.

Target foils may be prepared by occluding deuterium into titanium which has been evaporated onto a molybdenum or tungsten backing. The technology here is already quite common (Reference 13). For the maintenance of a high vacuum within the sealed-off tube, an equilibrium pressure less than 1×10^{-6} mm Hg is desired; this can be obtained at room temperature within the γ-phase region of the titanium-deuterium system (Reference 14) at concentrations up to $TiD_{1.8}$. Efforts to use lithium systems were not fruitful.

The thickness of the titanium layer should be at least 3 microns, exceeding the range of the highest energy tritons which can be accelerated. A proposal by Grams (Reference 15) for getting the layer of titanium directly on the inside of the Kovar cup at the target end of the tube has been found convenient. A single turn coil of graphite is mounted on two posts brought through vacuum-tight insulators on a steel plate (Figure 5). The tube body, previously cleaned and sand-blasted on the inside to improve its high-voltage creepage strength, is used as a bell-jar clamped over the plate. The inner glass surface of the tube, but not the Kovar end cup, must be shielded from the graphite filament by a clean cylinder. The graphite heater filament on its upper face is dished to contain bits of titanium wire. When the filament is first heated in vacuum it reacts exothermically with the wire to become titanium carbide. Thereafter, the carbide filament is used as a boat from which the weighed bits of titanium wire are evaporated to deposit uniform films of titanium on the inner surface of the Kovar cup forming the target end of the tube envelope. After evaporation the bell-jar may be let down to atmospheric pressure with hydrogen, or, better yet, with deuterium.

The various stainless steel parts of the neutron tube, such as the source shield and the pill box of the electronic pump, are all subjected to hydrogen-firing and vacuum-firing before assembly. After the tube assembly is closed by shielded-arc welding, in an argon atmosphere, the tube is evacuated and baked at 450°C for two hours before the source and target are loaded for the final time.

The deuterium and tritium are stored in uranium or titanium beds contained in quartz bulbs sealed to a glass system. A mask and protective gloves are worn when tritium is evolved into the system, to which the neutron tube is sealed for processing. First, the deuterium pressure is raised to several centimeters of Hg. The titanium surfaces of the neutron tube are deuterium-loaded while the tube is being cooled slowly from 450°C to below 200°C. The excess deuterium is reabsorbed in the bed, and that deuterium which is contained in the source is evolved by heating the source with the spiral nichrome filament. Next, the tritium pressure is raised in the tube and the source is temperature-cycled several times to assure its proper loading. After excess tritium has been returned to its bed, the tube is evacuated and baked at 200°C. A proper enclosure, and venting of the pump which handles waste tritium, are esential. Before the conclusion of the bake-out, the electronic pump appendage is heated with a radio-frequency coil to a dull red temperature to free its surfaces of gas. The getter wire is preheated but not flashed before the final seal-off.

The processing hood was designed such that a tube could be oil-immersed and pulsed, to obtain neutron output, while still on the pump. This permits a beneficial high-voltage "aging" of the tube walls and the

removal of some additional unwanted gas before the seal-off. A sealed-off tube suitable for 10^7 neutrons/pulse output is shown in Figure 6. Both smaller and larger tubes have been constructed; from the latter size it was possible to attain as much as 10^9 neutrons/pulse for short pulses using a target voltage of 300 kilovolts.

ACKNOWLEDGEMENTS

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FIGURE LEGENDS

Fig. 1. A design of sealed-off tube.

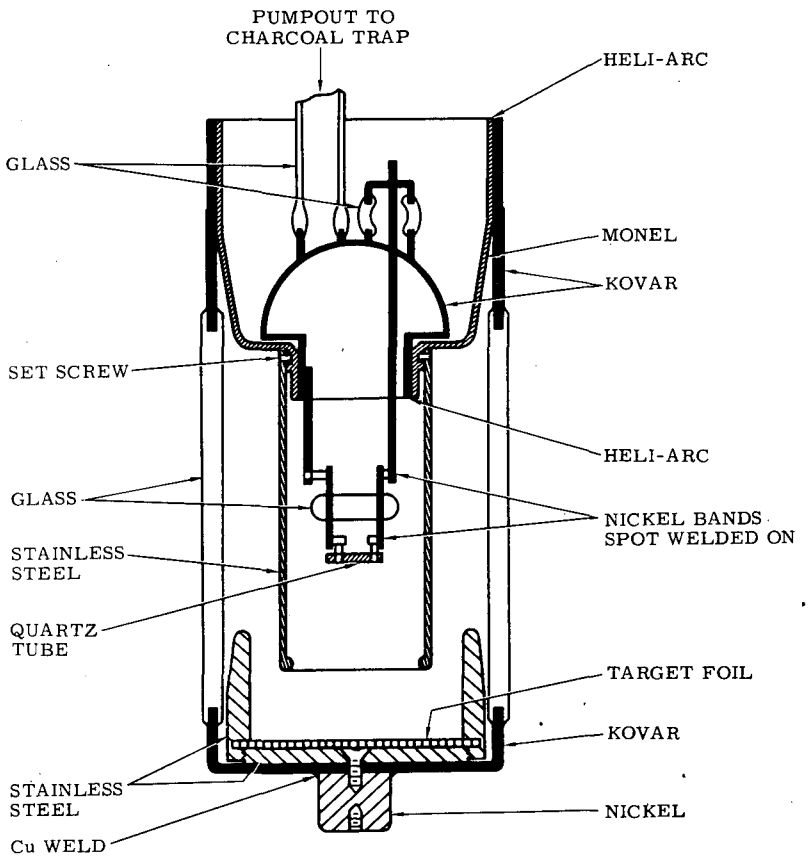
Fig. 2. Tube-driving circuit.

Fig. 3. Improved tube with barium getter.

Fig. 4. Spiral evaporator for titanium.

Fig. 5. Special evaporator heating coil for depositing titanium inside Kovar cups.

Fig. 6. Tube suitable for 10^7 neutrons/pulse output.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CHARCOAL TRAP
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GLASS
MONEL
KOVAR
SET SCREW
HELI-ARC
GLASS
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SPOT WELDED ON
STAINLESS
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TUBE
TARGET FOIL
KOVAR
STAINLESS
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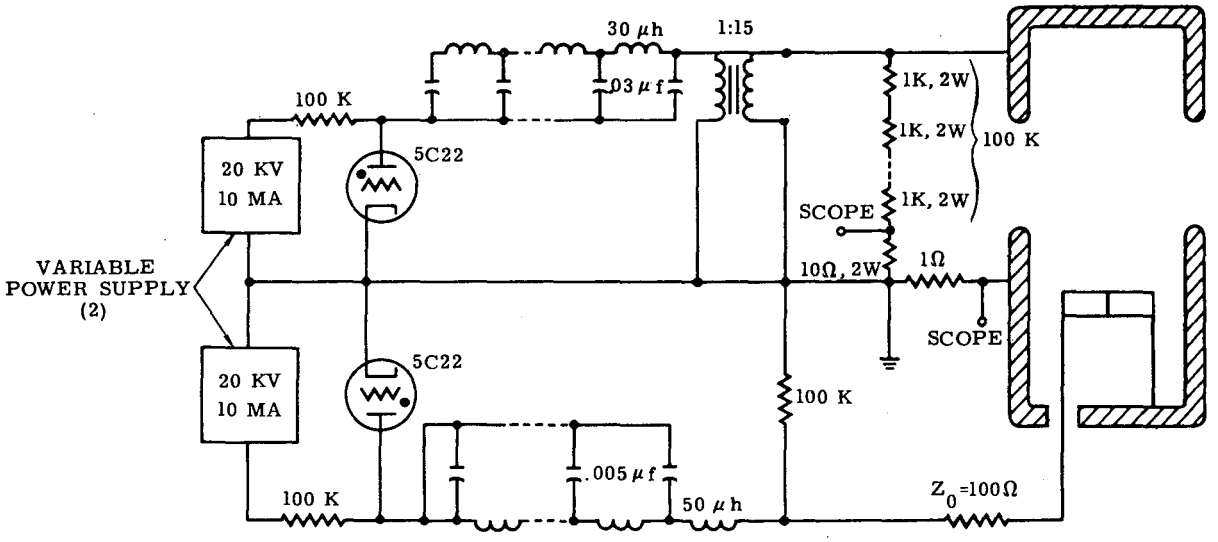
NICKEL
Cu WELD
MU-18443

Fig. 1



MU-18444

Fig. 2

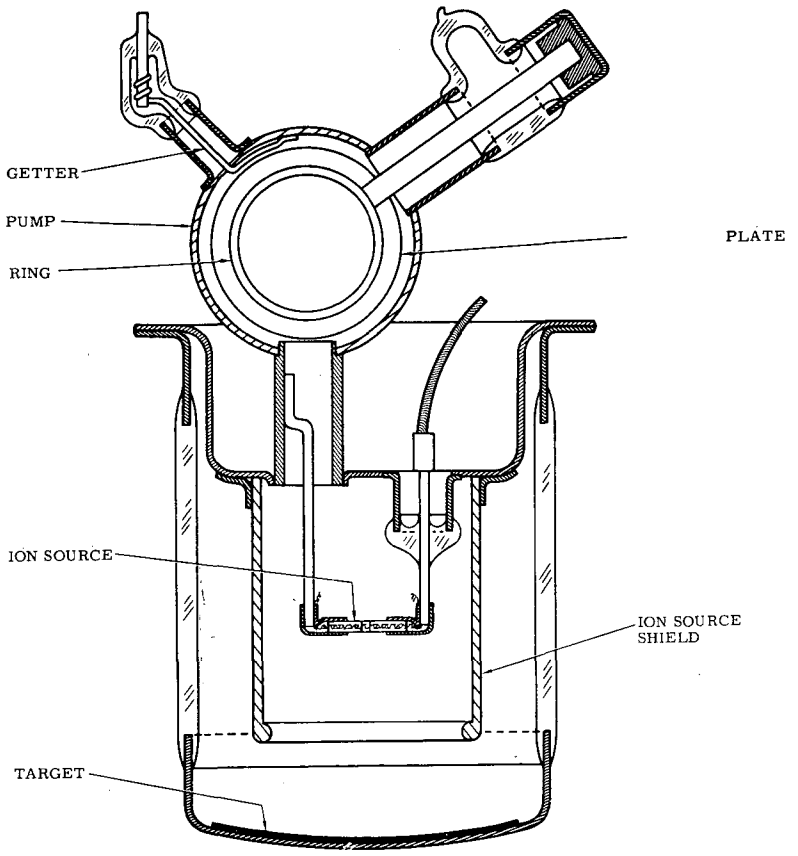

GETTER
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PLATE
ION SOURCE
ION SOURCE SHIELD
TARGET
MU-18445

Fig. 3

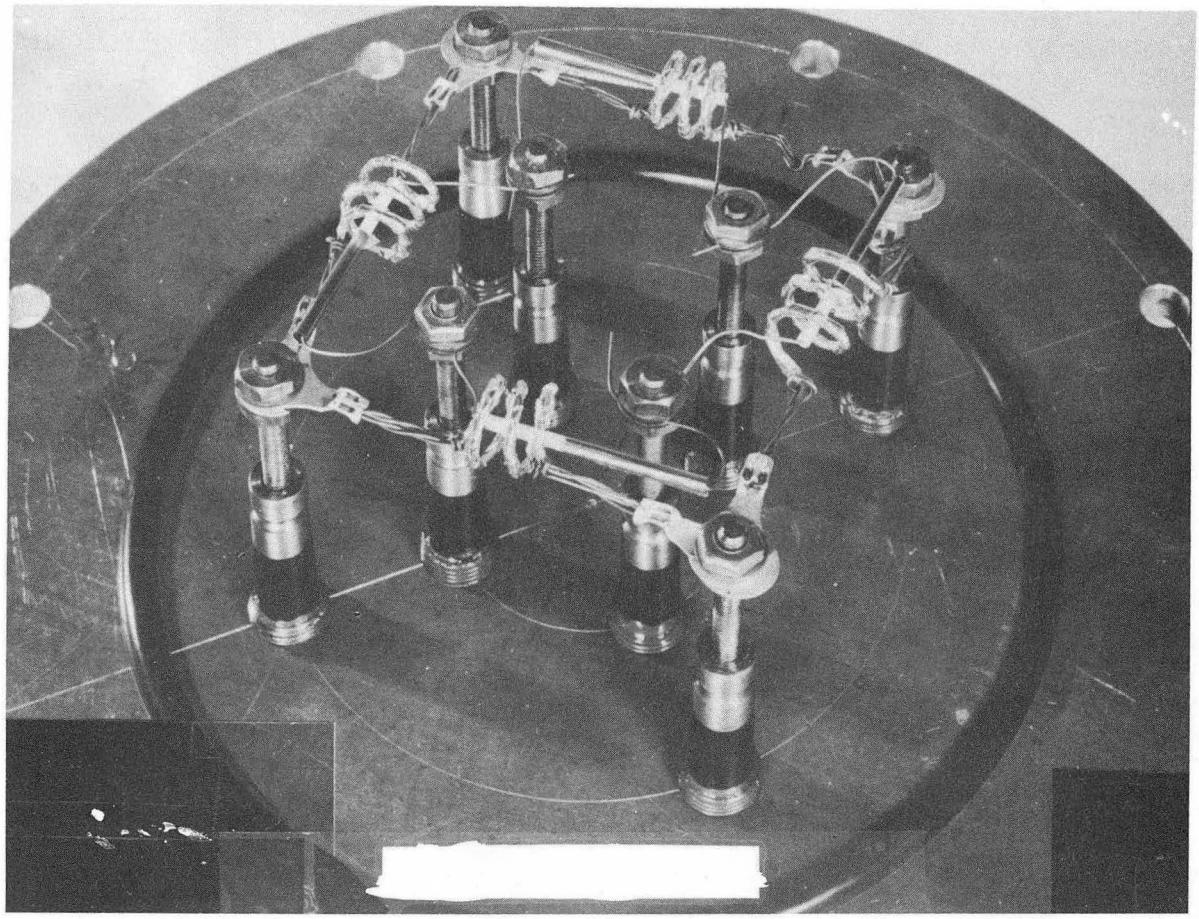

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Fig. 4

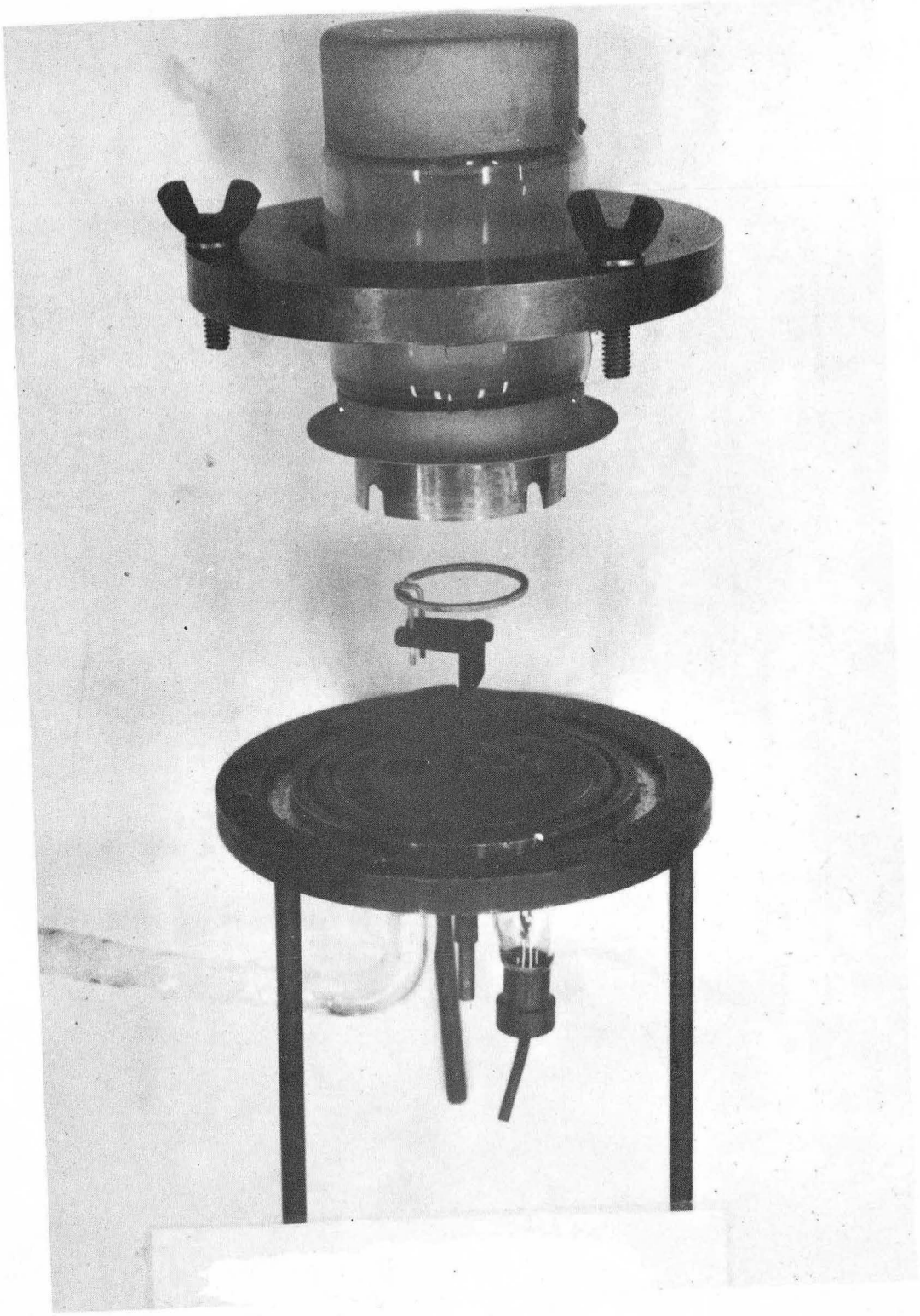

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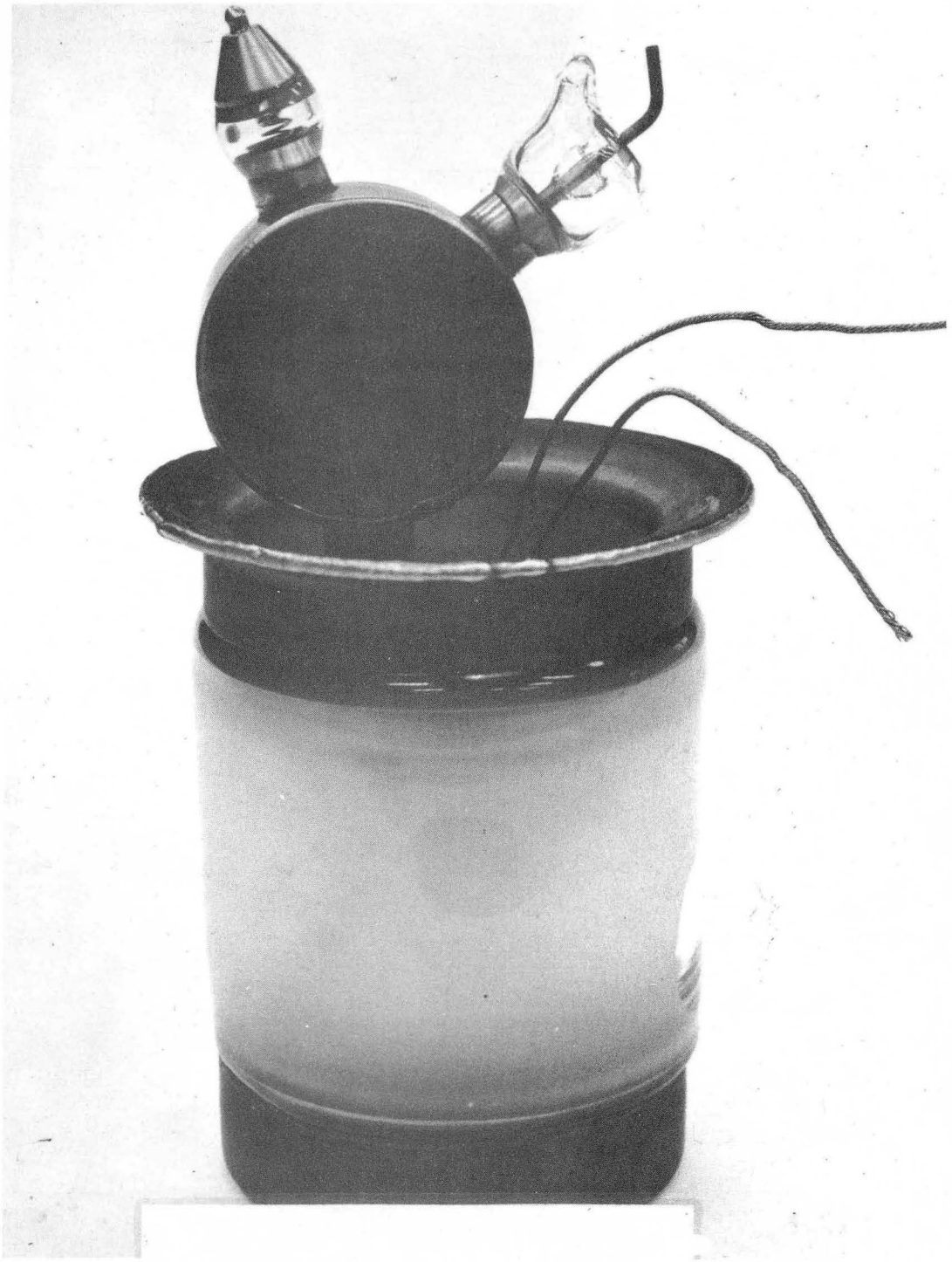
Fig. 5



ZN-2269

Fig. 6