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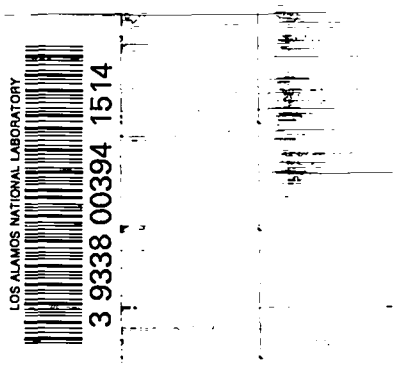


INTRODUCTION

Roy Reider
Safety Director

A few weeks ago I was approached by Robert Sherman of Q-26 with the suggestion that a committee should be established to sponsor a forum for exchange of information on tritium operating and safety problems. We agreed that Los Alamos was uniquely competent in this field, and tritium was used so widely that such a forum would serve many tritium users and potential users.

Little enthusiasm developed for another committee, but considerable interest was avowed for a single session. The interest was so real that every principal approached agreed to contribute, and it became only a question of selecting a time and place for the seven speakers to assemble. A letter was sent to divisions and groups interested in the subject and an announcement was placed in the LASL Bulletin. Attendance was 115.

Material Use (Tubing, Lubricants, etc.) - James Anderson, CMB-3

I want to discuss how we handle tritium at CMB-3 and what we use in the way of materials. The best way to talk about such a general term as "materials" is to tell you how we do things, what we use, and why we use it.

We work with large quantities of tritium in the form of gas and as solids. The solids are usually chemically reactive metal tritides. Our first goal in setting up a piece of equipment for working with tritium is to devise a secondary containment system. We have a primary system that contains the tritium, in the form of solid tritide or gas, contained within some other (secondary containment) vessel separating the tritium from the operator. If a leak develops in the system, there is thus some back-up containment before the operator starts inhaling the material.

We have chosen to use as our secondary containment vessel a dry-box system (Fig. 1), because an inert atmosphere must be maintained at all times when working with metal tritides. We have also chosen to put our gas systems within the dry box. If the gas system develops a leak we contain the tritium in the dry box where provisions have been made for removing tritium from that atmosphere, oxidizing the tritium to water, and collecting the water at another location in the building. The tritium that escapes into the dry box can then be rather rapidly recovered. Within the dry box we have a vacuum system for working with tritium gas. It includes standard volumes, gauges, pressure transducers, storage volumes, a uranium bed for storage and purification of tritium gas, and a vanadium tritide bed for boosting tritium gas to high pressure (103 MPa).

The only portion of the tritium handling system that is outside the dry box is a short run of stainless steel tubing. This goes from the dry box (and the gas line in the dry box) to the container in which tritium gas is received from Savannah River.

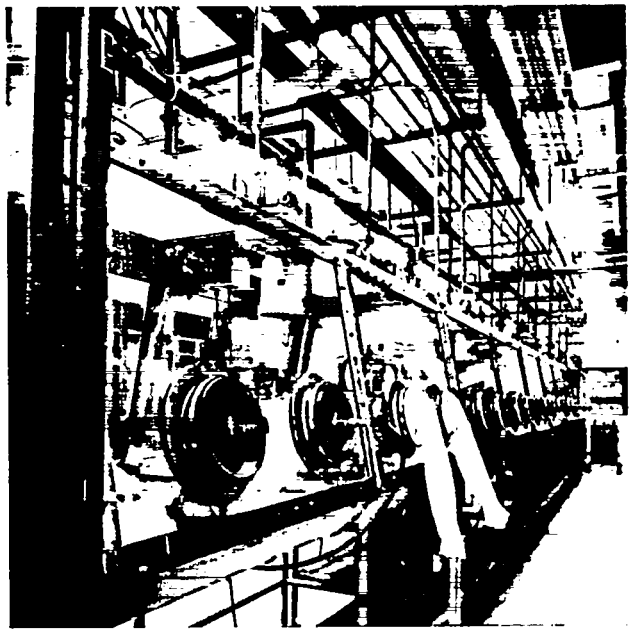


Fig. 1.
Dry box system.

What materials do we use in our system? First of all, we strive as much as possible to have an all-metal system. It cannot be completely achieved, but we strive toward that in all instances. We prefer to have welded or soldered joints wherever possible. This also is not entirely possible, because in any system there will be joints that will have to be opened, some with more frequency than others. For nonpermanent-type joints, we prefer to use Cajon fittings with metal (aluminum or nickel) gaskets. We do, on occasion, use Swagelok fittings.

Once the system is assembled, we helium-leak-check it as thoroughly as possible. Also, we incorporate into the system a double valving and purge system, especially for those parts that will require frequent maintenance. Figure 2 illustrates this. In

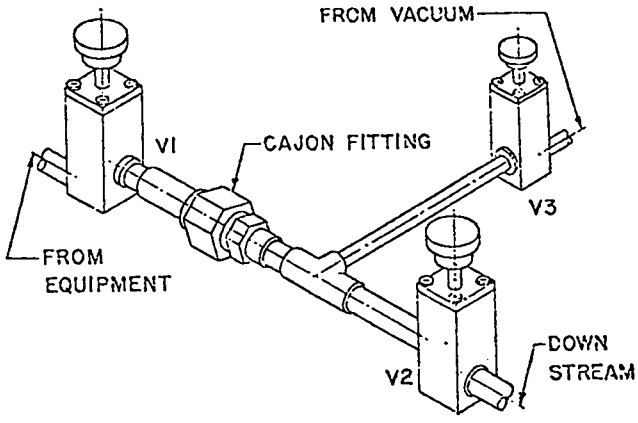


Fig. 2.
Valve and purge system.

Fig. 2, the piece of equipment on the left is connected to the next portion of the line by a valve. V1. Then there is a purge system through valve V3 that goes to a vacuum line, and a third valve, V2. If this piece of equipment has to be taken out, we can close valves V1 and V2, then open V3 and pull a vacuum in that portion of the line.

When we get a good vacuum, we can backfill with air or inert gas as many times as necessary to achieve a clean atmosphere. Then we shut valve V3 and can open the fitting. Not all contamination can be eliminated in this way, but personnel exposures can be reduced tremendously. We use this technique throughout our system wherever we think joints will have to be opened or wherever tritium-contaminated parts will have to be handled.

We use a lot of stainless tubing and we prefer 316 stainless where possible; 304 is also acceptable. Any of the low 300 series stainless steels is adequate for tubing or for other parts of the system. We prefer the 316 to the 304 because there is some evidence that tritium will tend to leach carbon out of the 304 somewhat faster than it will from the 316. This means that within a system one will occasionally have traces of tritiated methane and ethane; the 316 seems to be a little better in that respect. Another acceptable stainless steel which is used in tritium systems is 21-6-9. Avoid the 400 series stainless steels because they tend to show some hydrogen embrittlement. Recently Group WX-5 had failure in old copper tubing in Stoll's system.

We use metal gaskets wherever possible. Aluminum, copper, or nickel gaskets have been used. At some places in our system, especially along the manifold that feeds the dry-box system, we do not use metal gaskets. In those instances we have gone to a Grafoil gasket. Grafoil is a pure carbon material available from Union Carbide. It comes in sheets of varying thicknesses from which appropriate gaskets can be fabricated.

Elastomers are used in some places in our system. and with tritium one has two problems with them. One is the radiation damage resulting from the tritium decay. The other problem is that tritium will exchange with hydrogen in the elastomers. There are two considerations: 1) how much exchange will there be with hydrogen in the material, and 2) how much radiation damage will occur from the tritium decay. When these two problems were considered, we decided that Viton elastomers would be acceptable. Radiation chemistry books will say that Viton. which is a fluorinated hydrocarbon, does not have very good radiation stability. We have found that it works quite nicely in low-level tritium systems because one does not get the other problem of tritium exchange with the hydrogen. Viton is used for O-rings and we use some valves that have Viton seats. Occasionally we use neoprene in our system, particularly in the dry box. Neoprene gaskets are used around windows in the dry box, and neoprene gloves are used on the dry box. The neoprene has a finite lifetime, especially if large surfaces are exposed to a tritium atmosphere, but we have used neoprene with some degree of success.

Where one cannot use a metal valve, one can use valves having seats of Teflon, reinforced with 15-20% fiber glass. These hold up farily well. Teflon is not considered to be unaffected by radiation but, because it is a fluorinated hydrocarbon, there isn't much tritium exchange. Use of the glass-reinforced Teflon also alleviates the problem of creep. We use a lot of Worcester ball valves with fiber-glass reinforced Teflon seats; they work adequately and hold up quite well.

We don't use lubricants and vacuum greases very much, so I can't say much about them. We have used both silicone lubricants and vacuum grease and we have used the Apiezons. I can't elaborate because we don't use these materials in long-term experiments. Occasionally they are used in short-term experiments, where we have found both the silicones and Apiezons to be adequate.

Regular use is made of off-the-shelf mechanical vacuum pumps and regular off-the-shelf Duo-Seal pump oil. We do have a system for changing the pump oil. Referring to Fig. 3, I will describe briefly what we do. At the top of the figure note the word "suction." That is our experimental station. It can be an airlock or it can be whatever experimental station we want to evacuate. Typically, we will evacuate through the vacuum pump, exhausting to the effluent treatment system, which is a system for

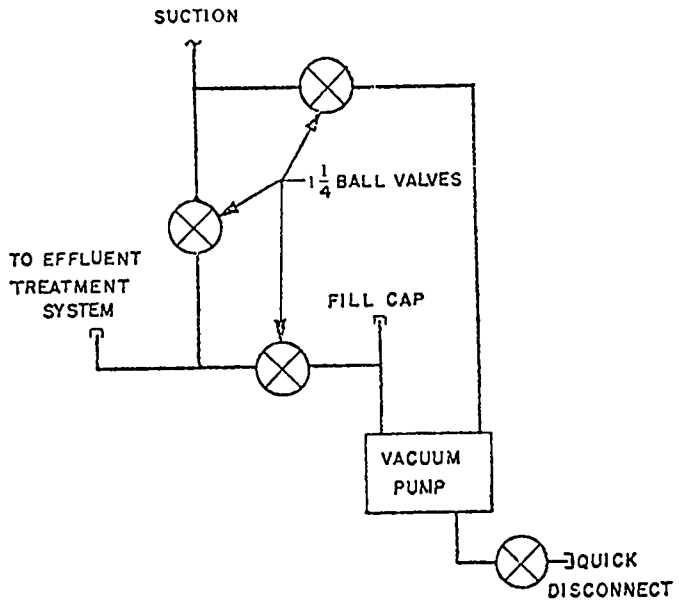


Fig. 3.
Schematic of evacuation system.

cleaning up all of our effluents by scrubbing tritium from the effluents before they are released up the stack. When we get ready to change oil in this pump, we use the quick disconnect drain and a fill cap on the exit line. We can isolate the pump and have a container which can be evacuated and connected to this quick disconnect. Then the valve is opened and the oil flows from this pump into the evacuated container and is collected in this way. We can then come back to our fill cap and add the prescribed amount of oil. Thus the oil can be changed without any tritium being detected in the room and without any exposure of personnel. Vacuum pump oils can be highly contaminated. Mound Laboratory has studied this quite extensively and has found that pump oils tend to saturate at about 30 Ci/l. That is a lot of tritium to end up in the pump oils. Figure 4 shows the system in operation. We have a vacuum pump with a hose running up to the evacuated container, which is a standard 30-gal drum filled with vermiculite. One valve is connected to a vacuum system to evacuate the tank. The second line goes over to the quick disconnect on the vacuum pump. We evacuate the container, open the valve, and pull the oil into the container, where it is adsorbed on the vermiculite. The valves are then shut off, and oil can be added through the fill cap. Not only have we changed the oil, but we've also collected the contaminated oil as a solid adsorbed on vermiculite. This container can then go straight into an asphalt-lined 55-gal drum and can be taken to H-8 for burial. For diffusion pump oils, we prefer the silicone fluids, i.e., the standard DC-704 type pump fluids.

We like to get tritium into the dry box as soon as possible after receiving it so that we do not have a container of tritium gas stored in our laboratory. We have a large uranium bed in the dry box as part of our gas system. Tritium is brought into the dry box as a gas in the vacuum line, is measured, and then is absorbed on the uranium. This technique achieves several things: (1) tritium is stored as a solid, (2) it is kept in the dry box, where we have better control over it, and (3) the method also helps purify the tritium. Any oxygen or nitrogen contamination in the tritium stream will also be absorbed on the uranium bed. When ready to take the tritium off the bed. we heat the bed to 450-500° C. The oxide and nitride which have been formed will not decompose, but the tritium is readily released. This also gives one an opportunity to purify the ^{3}He decay product. When a uranium bed is used for storage, the ^{3}He will not be absorbed. Tritium can also be purified by diffusing it through a palladium diffuser. Tritium will diffuse rapidly through a heated palladium membrane, whereas oxygen, nitrogen, and other gases will permeate the palladium much more slowly. Also, the ^{3}He can be removed this way because it will not diffuse through the palladium membrane.

We have a large effluent treatment system which we use to scrub all effluents generated in our facility before they are released up the stack. It is a rather large, elaborate system, but anyone setting up a small experimental system using tritium can very simply do the same thing. To clean up the effluents, all one needs is some system by which the tritium gas can be oxidized to water. There are several ways to accomplish this. Copper oxide heated to 350°C will oxidize hydrogen and convert it to tritiated water. There is also a commercial material, made by MSA and called Hopcalite; it is a copper oxide-manganese dioxide mixture. At 350-400°C, Hopcalite will oxidize the hydrogen isotopes to water. One can also use precious metal catalysts to achieve this oxidation. The "Deoxo" unit used for purifying oxygen tanks is simply a precious metal catalyst. One can get catalysts from several places - MSA or Engelhard, for instance. These catalysts are platinum or palladium particles finely dispersed on a ceramic substrate. The precious metal will catalyze the $H_2 + 1/2\ 0_2 \rightarrow H_2O$ reaction. One must be sure that sufficient oxygen is available to react with all of the tritium. The precious metal catalyst has the advantage that it will also catalyze the oxidation of hydrocarbons. If one has tritiated

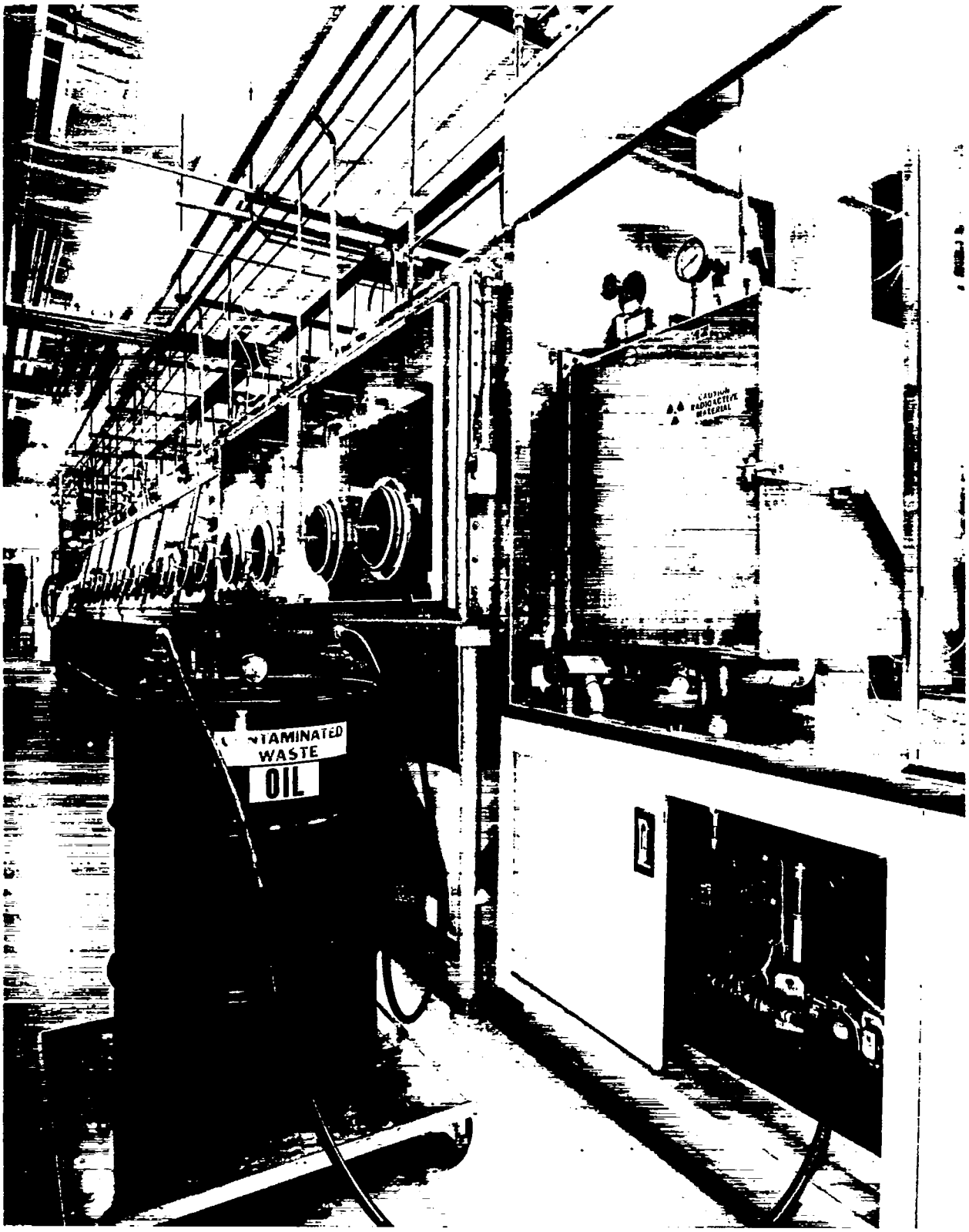


Fig. 4.
Oil waste collection system.

hydrocarbons, a precious metal bed operating at 500°C will oxidize the hydrocarbons to water and carbon dioxide.

These are simple setups to put together. As it comes out of the oxidizing bed, the exhaust stream must be cooled to near room temperature, and the gas stream must pass through a molecular sieve trap containing 4A or 5A molecular sieve which will adsorb water to the 1-ppm level. With a simple setup such as this, one can scrub tritium from vacuum pump exhausts in a small system. In a larger system, one would use the same philosophy but scale everything up in size. This is what we do at DP-East. At CMB-3 we use a large precious metal catalytic recombiner to convert all tritiated materials to tritiated water which is, in turn, adsorbed on molecular sieve. That water can either be buried or sent to an off-site recovery.

These are some of the things we do at CMB-3, and if you are interested, come out and take a look at this facility. We will be glad to show you around.

Hardware Selection (Valves, Fittings, Pumps, etc.) - R. Sherman, Q-26

As Roy said, I was the instigator of this symposium and it was my hope that we would be able to air our "dirty linen"; specifically, problems and incidents that we have had and how we have solved them so that we may learn from each other. If there has been a system failure it is folly to keep the information to ourselves when we are dealing with toxic materials such as tritium.

Our concerns in Q-26 span the spectrum of tritium usage from small-scale research to large-scale research to commercial-scale continuous operating systems. In the last case I am referring to the low temperature distillation system for separating tritium from a DT-in-D_2 mixture for the Intense Neutron Source (INS) facility.

Hydrogen embrittlement is a problem not only of hydrogen, but of deuterium and tritium. We have, for example, experienced pressure gauges being tested to 34 MPa and rupturing at 3.4 MPa on the second use. One must be careful with the selection of materials, avoiding carbon steels, 400 series steels, certain high nickel steels, and Inconel. On the other hand, the 300 series steels work quite well. Much of the embrittlement work was done by Fred Edeskuty and Bob Mills of Q-26 whom I consult when in doubt.

Elastomers, in general, do not stand up well in a tritium atmosphere. The tritium probably causes a rupture of the cross linkages in the elastomer molecules with a resultant loss of elasticity. Pump oils will harden to a Bakelite-like material in a year or so.

Pumping of tritium has always been a problem. One of our biggest successes has been a rotary vane pump (similar to a Welch Duo-Seal) designed by Bob Mills. This particular pump has two stages and has a capacity of about 1 ℓ/s. It will pump a vacuum of ~ 1 Pa (10^{-2} torr) or pressurize to 0.7 MPa (110 psi). This pump uses only about 1000 mm^3 of oil (1 cm^3) and so the exchange problem is not too great. Don Coffin has recently redesigned the rotary shaft seal to incorporate O-rings with great success.

Metal Bellows Corp. has a series of pumps which seem to have some potential, although there appear to be some major problems with obtaining an adequate seal at the head. They have made an all-welded double containment version for LLL, but at about a fourfold increase in price.

We have had very good experience with the diaphragm compressors such as are marketed by American Instrument Co. If one uses the Nylon(?)-faced check valves there is need for replacement on perhaps an annual basis. There are models ranging from 68 to 207 MPa with a compression ratio of about 15/1. The model we have operates from 0.55 MPa of compressed air as power source.

For circulation only, the French SRTI pump appears to be a superb instrument. We have the small model (PR-9). The pump uses a diaphragm attached to a wobbling bellows fitted loosely into its housing. The pump rotates (or wobbles) at 3000 rpm and will circulate 100 ℓ/min at a pressure head of 1.3 x 10^4 Pa (80 mm).

At lower pressure levels, the Edwards 2M4B mercury diffusion pump is a remarkable device. It will pump a good vacuum with a backing pressure of up to 6.5 x 10^3 Pa(40 mm). With a 51-mm throat, it has a pumping speed of 70 ℓ/s. Such a pump has been used as a circulator in a system which did not require a high operating pressure.

On the subject of fittings, we have had fair luck with Swagelok if one is careful. We are starting to convert to the Cajon-VCR fittings. At the present time we stock only a few sizes of simple straight couplings so each of us has had to lay in his own stock. I volunteer to coordinate the requests, and the stockroom has agreed to expand their line. The principle is very similar to that of the Varian vacuum flange—a deformable metal gasket, which is a cheap, throwaway component. The standard fittings are asexual and of 316 stainless steel. Gaskets are available in nickel, copper, and aluminum, among other metals. Our experience has been that they are easy to put together. The gasket fittings require 1/8 turn to tighten, and the joint is leak-detector tight.

We have shock-tested several of these fittings to liquid nitrogen and liquid helium temperatures with no leaks or failures. Our technician has been unable to make them leak. (On the other hand, the Main Shop has been unable to make them tight.)

Valves have always been a problem. The Hoke toggle valves work for about a year and then the valve insert assembly needs to be replaced. Likewise, the Hoke HGP valves work well with annual changes of the bellows assembly and gaskets. Don Coffin will have some words to say on newer seat materials. Until about two years ago my experience with the Hoke bellows valve (4171 M4B) was good. There must have been a change in the bellows supplier, however, because now I have a 20-30% failure rate. We have had good luck with the Model 4251F2Y Teflon-gasketed stainless steel valve and especially with the Model 4213Q6Y all-welded stainless steel (316) construction valve. Recently, at my request, we began to stock several of the NUPRO welded SS diaphragm valves with Swagelok fittings (SS-4H). I have yet to have a failure in one of these valves.

One further topic I would like to cover is on overpressure relief systems. Ordinary relief valves either leak too much for use with tritium or the elastomer seals embrittle with a resultant failure. This leaves one with the last resort of rupture disc assemblies.

We have used rupture disc assemblies manufactured by BS&B (Black, Syvalls, and Bryson). We used their threaded union and made lead gaskets to effect a vacuum seal. In conversation with others, however, I find that we are not alone in a communication failure with this company. We ordered discs to rupture at a differential of 30 psi, using the specifications quoted to us by a factory engineer, only to find that the discs as received rupture at 15 psia. The factory rated them at 30 psig! This came as quite a shock and caused some very tense moments.

Another firm which custom-makes a high class of rupture disc is Calmec, a subsidiary of Amatek. In their design a self-actuating spring-loaded cutter can be set to shear the disc within 1% of a preset pressure with an adjustment range of about 15%. They are more expensive, but can be sized much smaller than the types which operate on a mechanical failure principle. They are reusable upon insertion of a new disc and since the discs are thicker - quite reproducible.

Biological Effects - J. Lawrence, H-1

The biological effects of tritium result, primarily, from the energy deposition of the tritium beta-ray energy in tissue and from the associated ionization in tissue. There has been some speculation that significant tissue damage would be caused by the transmutation of the hydrogen isotope into helium, thereby breaking biochemical bonds. Experimental evidence indicates that transmutation damage is a very small contributor to the total cellular damage.

Biological damage has been observed by experimentation on small mammals and other animal organisms. The techniques used are to inject the animals with relatively large amounts of tritium (in the form of various compounds) and to sustain that quantity of injected material for a long period of time. The observed biological effects include: atrophy of the spleen and thymus in mice, rats, and hamsters; mortality (for sufficiently high exposures); incidence of tumor formation in hamsters; and the killing of spermatogonia in mice. All of these experiments have indicated that a sustained exposure to the extent of at least 1 μCi per gram of body weight was required.

That is a rather large quantity of tritium. If one extrapolates "1 μCi per gram of body weight" to a standard 70-kg man, that says that the man would have to sustain 70 mCi of tritium in his body **continuously** for any effects to be observable. For those of you who are not acquainted with the accepted "permissible exposure level," 70 mCi is 70 times the presently accepted level for continuous exposure. That is to say, 1 mCi of tritium sustained in the body will deliver to the body of a standard 70-kg man 5.0 rem/yr.

Since tritium is an isotope of hydrogen, and is readily oxidized to water, the earliest concern about the possibility of biological damage centered on tritiated water. Tritiated water behaves in mammalian systems as normal water. In man it is rapidly equilibrated with the other water in the body, which comprises about 60% of the body weight. As normal water is eliminated from the body in the form of urine, sweat, or exhaled air, so is the tritiated water. In man, the half-life of elimination is about 12 days.

At a somewhat later date, concern was expressed over the possibility of tritium becoming incorporated in the various protein structures, and thereby being bound in the body for longer periods of time, that is, exhibiting a longer biological half-life. One particular concern was that of tritium being bound in the DNA and the possibility of enhanced genetic damage. Experiments were performed injecting tritiated thymadine (a precursor to DNA) into small mammals. While it was found that the tritium was bound for a longer period, the concentration required for detectable biological (genetic) effects was found to be at least 1 μCi per gram of body weight. This is the same concentration level required for other biological effects.

One of the factors that enters into the estimate of whole body exposure to man is the quality factor (previously called the relative biological effectiveness, RBE) of the tritium betas. In 1966, the International Commission on Radiological Protection (ICRP) recommended an RBE of 1.7 for tritium beta exposure. It was based on considerations of the linear energy transfer of the tritium beta particle and on experiments done on small animals. In 1969, primarily because of the uncertainties in the various RBE values that had been derived from the small animal experiments, and in the interest of simplicity, the ICRP reduced the quality factor (RBE) to unity. This has resulted in some dissension in the community of health physicists who have to calculate whole body exposures from ingested tritium. Johnson (1971), in England, has reviewed the various RBE experiments with emphasis on the uncertainties involved and has concluded that for radiation protection purposes a quality factor (RBE) of two was probably more appropriate. This was consistent with Russian investigations, which indicated RBE's between 1.45 and 1.98.

At the present time, ERDA is still using the maximum permissible concentrations that the ICRP recommended in 1966. These were based, in the case of tritium, on an RBE of 1.7. Since LASL complies with ERDA regulations, LASL is retaining the use of the 1.7 quality factor.

Tritium is alleged to have been the cause of two human deaths. These will be discussed by Roland Jalbert. Here at LASL we expect to prevent such occurrences by providing appropriate protection for those personnel who work with tritium. The primary protection is provided by containment of the tritium. Any deficiencies in containment should be detected biologically by the routine urine sampling program, which will readily indicate levels less than 10% of a sustained 1-mCi body burden. By using this early detection capability, the Health Physics Group has the option of removing the personnel from further exposure or of eliminating immediately the situations which would have caused higher exposures, had the man stayed in the exposure situation.

Since 1951, when the tritium urine assay program was initiated at LASL, the highest annual exposure recorded for any one person has been 7.3 rem. The average of the highest tritium exposures for the 23-yr period has been 3.3 rem. There have been many tritium exposures below these levels—well below these levels—but these represent the highest that LASL has recorded.

The tritium urine sampling program at LASL works in the following way. All persons who work in areas where tritium is handled in curie, or larger, amounts are scheduled for routine tritium urinalysis either every two weeks, or monthly, depending on a judgment of exposure potential. In addition, whenever a worker himself or a health physics surveyor suspects that a significant tritium exposure is occurring or has recently occurred, the persons involved are asked to immediately empty their bladders. That urine voiding is discarded. Those persons are then requested to provide another voiding about 2 h later. This is the initial voiding. It is analyzed for tritium content, and is used for the initial estimate of the man's exposure.

Following the initial urine assay in the case of an acute exposure, a regime for collecting additional samples has been established. The regime for additional collection depends on the result of the immediately previous analysis as indicated below.

Previous Result (μCi/liter)	Procedure
<1	Submit no more samples until next suspected exposure or next routine (every two weeks) sample for those working where potential exposures exist.
1 to 10	Submit next sample within 1 month of time of previous sample, unless person works in area where potential exposure exists, then submit samples every 2 weeks.
10.1 to 100	Submit weekly samples.
>100	Submit daily samples, including weekends. (Normally this would be the result of an acute exposure which would be individually followed, depending on the magnitude.)

Having obtained urine assay data over a series of analyses, we apply a procedure which we believe makes a reliable estimate of the whole body radiation exposure received by the persons submitting the samples. This computerized procedure examines successive pairs of sample results, calculates the actual elimination half-time between the samples and, using this half-life, calculates the exposure the person received between submission of the samples. The total exposure received over a series of urine voidings is the sum of the exposures between successive pairs of samples.

In applying this procedure, if the second sample of a pair is larger than the first, it is assumed for the whole body exposure calculation that the body burden has remained constant during the period between the samples. Basic to this procedure is the assumption that a significant exposure will be detected on the day the exposure occurs.

In those cases where the urine results remain constant, a quick estimate of the person's exposure in millirem is obtained by the product of one-half the value of the urine results in microcuries per liter times the number of days the urine results remained constant. The procedure for calculating the exposure in the case of diminishing urine results is a little more complicated, and I think I will omit describing it to save some time.

This exposure calculation procedure does give us some problems. If employees do not submit urine samples according to the schedule given earlier, significant overestimates of exposure can result. For example, consider a man whose urine level diminished to, say, 9 μCi/liter and then he left town on a month's vacation. If he did not submit a sample immediately upon returning to work, but waited until his next suspected exposure, say another month later, and the urine assay indicated 10 μCi/liter or more, the calculational procedure would assume that the previous level of 9 μCi/liter was maintained for the 2-month period, and attribute an exposure of ~270 mrem. This result is an obvious overestimate of the exposure actually received. But, that's the amount that would be added to his personal radiation exposure record.

Here at LASL, tritium vapor accounts for most of the personnel exposures. As a gas, $^3H_2$, tritium is not particularly toxic. Considering airborne tritium gas and tritium water vapor, equivalent exposures occur when the gas is 400 times more concentrated. Work of Pinson and Langham in the early 1950's indicated that when a human breathes tritiated water vapor, essentially all of the tritium inhaled is retained in the body. Their experiments also indicated that for prolonged exposures in a tritiated water atmosphere, there is a significant skin absorption. If a man works in such an atmosphere for approximately half an hour, transfer of tritium water vapor across the skin will equlibrate, and an amount of tritium will be deposited in the man's blood stream approximately equal to that inhaled by breathing the atmosphere.

One important source of exposure at LASL results from the handling of tritium-contaminated hardware. Most hardware becomes contaminated by tritium gas occluding to the surface. Over a period of time some fraction of this tritium oxidizes, and the subsequent handling of the hardware results in significant transfer of tritiated water across the skin. Some of the larger exposures have resulted from handling tritium-contaminated hardware without benefit of rubber gloves or some such barrier material.

Changing of tritium-contaminated vacuum pump oil has been another source of large exposures. I was very pleased to see Jim Anderson's technique for changing vacuum oil at DP-East. I hope the technique will be adopted throughout the Laboratory and that we will not have a recurrence of the situation experienced 4 or 5 yr ago at the large Van de Graaff. An employee was changing the oil in a vacuum pump at the large Van de Graaff. He made an effort to be "upwind," but inadvertently spilled a few drops of the oil on his shoe. He didn't notice the slight spillage at the time, and over the next day or so significant tritium was absorbed through the leather into his body. His exposure from this incident amounted to 4.5 rem, or nine-tenths his allowed annual exposure.

In the case of significant exposures, there is something that can be done to reduce the whole body exposure. That something is to increase the body water turnover rate. A standing joke in the early 50's—I hope it was a joke—involved the purchase of a keg of beer to enhance the water turnover following a sizable tritium exposure. It undoubtedly was effective if the exposed employee was much of a beer drinker. By reducing the nominal biological half-time from 12 to 6 days, the total body exposure will be reduced by a factor of two. It is somewhat more difficult to reduce the body turnover time much below 6 days without incurring undesirable side effects. By undesirable side effects I mean kidney damage, not alcoholic inebriation. In the event that an employee does experience a significant tritium intake and it seems worthwhile to reduce his exposure (consistent with policy of keeping all radiation exposures as low as practicable), the Health Physics Group in concert with the Industrial Medicine Group will advise a prescribed regime of fluid intake—not beer, just water.

A problem, which we have not observed at LASL but which appears repeatedly in the literature, concerns the fixation of tritium in the organic constituents of the body. Small animal experimentation indicates an upper limit of 2-4% for the exchange of tritium from water to organic molecules. If fixation in organic molecules does take place, the problems arise because the biological elimination half-time increases, and because urine assays do not indicate the total amount of tritium in the body. This makes the estimation of whole body radiation exposure much more difficult.

Another form of tritium that has been troublesome in recent years is insoluble tritium compounds, some hydroxides. Exposures to this material by inhalation would result in the material being deposited in the lungs. Since it is insoluble, urine assays would not be suitable for determining personnel exposure. To my knowledge, there is no reliable way to estimate a man's exposure after he has worked with these materials. Happily, work with these insoluble tritium compounds has not been undertaken at LASL.

High Pressure - Don Coffin, WX-5

This talk will cover the scope of our experience at TA-33. Since 1955 we have been handling gaseous tritium at pressures of up to 200 MPa. (30,000 psi or 2,000 atm.) The temperature ranges from that for liquid nitrogen (77 K) to the temperature useful for diffusing hydrogen isotopes through palladium or nickel (700 K). This is one of the techniques that is now being used quite a bit. I'll first discuss design problems and materials, and then move quickly through some suitable schemes for compressing tritium gas.

Strength of materials is usually not very critical at low pressures. You can use just about any kind of a metal system from vacuum up to a few atmospheres. Normally, you don't even have to concern yourself about hydrogen embrittlement because your vessels are so overdesigned to make them strong enough to support their own weight that the pressure is not a great additional stress. When you get up to pressures of 200 MPa, however, hydrogen embrittlement is a very real problem, and there are very few materials that you're free to use. Radiation compatability is a real concern with tritium because at high pressures you can get concentrations of 2,000 Ci/cm^3. The average energy of a tritium decay beta particle is about 4 keV, which is sufficient to break down or cross-link any kind of an organic bond. This means you can't use elastomers or plastics, as has already been noted. Thus for static and moving seals you have to figure out how to design without elastomers. Extremely low leak rates are detectable for tritium, and the tolerance for tritium leakage is correspondingly low, so the problem of high pressure leaks is much more severe than for any other material. Tritium is a material that has to be accounted for just as do plutonium or uranium. The only way to assay it at high pressures is to accurately measure its pressure and temperature; so you try to do precise gauging which, in turn, requires stressing deformable elements to near their elastic limits. You overcome safety factors more rapidly than you gain accuracy, so there is a trade-off here that you have to make in your design. Double containment, such as Jim Anderson discussed, becomes a far more difficult problem at high pressure, as will be seen when we look at some of the equipment required for gas compression. Remote control is desirable for several reasons. First, the high pressure itself is hazardous enough to warrant getting operators out of range of the pressurized equipment, and there is the additional hazard of radiation in the event of leakage or failure.

I am going to go very quickly through materials because most of them have already been mentioned. Beryllium copper hasn't been mentioned, and it is a suitable material for resisting hydrogen embrittlement at high pressures. It is a high strength material, and so is 21-6-9 stainless steel, which has all the hydrogen-embrittlement resistance of the 300-series stainless steels, plus being about twice as strong. Mercury hasn't been mentioned, but it has historical importance in some of the compression schemes we'll be looking at a little later. As to plastics and elastomers, we're finding polyimide (duPont's Vespel) to be a very useful material for soft gaskets, valve seats, and packings. We're now using Vespel to replace all our high pressure valve packings, which come standard with a cylindrical Teflon packing. I can't leave the subject of plastics without making a special point about Teflon, Kel-F, and Viton-A. In low radiation fluxes these halogenated plastics may continue to seal for a long time, and we used a lot of valves containing these materials back in the late 1950's. Then we had a spate of metal failures in our system which were ultimately traced to the fact that Kel-F and Teflon, under irradiation from tritium, were decomposing into tritium fluoride, which then attacked the metal components of the system. We were getting holes in metal valve bellows even while the Kel-F seats were still sealing. I'd like to throw this out to Jim Anderson and his people to consider: This is a long-term effect; we had these valves in an atmospheric pressure system for 5 yr without a failure. Then we

started having failures about once a week until all the valves were replaced. At that time, we replaced the Kel-F and Teflon with Nylon, but today we use Vespel whenever possible.

Now we want to go through some pumping schemes for tritium, arranged in historical order. Figure 1 shows an L-shaped mercury pump that was devised by Fred Edeskuty in the 1950's. It has a piston which operates horizontally, stroking a vertical column of mercury, which covers up the seals and protects them from the tritium being compressed. This was one of the early schemes, the earliest of several modifications, and worked very well. Later versions were U-shaped, and pistons were driven by a crankshaft. Figure 2 shows a modern version of the same idea. It is another mercury compressor that is commercially available (Aminco) in which the mercury is pumped with a standard hydraulic system. The main drawback to these liquid metal pumps is that you have to deal with mercury. If mercury doesn't bother you, these are fine pumps. They're simple and reliable, and most of their seal problems are solved. All you have to worry about is hydraulic pressure generation, which is a well-developed art. If you can't stand mercury, and we couldn't because we were often asked to fill devices having copper liners, you have to look for other schemes. It is axiomatic that if you have mercury anywhere in your system, you have it everywhere.

Two other schemes still in use are shown in Fig. 3. If you want to generate high-pressure tritium and you don't have time to order a lot of equipment, anyone with a machine shop can throw together one of these devices. With the cryopump you get a 4-to-1 compression ratio simply by cooling the gas to liquid nitrogen temperature, valving it off, and letting it warm to room temperature; adding an adsorbent (molecular sieve) increases the compression ratio. The metal hydride pump is the scheme being used at CMB-3, where Dean Carstens has developed a vanadium hydride pump with which he can generate 70 MPa. Of course this system works as well for tritium as for any hydrogen isotope, except that tritium decays into helium-3, which eventually blocks the hydriding action if not removed. The other problem with either of these pump designs is that they have to be sized to the job. If you have to fill a large vessel to high pressure, you quickly get into some large cylinders when you consider the wall thickness required to withstand the pressure. A similar idea that's been used is the thermal compressor (Fig. 4). It is simply a series of electrically heated cylinders, which are heated sequentially and

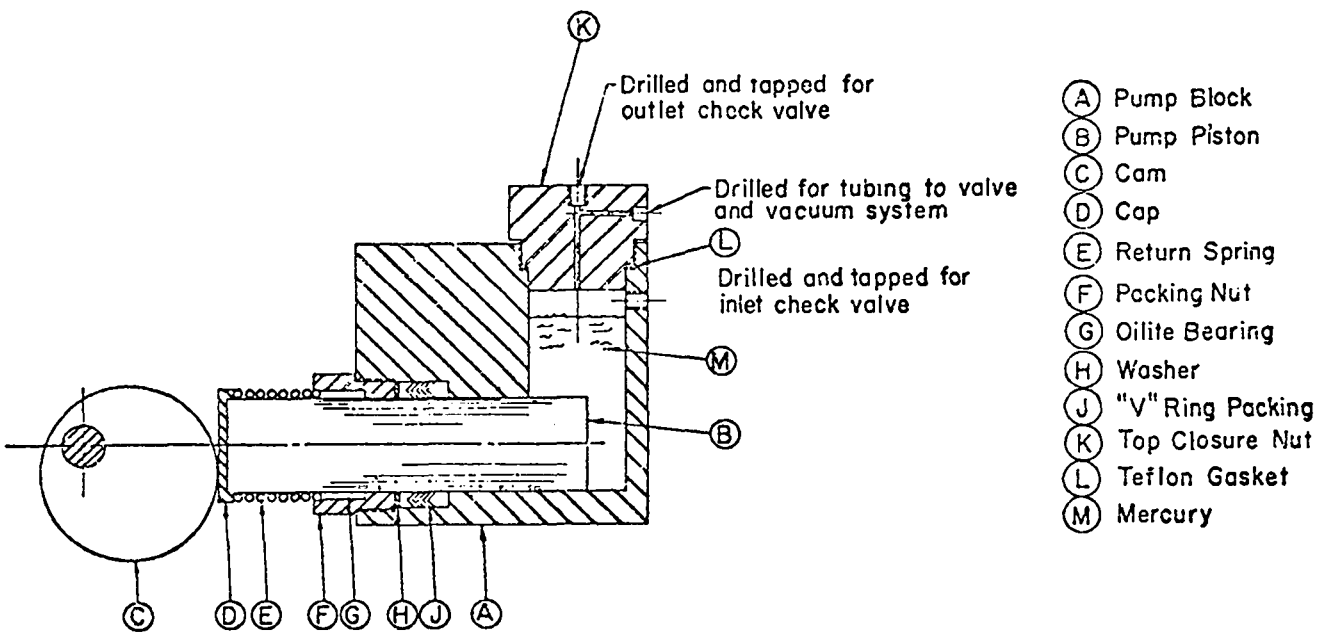
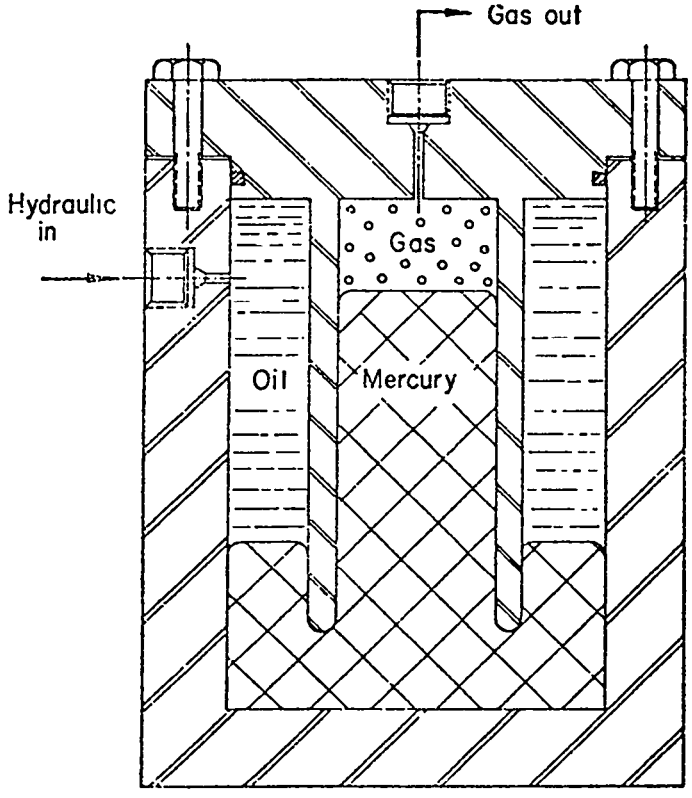


Fig. 1.
Mercury piston pump.



Advantages

1. No moving or static gas seals
2. All pressurization hydraulic

Disadvantages

1. Complex level-interlock system
2. Mercury upstream-materials problem

Fig. 2.
Mercury column pump.

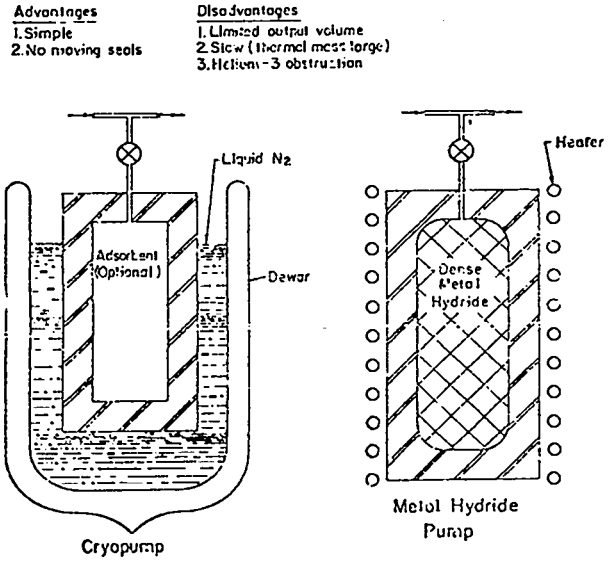


Fig. 3.
Sealless pumps.

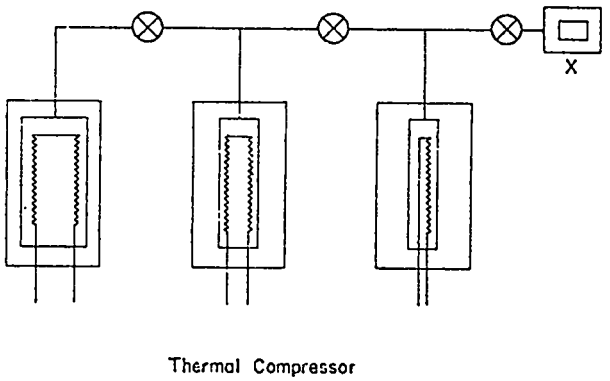


Fig. 4.
Thermal compressor.

valved off. It is reasonable to get about 2-to-1 compression ratios for each stage each cycle. Here the limitation will be the speed, because you have to heat up the cylinders as well as the gas, and then have to wait for cooling between cycles. However, if you have lots of time, this is another very simple system to build because you have no moving seals. The only important seals are for electrical leads, and many methods exist for making those.

The design shown in Fig. 5 is the design that we depend on, and is from my point of view the best way to compress tritium—the welded-diaphragm compressor. The advantages are that it's fast and reliable. When you turn it on it compresses gas, and when you turn it off it stops. The maintenance has been low even in continuous tritium service over a period of 15 yr. The main disadvantage is that it requires interstage check valves because of the speed at which it operates. When you have check valves, you have only two problems—if they don't work, and if they do. If they don't work, you have to replace them. If they do work, tritium is still trapped in all stages when you're through pumping, because the check valve closes below a certain differential pressure. We have ideas for solving both of these problems. Our check valves do work all the time now, due to a changeover to Vespel for the cage gaskets as well as for the poppets themselves. We've not had a check valve failure since we've gone to this new material. With only elastomers or Teflon, we previously had to replace poppets every few weeks in tritium service. One other tritium pump which we've built, although it is not in service yet, is shown in Fig. 6. It is a bellows pump which works on the same principle as the one Bob Sherman discussed. This one is a balanced design in which the bellows is driven hydraulically, so that there is never any pressure difference across the rather fragile bellows. It displaces about 50 cm^3, and a block is welded inside it so the dead volume is near zero when the bellows is completely compressed. We hope to attain something like a 50-to-1 compression ratio.

Figure 7 is a general picture of our overall system. The basic device is a four-stage diaphragm compressor which by itself is capable of pumping from about 1/2 atm to 2000 atm. On the output manifold is a one-stage bellows pump, which gives the system the capability of going to higher pressures if one of the stages of the main compressor is not working. Another capability of the bellows pump is much finer pressure control for filling small things, because we can fill the bellows pump and stroke it very slowly to get to the exact pressure that we want.

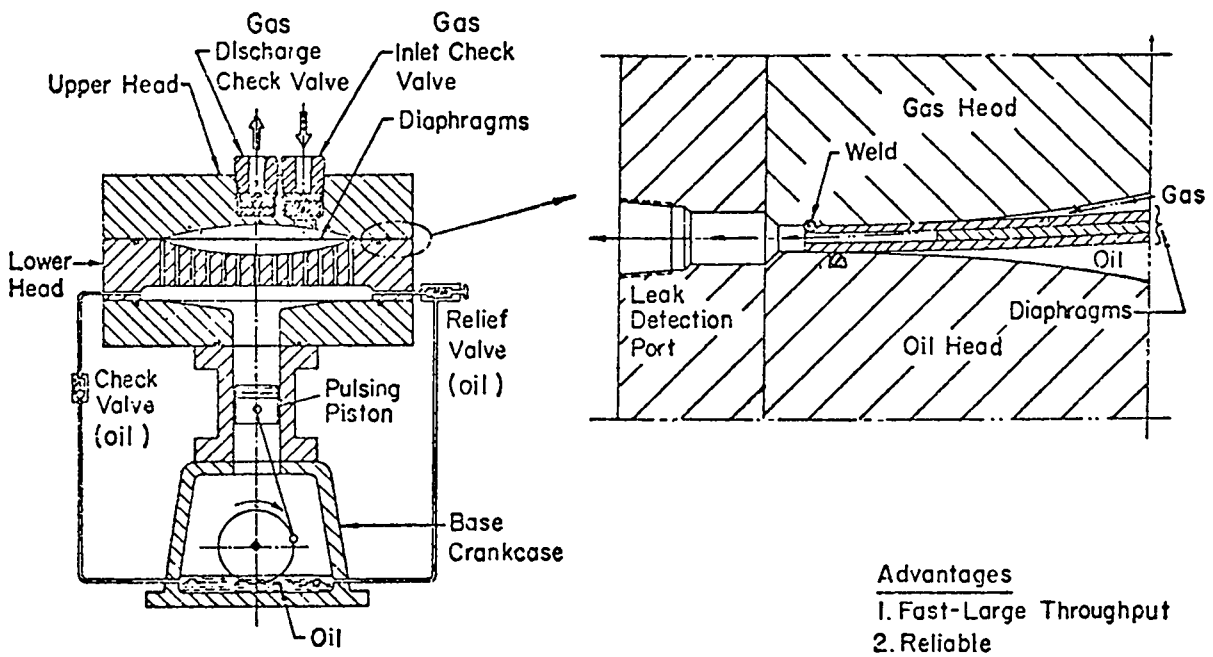


Fig. 5.
Diaphragm compressor.

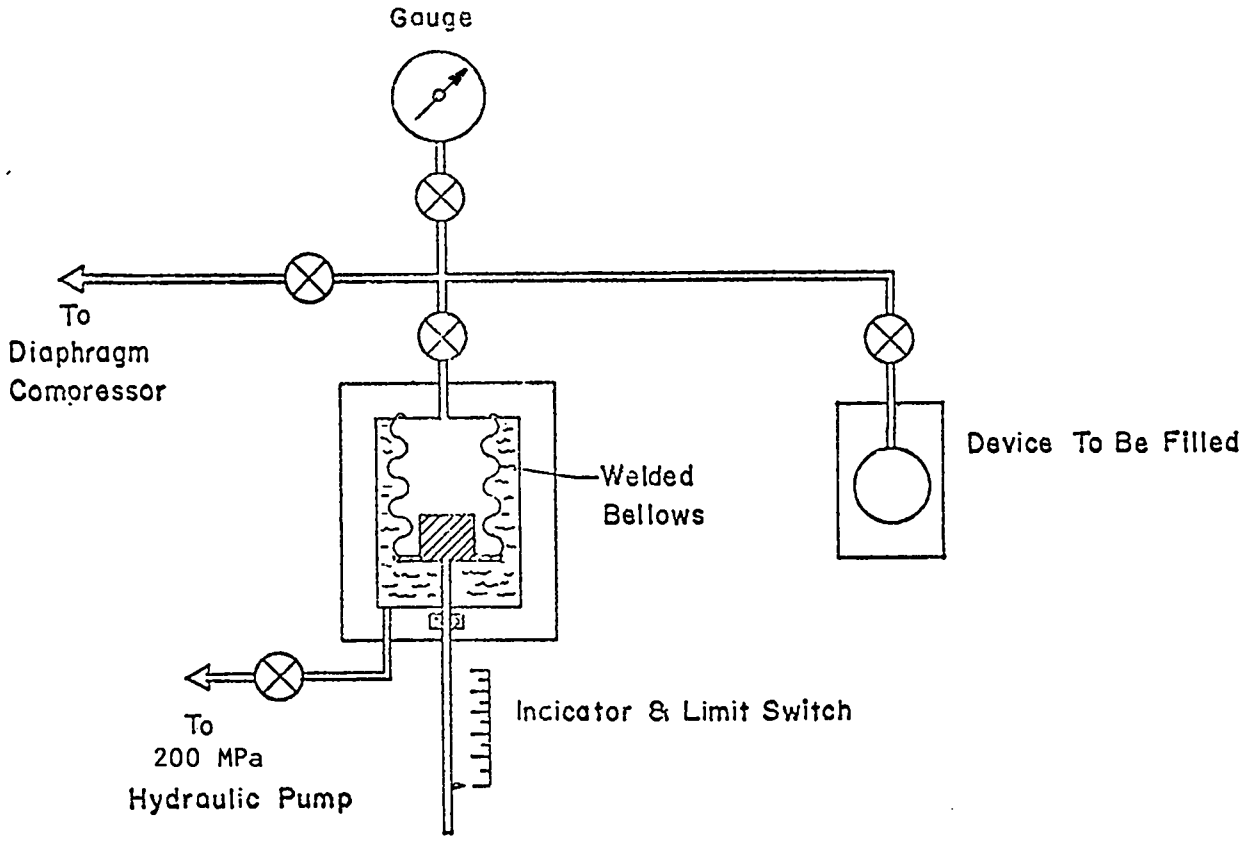


Fig. 6.
Bellows compressor.

Also on the output manifold, in addition to a selectable precision gauge manifold, we have two schemes for containment. One is a simple high-pressure commercial valve with our own remotely controlled valve operator. The other is a welded-closure scheme in which the thick-walled capillary tube through which we fill is sealed with a resistance pinch weld. This gives us an all-closed system with no valves or packings. These welds have leak rates of the order of 10^{-13} cm^3/s (1 pCi/s) or less.

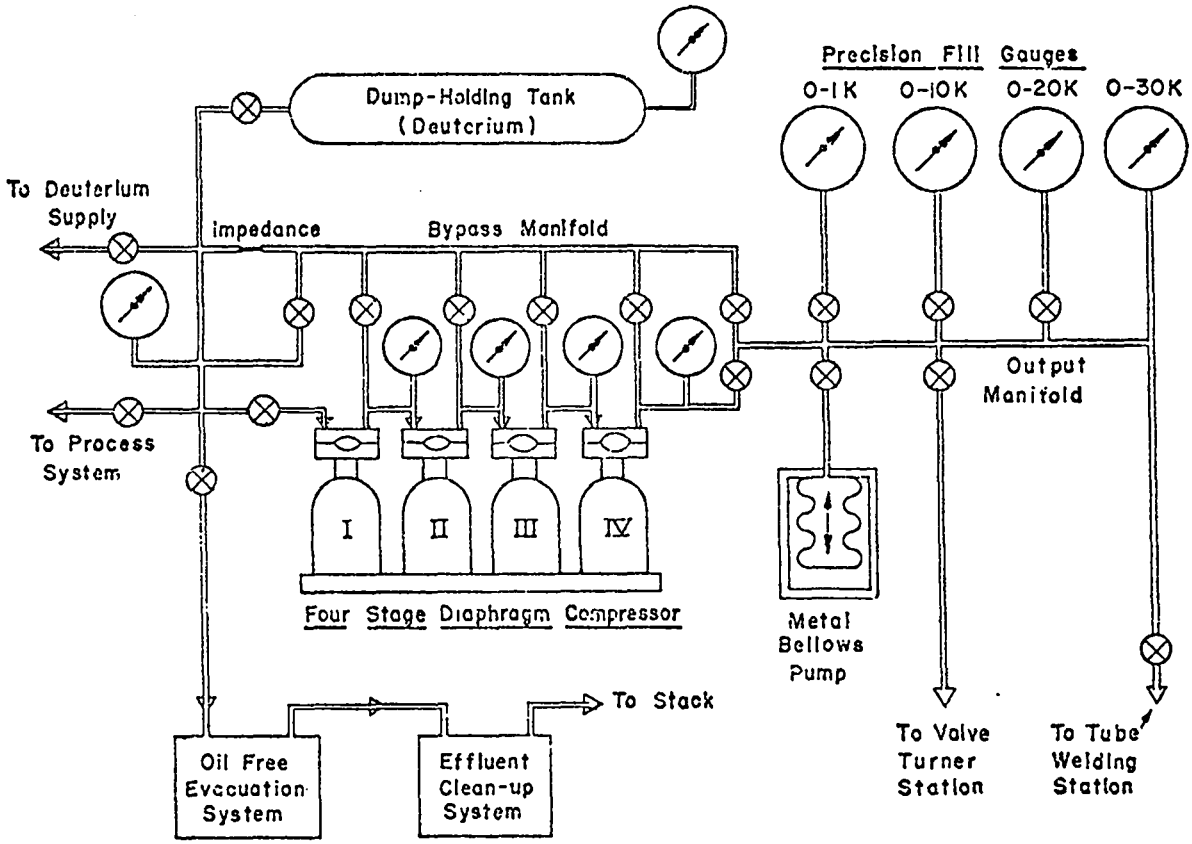


Fig. 7.
Pressure system schematic.

Operating Procedures (High Pressure Tritium Experiment at LLL) - J. D. Seagrave, P-DOR

I'm not sure whether Roy put me on the program for a comic relief or to represent the fact that a naive user at this Laboratory can request large quantities of tritium and depend on the here-assembled experts to survive, or whether it was to provide insight into how things are done at a different laboratory.

As a point of reference, in 1952 when I first came here, the setup shown in Fig. 1 was a bulk tritium experiment. The safety considerations were not much more than the set of shoe laces that backed up the wires holding the meter-long high-pressure transmission cell. I remember that since I was new, I got to change the sample. It was warm to the touch and was filled at about 14 MPa.* I think the pressure test consisted of running it up to 28 MPa of helium: it didn't leak, so that was good for 14 MPa of tritium. The other provision was that when the machine wasn't running, we used a big tank connected to a pump. We wrapped the meter or so of hypodermic tubing around the sample, stuffed it all in the can, and I sat on the lid until the pumps took hold. And of course it was all very secure, because at this time 2 moles of tritium was a classified quantity and there were security guards around to guarantee that it was a safe experiment. Later, things got harder.

In 1960 we tried to again with thin-walled (0.25 mm) stainless steel spheres in which we had a condensed form of tritium—calcium tritide. We had about a quarter of a mole of T_2 at that time. We also did a ^3He experiment with a high-pressure cell. In 1967 and 1969, through the good offices of Bob Sherman and Gene Kerr, we built a marvelous chariot to contain 1 mole of liquid tritium. We had then the first SOP, I think you'd say. However, I'm not going to talk in detail about that. It consisted of one nice plumbing diagram, three pages of description of it by Gene Kerr, eight pages of procedural outline and call lists, and 82 pages of correspondence with Ted Ehrenkranz. Now this went off, it's only fair to say, just beautifully. I think it was right **after** that that the SOP requirements really got stiff. Nonetheless, the care with which that equipment was prepared and checked out stands in testimony to the skill of the people on whom we depended.

*1 MPa = 10 atmospheres.

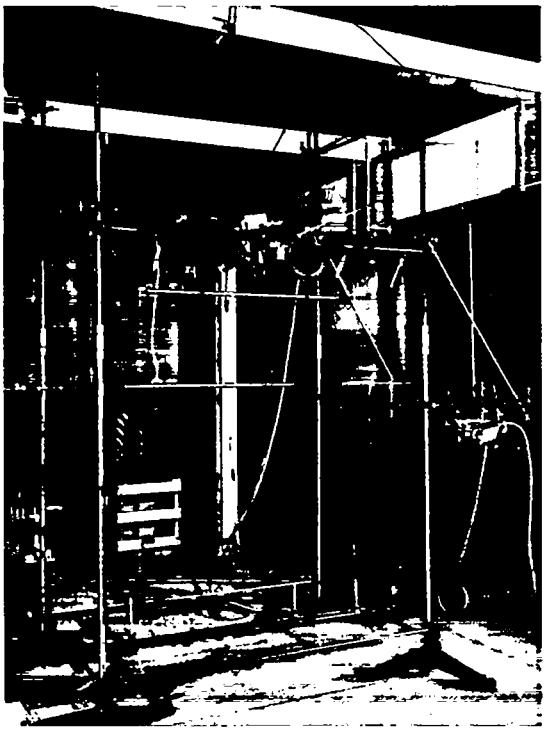


Fig. 1.
Tritium transmission cell setup at P-9 in 1952.

In 1972, I learned that the people at Rensselaer Polytechnic were doing a transmission cell measurement with ^{4}He, and I asked them "Why don't you do it with ^{3}He, too?" And they replied, "Well, you just bring the 'duck' and we'll do it." And that's what we did. Things went without the slightest hitch, so smoothly that I seriously proposed (not knowing what I'd get into): "Why don't we do it with tritium?" We wasted about 6 months talking to the AEC and the State of New York—mostly about en-environmental impact—and gave up. By that time Livermore's Linac was also available to make a continuum source of neutrons from an electron target source. The Bureau of Standards had one too, but that was in an even larger center of population. We then undertook what turned out to be a 2-yr program of getting ready to do at Livermore what we'd just done at Rensselaer. The Livermore Linac is all underground, and except for a building that contains most of the RF generating power supplies, and the offices and control room, about all one can see is a "silo." The electron beam comes along horizontally underground and shoots straight up to hit the target in the silo. Then for neutron time-of-flight studies there's a 250-m evacuated tapering flight pipe, and a second flight pipe which we used for a supplementary neutron beam monitor. The philosophy that went into proposing this experiment was in three parts. First, the 3-1/2 moles of tritium would be outside the main building (we cheated a little and built a new thin-walled shack, with a removable roof to get the stuff in and out); the shack was located just outside of the main source silo and connected only by a collimating system. *Second*, there would be a total containment of the high-pressure cell. This was a meter long 2.5-mm-diam cell for a design pressure of 17 MPa contained in an evacuated container, collimated with thin windows in line with the beam. If the pressure cell containing the tritium and the reference cell containing either hydrogen or deuterium should both completely vent in the container, the internal pressure would still be below 1 atm. Livermore was very fussy about the metallurgy and required an elaborate procedure of welding, testing, and certification. The *final point* was that we would design the hardware and Livermore would test it, which was a nice touch: nobody had a vested interest in how the tests went. (Since they were going to hold the bag, they were going to test it pretty thoroughly.) Figure 2 shows the key element of the transmission cell design: an ellipsoidal end which was built in the shops with a computer-programmed mill following the mathematical relationship shown. This material is a 0.75-mm wall, 21-6-9 stainless steel of a special heat-treat that Livermore had good experience with; we were able to get some chunks of it to make the transmission cells. They say that "21-6-9" material is very spotty; some heat-treats will be full of holes. This was why we had to stick explicitly with the material they had specified. We also built a series of test cells (Fig. 3). This was a little shortie which was intended to ascertain the burst strength of the ellipsoidal end. We made 10 ends. We needed three transmission cells—that took six ends—and we had the rest to make four ellipsoidal-end burst-test cells. We made some straight cylinders with the belly-band weld, and then we made some heavy-walled-at-both-end- cells for burst pressure tests of the weld. Figure 4 shows one of the cells with its classical petal failure at something over 96 MPa. The design pressure was 17 MPa. All of those tested had a safety factor—burst to design pressure ratio—over 5. The stress analysis was made by Harry Luke of WX-1 and it seems to have been confirmed in almost all details. Any minor discrepancies were on the conservative side, so we were happy with the situation. Figure 5 shows the sample changer which we referred to as "the garbage can"1.5 m long, 35 cm in diameter. It contained the three cells: the sample, the reference, and an evacuated dummy. There were 0.25-mm windows at each end of the containment vessel. Also

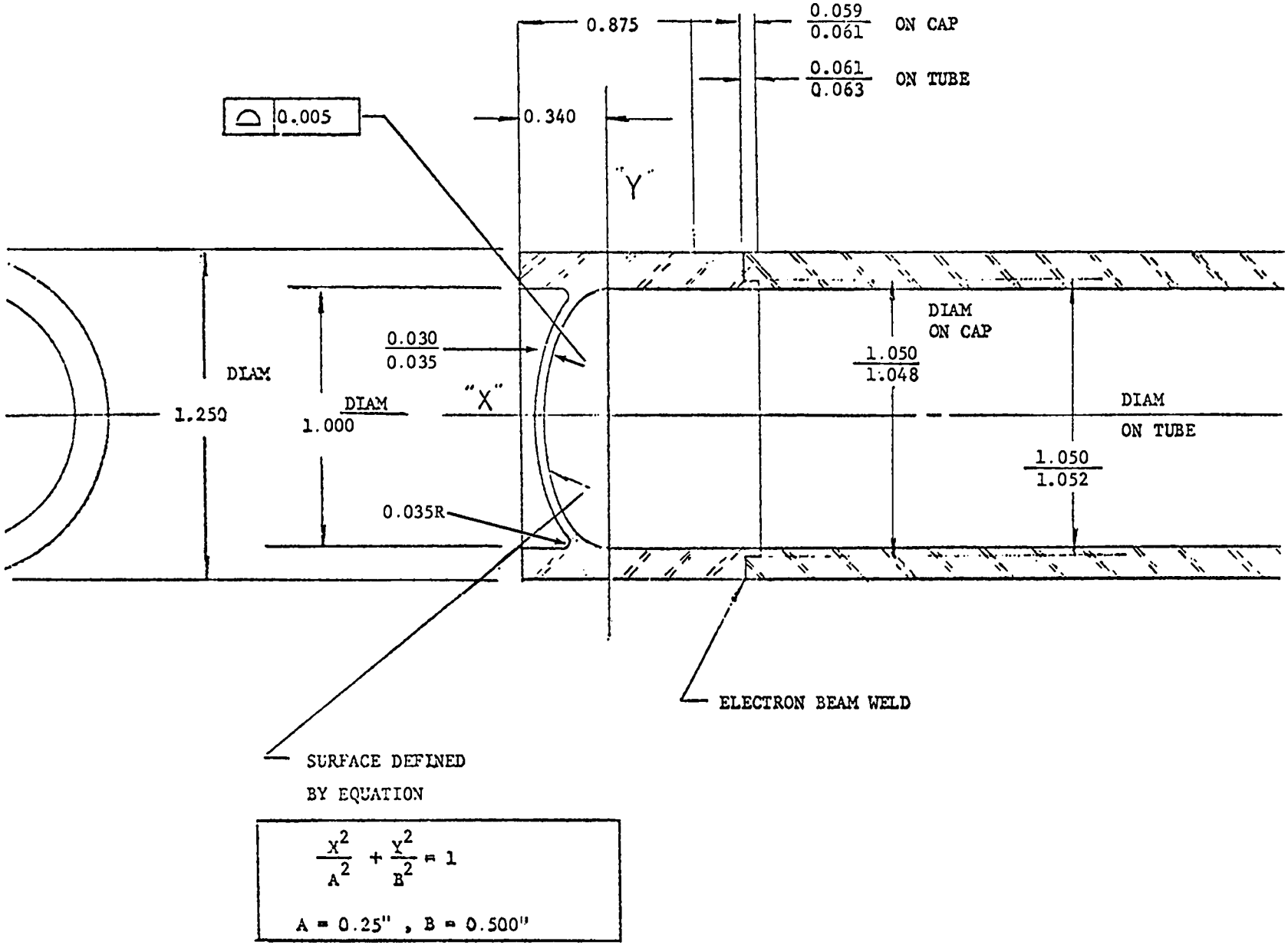


Fig. 2.
Drawing of ellipsoidal end of transmission cell.

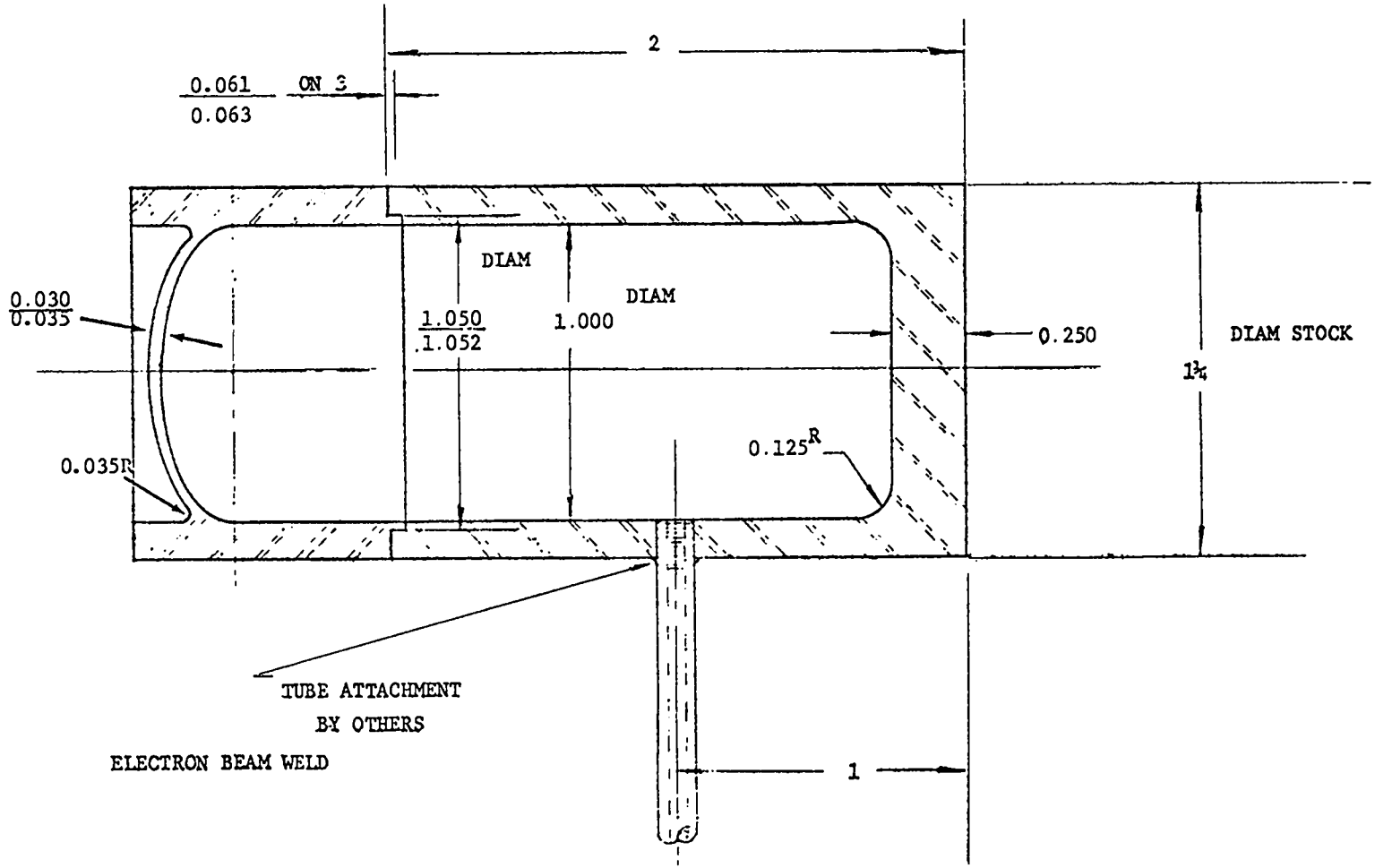


Fig. 3.
Drawing of burst-test cell.

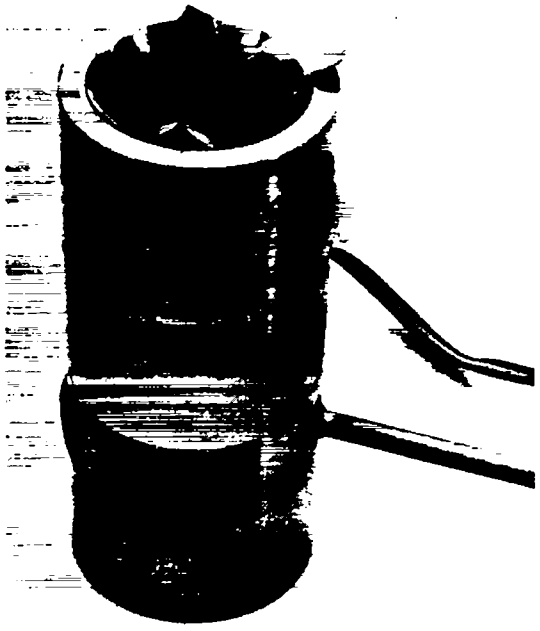


Fig. 4.
Rupture of Test Cell "F."

shown is a pneumatically operated geared-up system for rotating to the three different positions. Figure 6 shows the other end, at which there was a large access port so that the filled cells with their double valves could be placed within the chamber. There

Fig. 5.
Rotating total-containment sample changer.

Fig. 6.
Sample-changer with cells installed, viewed from access port end.

were only the flange closures and one chamber-evacuating pumpout opening. Once the tritium was in there, it was locked in and the rules required that if there was any rise in pressure we were to pack the whole thing back to the tritium handling facility. We did not have to do that, but there were a lot of dry runs and exercises on how to do it. A quick chronology (Fig. 7) shows the time scale of things. In 6/72 we did the experiment at Rensselaer and did not get around to deciding to try to do the one at Livermore until early in '73. Harry Luke made the stress analysis on which the chamber design was based. We agreed to use the special heat-treat 21-6-9 material as required by Livermore. We designed the pressure vessels on that basis and Harry's design. We got the stock and fabricated the basic parts. Livermore made some preliminary tests and revised slightly the electron-beam welding specs about the joints, so that all the rest of the samples then corresponded to the best possible experience. We sent them the 10 elliptical ends. We used the thinnest and least uniform ones for the test cells. Then we made the open tubes and the closed ones and started the changer design. That design then was finished but cost-estimated too high, so we changed the materials here and there and revised it a bit more. The changer was fabricated by Christmas 1973, but it took another year to get the experiment run. The documentation on the stress analysis and miscellaneous calculations proceeded. Then Livermore made the tubing and the heavy cell tests. Finally, all design safety factors proven, we made the sample cells. The first draft of

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TRITIUM TOTAL CROSS-SECTION
CHRONOLOGY EXPERIMENT

6/72 $\sigma_T(^3He)$ at RPI
2/73 $\sigma_T(^3T_2)$ Proposed at LLL
Stress Analysis by H. Luke (WX-1)

3/73 Special Heal No. 21-6-9 required by LLL

4/73 Pressure Vessels and Test Cells Designed

6/73 Stock Obtained and Sampled
Fabrication Starts

6/73 E. B. Weld Dimensions Specified by LLL

7/73 10 Elliptical Ends Fabricated

8/73 3 Open Tube, 3 Closed Cell Weld Test Units
Sent to LLL

9/73 Changer Design Started

10/73 Changer Design "Finished," Estimated,
And Revised

11/73 Changer Fabricated
Axles Reworked, Shipped to LLL

1/74 Stress Analysis and Designs Documented

2/74 Tube and Heavy Cell Weld Tests

4/74 4 Thin Cells Tested at 5.3-6.2 Safety Factor
3 Sample Cells Fabricated and Tested
First Draft of Safety Notes

5/74 Alignment and Dry Run

9/74 SOP Draft

10/74 Revised SOP and M.E. Safety Notes

11/74 D_2/H_2 Run
12/74 Final Run With T_2

Fig. 7.
Chronology of the LLL tritium total cross-section experiment.

the safety notes were next prepared, which partly involved just recollecting this documentation that had been building up. We made a collimator dry run, completed the SOP draft, revised it a little, and brought it up to date with our experience. We made a dummy run with deuterium substituted for tritium, using hydrogen as the reference. And then in December 1974 we made the final run with tritium. It went without incident except for one comical one. The list of people to call in case of emergency was made available to the fire department, and in the middle of one night (things were running very smoothly) there was a "tritium" alarm. The first thing that happened was that the girl at the fire station called all **50 people**—all the way to Berkeley—and they started showing up in phalanxes. The only real problem was clearing the people out of there to get the experiment going again. It was sort of an overkill but, gee, they'd got all this training and practice so the dogs had to come out and bark a little bit at three in the morning. The only real hangup was the automatic shutdown of the house ventilating system which was something we'd missed in the first draft of the SOP (I remember pointing it out to them). It turned out that the data computer started getting hotter and hotter during all the standing-around and talking about it, which may have invalidated some of the data. But that was a small price to pay for the assurance that the emergency SOP worked. The details of what went into the SOP are available if anyone wants to consult them.

The outline (Fig. 8) of what was called the Mechanical Engineering Safety Notes-documentation of the design, with names of individuals responsible for the design calculations, specimen preparation, proof tests, welding tests, microphotographs, etc.—the whole thing became Appendix B of the main SOP which is outlined in the last table (Fig. 9). It looks more complicated, but it has only the obvious things: the scope, who's responsible, what the hazards are and what to make of them, what specific hardware preparation is necessary, what controls there are, what alarms, and who's responsible for seeing that everything is worked out, that emergency call list (which worked only too well), the mechanical notes, some details about how the tritium was to be handled, and the call list in case there was any kind of problem.

I was impressed that when the Fire Department Supervisors came around to be briefed on the nature of the scene they were carrying a portable color TV recorder. They did short interviews with the chief perpetrators and other people you might find in the middle of the night running around the place. The

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TRITIUM TOTAL CROSS-SECTION EXPERIMENT STANDARD OPERATING PROCEDURE - APPENDIX B
4/74 MECH ENG SAFETY NOTES - REVISED 10/74

I. SYSTEM DESIGN
- A. Pressure Vessel Description
- B. Hazards
- C. Stored Energy
- D. Calculations
- E. Associated Components
- F. Fabrication and Welding
- G. Proof Testing
- H. Safety
- I. Comments

II. APPENDIXES
- A. Stress Analysis
- B. Sample Changer and Containment
- C. Self Heating
- D. Material Certificates
- E. Weld Qualification
- F. Weld Inspection
- G. High-Pressure Test Procedures
- H. Cell Test Summary

Fig. 8.

Outline of the mechanical engineering safety notes.

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TRITIUM TOTAL CROSS-SECTION EXPERIMENT
9/74 STANDARD OPERATING PROCEDURE - REVISED 10/74

I. SCOPE OF THE EXPERIMENT

II. RESPONSIBILITIES

III. HAZARDS

IV. HAZARDS ANALYSIS

V. PREPARATION OF HARDWARE
- A. Sample Changer
- B. Transportation
- C. Sample Holder
- D. Sample Changer Stand and Building
- E. Safety Controls and Alarms

VI. OPERATIONAL CONTROLS
- A. Filling and Loading of Samples
- B. Pressure and Radioactivity Checks
- C. Console Controls
- D. Local Alarms
- E. Area Controls
- F. Security Controls

VII. HAZARD CONTROL RESPONSIBILITIES
- A. Fire/Explosion Safety and Protection
- B. Checks of Safety Devices and Alarms
- C. Verification of System Integrity
- D. Overall Hazard Analysis
- E. Instructions

VIII. EMERGENCY CALL LIST

IX. APPENDIXES
- A. Health Physics Calculation
- B. Mechanical Engineering Notes
- C. Tritium Handling
- D. Tritium Monitoring

Fig. 9.

Outline of SOP for LLL tritium total cross-section experiment.

entire fire department subsequently had the benefit of walking through interviews with the personnel and an explanation by the scientists as to what they were trying to do without 50 people having to get it individually or in too large a group. When we were running, those tapes were shown again to each shift. It wasn't very long, perhaps 10 min of edited tape. It certainly seemed to me a valuable resource. At that time LASL didn't have such a thing in the entire Laboratory, let alone in the Fire Department. Videotape is a very useful tool to make personal contact and communicate to the emergency personnel what it was the experimenters were trying to do when something went wrong.

Now to conclude I'll show a few (color) slides of how the operation actually went off. In Fig. 10 is the blue garbage can sitting on a stand. There's a computer control that permits this pneumatic system to make all the changes by complete remote control. There's a 10-mm-diam collimator through the 3-m shield wall, collimating the beam so that it sees only the 0.75-mm ends of the chamber and the two 0.25-mm ends of the containment vessel. There is a big monitoring carbon sample in the beam and the neutron detector overhead. A pressure gauge is also visible. Figure 11 shows the same scene from downstream looking back, showing the drive mechanism geared up. At the top is the beam position with a thin window. The windows had 0-ringed brass plugs; when the changer was handled, the windows were protected by a heavy shield. There are three television cameras just barely visible that look at the gauge system in all three positions so that at no time were we out of direct visual contact with the position and the gauging. In addition, there was an electronic transducer which was connected to alarms and abort interlocks. Figure 12 is a view looking downstream from the changer. There is another thin window in the 250-m flight path. The tritium monitoring systems are at the left, also interlocked. It was one of these that plugged up its airflow in the middle of the night and set off that false alarm. Figure 13 shows the 250-m booth. The flight path, you'll notice, has enlarged considerably at this distance. There's a "six-pack" of photomultipliers located around a scintillator package. The big window in the flight tube was necked to a somewhat smaller diameter and then closed with a 1.5-mm aluminum diaphragm. It is a man-swallowing aperture when the flight path is evacuated. The procedure required that we crank in from outside the building a heavy aluminum shield before entering because if that diaphragm ever failed, it would suck up the apparatus and experimenter and put them through two or three collimators on the way back upstream. Figure 14 shows the monitoring beam pipe, which is much simpler. The corresponding pipe window is thinner and is outside the building. Another carbon disc scattered monitored neutrons into the

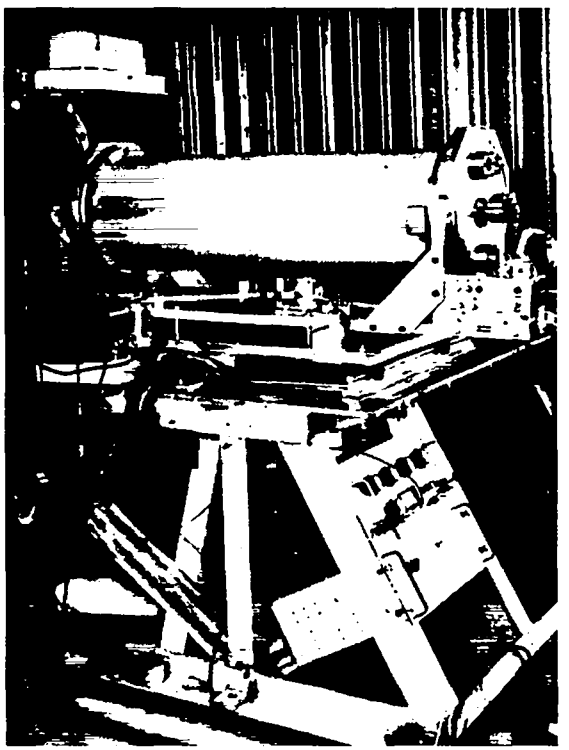

Fig. 10.
Sample changer on alignment stand, with controls and detector.

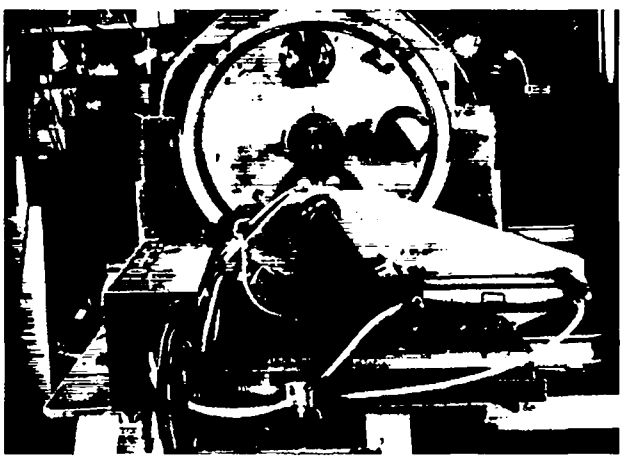

Fig. 11.
Sample changer viewed from downstream.

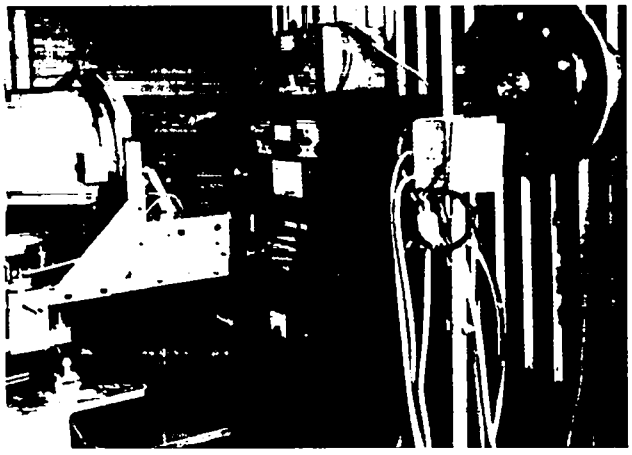

Fig. 12.
Downstream view from the changer.

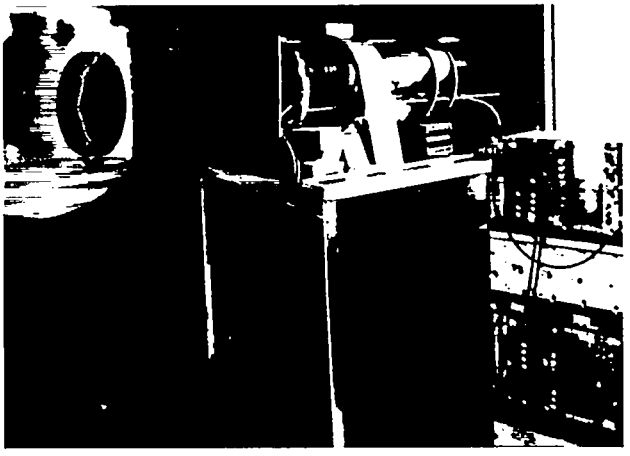

Fig. 14.
Upstream view of monitor booth, showing graphite scattering sample.

detector—an experimental technique to get a better signal- to-noise ratio. Figure 15 shows the conclusion of the experiment: a quarter of a million curies going out the roof. (I thought you'd want to see that.) The rigger didn't, but the operation went very smoothly. Notice the peculiar wheels. These were recirculating ballbearing sleeves that slid sideways, so that for some alignment operations we could move the apparatus out of the beam, align the collimator with a laser, and pull the sample changer back to a definite stop. And finally, Fig. 16 shows the crane with the roof off and everybody breathing a big sigh of relief. That's the travelogue. I do have both the 96 pages of notes from the previous operation and the detailed specs of the recent experiment the way they're written up for Livermore's requirements, if anybody wishes to avail himself of them.

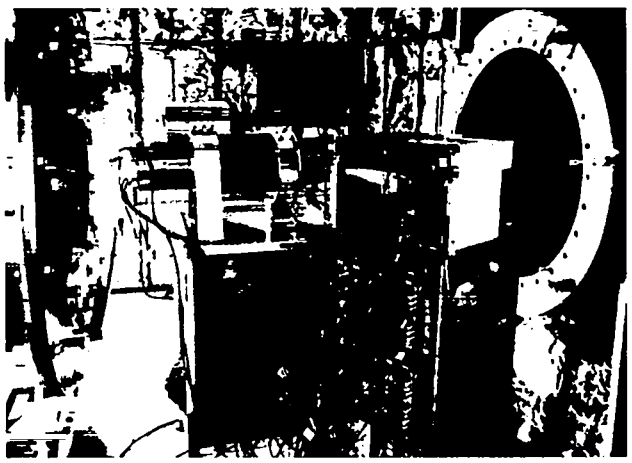

Fig. 13.
Detector "Six-Pack" in 250-m flight booth.

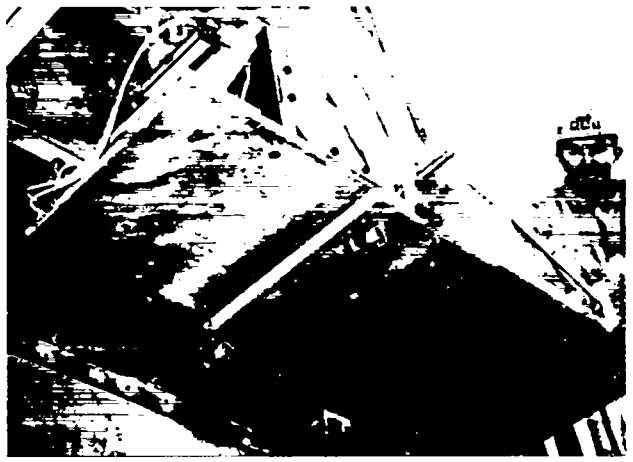

Fig. 15.
Removal of the sample changer through the roof of the shack.

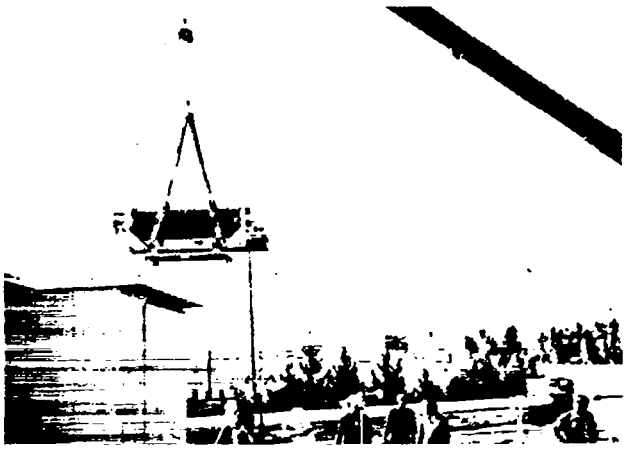

Fig. 16.
Crane moving sample changer.

Incidents - R. Jalbert, H-1

If I were to receive a call that we just had a tritium incident, I would wonder immediately in which category it would fall: would it be one in which we had umpteen curies of tritium gas that went up the stack with little or no measurable or calculable exposure to anyone either within the Lab or outside, or would it be one that would involve not the gas but the oxide, with perhaps a few people receiving a significant exposure. Here at Los Alamos we have had our share of both.

In the first category, our gas releases have been not too bad; on the order of a few hundred curies maximum. Years ago we might have exceeded this by a factor of 10 or so. In the other category, our exposures haven't been too serious either. Jim Lawrence mentioned a high dose equivalent of something like 7 or 8 rem. Compared to the experience of others, ours haven't all been that exciting.

Elsewhere, Livermore and Savannah River in recent years have released a little bit more gas than we have, on the order of 5 ft^3. This is the approximate number originally reported to the press in the case of the Livermore release. As far as exposures to personnel are concerned, the ones of greatest interest, and the ones that Jim Lawrence alluded to, are the two tritium deaths in Europe about 10 yr ago. I thought that I would talk about these rather than describe how a valve failed and curies were released and went up the stack, and where the most serious biological effect was the color change of the public relations man's hair from black to white.

The two deaths which I'd like to talk about occurred about 10 yr ago, within a year of each other, in two European countries. Both occurred as a result of work in factories which were engaged in the manufacture of luminous paint. Both companies had been using radium and thorium, later switching to strontium and, at least in one case, promethium, and eventually tritium.

The first case involved a chemical engineer who had worked with radium and thorium for roughly 18 yr before switching to work with strontium and promethium for about 2 or 3 yr. Then, when he was about 60, he started working with tritium, which he handled for about 8 yr. During the first 4 yr, he worked with only about 600 Ci, whereas during the last 4 yr he handled about 7000 Ci—a total of about 7500 Ci. Apparently he was a pig-headed individual. He refused all medical counsel to quit work when it was discovered that he had been exposed and had already received a fairly high dose. He continued to work almost until his death. He terminated his job on October 14, 1964, entered the hospital on November 3, and died 4 days later after one blood transfusion. The health physics must not have been very good. Apparently the tritium monitoring was nonexistent in the early years and possibly during most of this period. This is suggested also by the fact that urinalysis was not begun for the man until early 1963, the year before he died. He worked 6 yr with tritium without any urinalyses. In the first of those last two years his urine tritium levels ranged from 140 to 1120 $\mu Ci/\ell$, and, in the second year, 50 to 800 $\mu Ci/\ell$.

It is difficult to compute the dose that he received from tritium because it is based on the data of only the last 2 yr. However, this detail does not hamper the intrepid health physicist faced with this task. It is estimated that the man received somewhere between 300 and 600 rem. This dose was spread out over 4 to 8 yr, but most of it in the last 4 yr. This dose includes that due to approximately 7 to 14% tritium incorporation into the tissue, which Jim Lawrence alluded to. This fraction is much lower in an acute exposure and becomes higher in a chronic exposure, such as this one. Blood samples were taken, and in 1961 (about 2 yr before his first urinalysis) his white blood count was normal and remained so until just before his death. His red blood count, however, decreased steadily. His hemoglobin also dropped, but not quite as rapidly as the red blood count. He died of an advanced case of panmyolopthisis, a degeneration of the blood-forming elements in the bone marrow.

The first question that comes to mind is "What about the radium?" since he had worked with radium for about 18 yr. It turned out that his body

burden of radium was 3 to 4 nCi, compared to the maximum permissible body burden of 100 nCi. The next question concerns strontium. Unfortunately, we have no data on strontium, so we don't know to what degree strontium was incorporated in his body. Radium is a bone-seeker and so is strontium. Although tritium is not a bone-seeker, it does get into the bone marrow, since there is water there, and is exchanged in the marrow with organic molecules. The concentration of tritium in the bone marrow was higher than the concentration of radium in the bone marrow, but the disintegration energies are different. Other factors also make the comparison difficult. There could be a synergistic effect there between radium, strontium (if there was any), and tritium. In any case, here is one death that can probably be attributed, at least in part, to the incorporation of tritium in the body.

Parenthetically, three other people were involved in that case. One, a young lady, age 22, worked with him for several years and quit. Her dose was estimated to be on the order of 100 to 300 rem. She suffered from moderate hyperchronic anemia. Two other individuals continued working in the plant for a year after this fellow died. The health physics for these two apparently wasn't much better—they incorporated something like 189 μCi/ℓ in body water. The dose was not computed. They suffered no ill effects. Finally, the plant was shut down; or at least that operation was stopped.

Case number two, in a different country, was somewhat similar. Data are even more skimpy, however. In this case, a man went to work in 1959. He worked with radium and strontium for 2 yr and switched to work with tritium in 1961. Again, the health physics must have been poor. I might mention that in at least one of these two fatality cases, the man did wear a film badge, probably a holdover from the radium days. The film badge showed an apparently hard gamma exposure when he was working with tritium, which is a soft beta emitter. However, that can be explained by the fact that tritium will infiltrate the film badge and result in a blackening of the film, which then looks like a hard gamma exposure. The film badge may have been the only radiation protection that they had at the plant. Anyway, this fellow started working in 1961 with tritium and although we don't know exactly how much he worked with it, it looks like several thousand curies spread over about 3 yr. Urinalyses were begun 32 months after he started working with tritium. He quit several months later, in May 1964, and he died in mid-September 1965. During the 3-month sampling period, the tritium in his urine ranged from 53 to 117 μCi/ℓ, and was 70 μCi/ℓ when he quit work. Eighty-four days later, at the time of the next urine sample, the tritium level in his urine was 5 μCi/ℓ. Twelve days after that it was 29 μCi/ℓ. Here we see examples of some fairly wild fluctuations of urine tritium levels **after** the tritium incorporation had stopped. This was caused by tritium from the organic molecules feeding into the body water as a result of metabolic processes. These fluctuations continued until his death.

The estimate of the dose is even more difficult in this case because we have only that little bit of data for a few months and we have to extrapolate back almost 3 years, but that doesn't stop us. One estimate is something like 300 to 1000 rem, and another is somewhat higher, 1000 to 2000 rem. He died of the same disease as in the first fatality case.

At his death, his radium body burden was roughly 90% of the maximum permissible body burden—about 90 nCi. Two other people were involved, and we have little data on them, except that one had slightly over the maximum permissible body burden of radium.

In both countries, the governments' concern or control must have been very slight to have allowed this to occur. At Los Alamos we started urinalyses in 1951. We were monitoring for tritium with reasonably good instruments in the early 1950's. Our instruments were LASL-developed because no reliable instruments were commercially available. In those days, if we had had to rely on instrument suppliers, I think we would have been in a much worse way. But we built our own, which a private company can't easily do.

In conclusion, I wish, first of all, to thank Jim Lawrence for allowing me to infringe on his territory. Secondly, I did Xerox the report on which most of this talk is based, a report presented at the Tritium Symposium in Las Vegas several years ago. I have a few copies if anyone is interested, and I can make more if I don't have enough.

Emergency Planning - Roy Reider, Safety, H-3

Thank you, speakers, for your participation. We expected about a dozen people, with the principals taking in each other's wash, but there are 115 people here. I'd like to emphasize that you must have an SOP for tritium work; H-Division people will help you with it. I'm sure all these other people who have used one, Sherman, Coffin, Seagrave, and Anderson, would be happy to help you. You now know the people who gave these presentations, so if there's any need for information you can get it on a personal basis.

In emergency planning, dollar values must be considered. The unclassified value of tritium (see Fig. 1)

HYDROGEN

HYDROGEN-3 (Tritium) (12.3 years)

Produced by $^{6}Li(n,\alpha)\ ^{3}H$

Theoretical specific activity: 9.6×10^{3} Ci/g ^{3}H

Shipped as gas containing small amount of ^{3}He daughter at a concentration of 2.59 Ci/ml at STP.

Radiochemical purity: >99%

Up to 10 Ci shipped in glass ampules equipped with breakoff seals and packaged in nonreturnable container; larger quantities shipped in returnable metal cylinders.

PRICE:	Minimum order	$25.00
	0- 1,000 Ci	2.00 per Ci
	1,001-10,000 Ci	1.50 per Ci
	10,001-25,000 Ci	1.00 per Ci
	>25,000 Ci	0.75 per Ci

Plus $30.00 packaging charge per ampule or cylinder.

Fig. 1.

in small amounts is about $4/Ci; in large amounts, it costs less. You can estimate that tritium is worth roughly about $1/Ci, or $10,000/g. Some incidents have released as much as $100,000 worth of tritium. Aside from the cost, which we normally consider in emergency planning, the concern with radiation and the environment is what makes any work with tritium senstive beyond its value or beyond the real risk.

Of first importance in emergency planning is an alert signal so that you and others will know that something has happened. Next, you need instructions on where to go, what the control measures are, and whom you should notify. It's one thing to know it yourself, but it's equally important to inform your neighbors or anyone else who might be affected.

In your planning, you should arrange to notify your neighbors and the Health Physics Group and, of course, higher authority. As much as possible, emergency planning should be included as part of the Standard Operating Procedure.

Conclusion - Roy Reider, Safety Director

There has been enough interest in obtaining the information presented in this seminar to warrant the effort necessary to transcribe the tapes. Each speaker has edited his presentation and added figures where appropriate. Some interest has been expressed in future conferences dealing in detail with accident and near-incident experiences.

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