

ATOMIC ENERGY
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L'ÉNERGIE ATOMIQUE
DU CANADA LIMITÉE

**TRITIUM IMMOBILIZATION AND PACKAGING
USING METAL HYDRIDES**

**Immobilisation et emballage du tritium
au moyen d'hydrures de métal**

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Résumé

Le tritium provenant des réacteurs CANDU à eau lourde devra être emballé et stocké de façon sûre. Il sera récupéré sous la forme élémentaire T_2 . Les tritiures de métal sont des composants efficaces pour immobiliser le tritium comme solide non réactif stable et ils peuvent en contenir beaucoup. La technologie nécessaire pour préparer les hydrures des métaux appropriés, comme le titane et le zirconium, a été développée et les propriétés des matériaux préparés ont été évaluées. La conception des emballages devant contenir les tritiures de métal, lors du transport et durant le stockage à long terme, est terminée et les premiers essais ont commencé.

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ABSTRACT

Tritium recovered from CANDU heavy water reactors will have to be packaged and stored in a safe manner. Tritium will be recovered in the elemental form, T_2 . Metal tritides are effective compounds in which to immobilize the tritium as a stable non-reactive solid with a high tritium capacity. The technology necessary to prepare hydrides of suitable metals, such as titanium and zirconium, have been developed and the properties of the prepared materials evaluated. Conceptual designs of packages for containing metal tritides suitable for transportation and long-term storage have been made and initial testing started.

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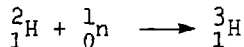
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Tritium Immobilization and Packaging Using Metal Hydrides

1. INTRODUCTION

Tritium is a radioactive isotope of hydrogen. It emits a low energy β -particle on decay to helium-3 with a half-life of 12.3 years. Tritium is present in the environment from three sources: natural tritium produced from cosmic radiation interacting with molecules in the upper atmosphere, testing of thermonuclear weapons and emissions from thermal reactors. There are various estimates(1,2,3) of the relative amounts of tritium from these sources. The approximate values are 7 kg of tritium from natural sources, 120 kg from weapons testing and 0.7 kg from thermal nuclear reactors.

In nuclear reactors employing heavy water as the coolant and/or the moderator, such as CANDU reactors, tritium is produced primarily by neutron capture by the deuterium atoms of the heavy water.



The tritium production rate in CANDU reactors is typically $2.4 \text{ kCi} \cdot (\text{MW(e)} \cdot \text{a})^{-1}$, ($89 \text{ TBq} \cdot (\text{MW(e)} \cdot \text{a})^{-1}$)(4). The exact value varies with reactor design. With an installed capacity of 5565 MW(e) in Canada the tritium production rate is approximately 13.4 MCi/a (0.5 EBq) or 1.4 kg/a . The total amount of tritium presently contained in Canadian reactors (both research and power reactors) is 6.5 kg . The tritium is contained within the reactor containment system and the emission of tritium from CANDU stations is maintained at less than 1% of the permissible limits(5). However in station operation, leakage of heavy water from various mechanical components occurs causing a tritium hazard for the reactor operating personnel. Because the tritium concentration in the water increases with time during reactor operation so does the hazard associated with heavy water leaks. To minimize this hazard steps are being taken to recover the tritium from the reactor's heavy water systems and to maintain a low tritium concentration.

A tritium recovery system to recover tritium from water of the Pickering Generating Station has been committed(6). This plant will recover $\sim 8 \text{ MCi/a}$ (0.3 EBq) at equilibrium conditions. The process to be used will be electrolysis of the water to give tritiated deuterium gas followed by cryogenic distillation to separate deuterium and tritium. The deuterium will be recombined with oxygen to return detritiated heavy water to the reactor. The tritium from the plant will be in the molecular form of hydrogen consisting of a mixture of

deuterium and tritium isotopes or perhaps as virtually pure T_2 . The product tritium from the tritium recovery system will be stored. The length of storage will depend on the development of a commercial market for it.

The objective of this program is to select, develop and demonstrate a method of packaging separated tritium in a form suitable for storage or disposal.

2. METHOD OF TRITIUM STORAGE

There are a number of potential methods for storing tritium. Tritium exists mainly in two molecular forms, water and hydrogen. Since tritium in the water form is several orders of magnitude more toxic than tritium as hydrogen(7) long-term storage in the water form is avoided unless the tritium concentrations are very low.

Tritium in the form of water can be stored in tanks but this is not suitable for long-term storage of concentrated tritium because of the potential for leaks. The tritiated water can be immobilized by incorporation into cements as a chemically bound hydrate and the cement packaged in a form suitable for storage or disposal(3,8,9). This is an effective, inexpensive method that is presently in use in various countries, particularly for fairly large volumes of low concentration tritiated water. The disadvantages of cements are the tritium can be leached out through chemical exchange with water(10) and can be driven off by heating to high temperatures above $500^\circ C$ (4). Impregnation of the cement with an organic polymer will reduce the leach rate of tritium by water(11), however, it is still sufficiently fast that the cement must be kept dry to retain its tritium.

Zeolites, such as molecular sieves and adsorbents, such as alumina and silica gel, also strongly hold water so are potential methods for immobilizing tritiated water. In these, like the cements, the tritium can be leached out by water, and heating them above $200-300^\circ C$ drives off the adsorbed water.

In contrast to chemically bound water as hydrates, hydrogen (or tritium) atoms bound to carbon in organic polymers are not subject to rapid exchange with water so the leach rate of tritium contained in these molecules is low(3). These compounds offer a medium that could be used to immobilize and store tritium. The drawback is that several processing steps are required in their formation, which in the case of concentrated tritium involves the handling of hazardous tritium-containing compounds in a complex processing scheme. At high temperatures, $>300^\circ C$, these polymers melt and decompose and eventually burn, releasing the tritium.

Tritium in the form of hydrogen, which would be the product from a tritium recovery plant, can be stored either directly as a gas in a container or immobilized as a metal tritide then stored in a container. Gas storage has the advantage that it is a simple well-established technology and the tritium is easily recovered. It has the disadvantage of a greater potential for leakage than for a solidified form such as a metal hydride.

Many metals react with hydrogen to form solid hydrides but only those of the transition metals have the required properties for the tritium storage application. The properties of these metal hydrides that make them suitable are very low dissociation pressures at normal temperatures, high capacity for tritium, ease of preparation and stability in air and water at storage temperatures. The hydrides of zirconium, titanium, hafnium and yttrium(3) as well as erbium have been suggested as useful for tritium storage. The dissociation pressures of titanium and zirconium hydrides are less than 10^{-20} atmospheres* at 25°C and less than 10^{-2} atmospheres at 500°C , but are completely dissociated by 1000°C , whereas the hydrides of yttrium and erbium are extremely stable with dissociation pressures of less than 10^{-3} atmospheres even at 1000°C . Titanium and zirconium hydrides would be suitable for recoverable storage whereas the hydrides of erbium and yttrium would be more suited for non-recovery of the tritium.

For all practical purposes there is no tritium partial pressure above these compounds at temperatures expected for storage, $<100^{\circ}\text{C}$. This means rupture of the storage container will not release tritium. Another advantage of metal tritides is their large capacity for hydrogen (or tritium). Density of hydrogen in some metal hydrides is similar to or greater than that of liquid hydrogen(12).

Tritium stored as metal tritides can be recovered as gas by heating to a sufficiently high temperature ($\sim 600^{\circ}\text{C}$ for Ti or Zr) and collecting the gas. This is not as convenient as simply opening a valve on a gas cylinder, but it does provide a simple method for separating the decay product ^3He from the tritium if that is desired. The ^3He can be pumped off the metal tritide at low temperature prior to heating to recover the tritium.

In comparison with gas storage, the use of metal tritides is considered to have some disadvantages. These are that a processing step is required between the recovery plant and the storage container to form the tritide, the tritium is not as easily recovered, and the metals are expensive. For this application, these considerations represent only minor disadvantages. The information in this report shows the preparation is a simple single step reaction. While the tritide

* 1 atmosphere, standard = 101.325 kPa

must be heated to recover the tritium, by having the tritium chemically bound to the metal it is much easier to separate the helium-3 from the tritium than with gas storage. Because of the high capacity of metal tritides and the small mass of tritium recovered the quantity of metal required is so small the cost is not significant. For example to hold an entire year's production from a plant extracting 800 g of T₂ (≈8 MCi ≈0.3 EBq) would require approximately 8 kg of TiT₂, which would cost less than \$500 for the titanium at current laboratory prices.

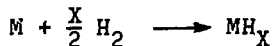
The hydrides of titanium, zirconium, yttrium and erbium have been studied as candidates for storing the tritium from a CANDU tritium recovery plant.

3. IMMOBILIZATION OF TRITIUM AS METAL TRITIDE

The objective of this work was to develop the necessary technology to prepare and package metal tritides and to evaluate their properties. Since the chemical properties of tritium (T₂) are virtually identical to those of hydrogen (H₂) and deuterium (D₂) and facilities for working with high specific activity tritium were not available, the initial phase of the work has been done with hydrogen and deuterium.

3.1 Preparation of Metal Hydrides

Metal hydrides are formed by the direct combination of the metal and hydrogen.



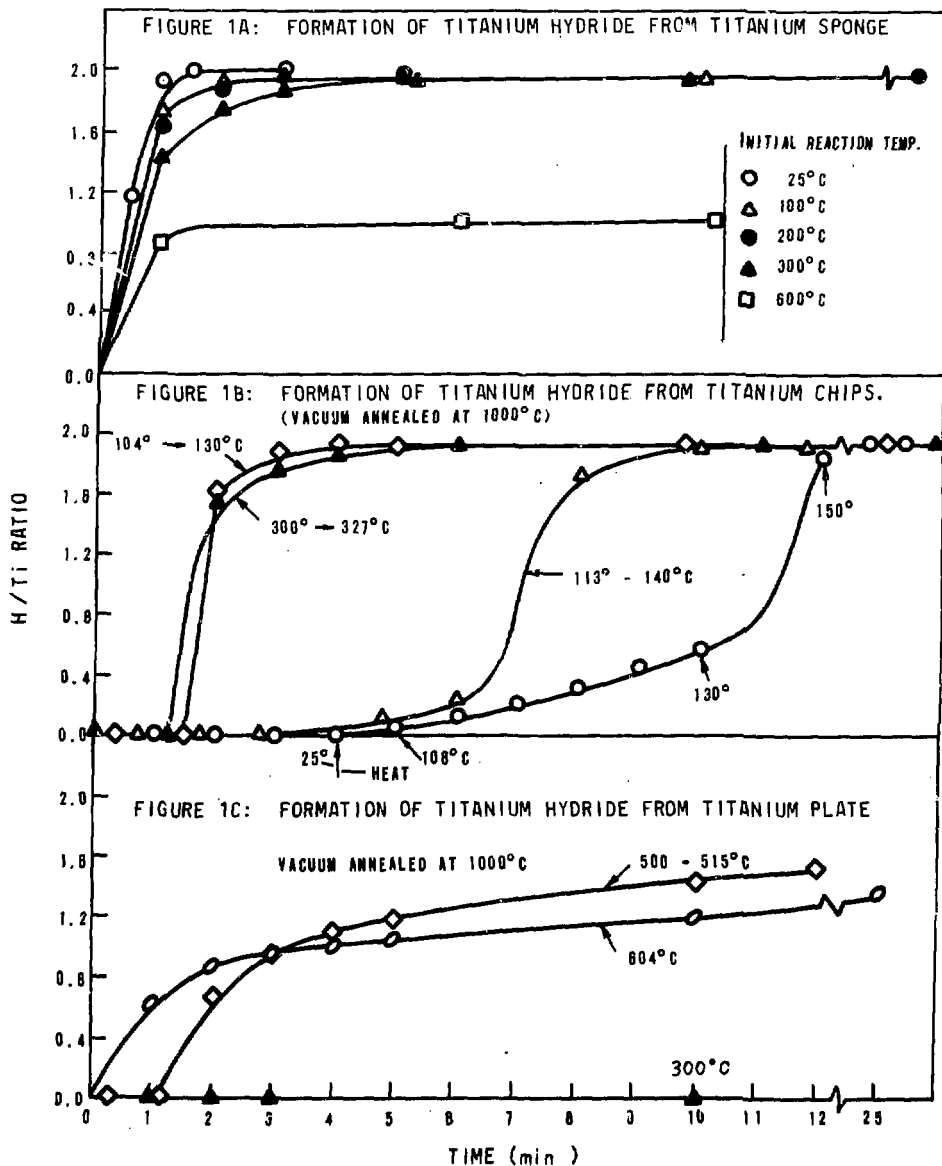
where M represents the metal and X the appropriate stoichiometry for the reaction of that metal.

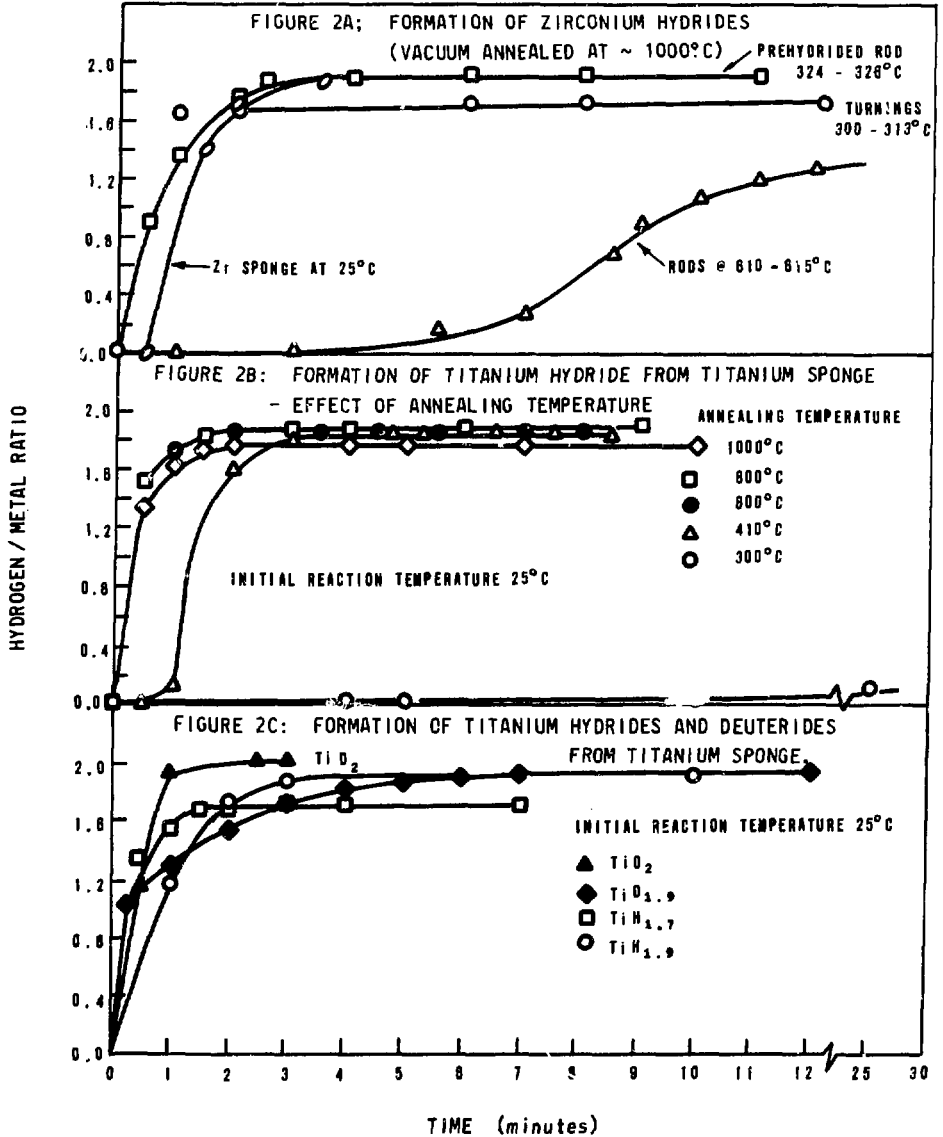
For this reaction to proceed quickly the metal surface must be clean and the hydrogen must be pure. The work has been done with titanium and zirconium in the form of metal sponge, turnings and rod or plate. The metal surface is cleaned by heating in vacuum (vacuum annealing) for a period of time to dissolve the surface oxide, then cooled to the desired reaction temperature. The hydrogen is purified by either passing through a hot palladium-silver alloy membrane or absorption/desorption on a uranium metal bed. This work was carried out on a conventional vacuum apparatus with facilities to measure the pressure temperature and volume of the hydrogen and the pressure and temperature of the reaction vessel. The reaction vessel in most cases was a quartz glass tube.

The rate of formation of titanium hydride from titanium sponge, titanium chips and titanium blocks is illustrated in Figure 1. For each of the three metal forms the metal was purified by vacuum annealing at 1000°C for a period of one-half to two hours, and the hydrogen was purified by passing through a hot palladium-silver membrane. Figure 1A shows the effect of reaction temperature on formation of titanium hydride from titanium sponge. The reaction rate is very fast with the reaction complete in less than 5 minutes. The reaction temperatures quoted are initial temperatures when the hydrogen was first introduced to the metal containing vessel. Because the reaction is exothermic, the temperature rises with time. The H/Ti ratio for the run at 600°C was limited to 0.9 because the dissociation pressure at this temperature equaled the pressure of hydrogen in the system. The reaction rate is slower with Ti metal in the form of chips or plate in comparison with sponge. At an initial temperature of 25°C the titanium chips did not react. On heating, the reaction started at ~100°C and proceeded quickly after 130°C. Similarly, a reaction with an initial temperature of 100°C showed a five-minute induction period before the reaction started. This sample was dehydrided by heating and pumping off the hydrogen, then rehydrided. The reaction rate on this second hydriding at 100°C was much faster. The faster reaction rate on the second hydriding is believed due to increased surface area in the metal sample as a result of the hydriding-dehydriding procedure. For a fast reaction rate with titanium chips or turnings an initial reaction temperature of ~300°C is required. As shown in Figure 1C, reaction temperatures of 500-600°C are required with titanium plate or bars.

The conclusion from these experiments is that the rate of the hydride formation reaction is dependent on the surface area available for the reaction, as might be expected. The sponge samples with a much higher surface area reacted more quickly and at a lower temperature than samples of titanium with lower surface areas such as turnings or bars. Because of the bulk form of the turnings and bars, diffusion of the hydrogen into the metal is believed to be the rate controlling step(13) in the reaction and hence higher temperatures are required. The distance that the hydrogen diffuses into a metal cylinder depends inversely on the square of the distance (14) and hence a more complete penetration of hydrogen at a particular time and temperature, i.e., a faster hydriding rate, is noted for sponge compared with that for the bulk samples.

Similar results were observed with zirconium samples and are shown in Figure 2A. The zirconium sponge samples reacted quickly at room temperature whereas turnings required a temperature of 300°C and rods were slow even at ~600°C. Again pre-hydriding a rod greatly increased its subsequent reaction rate.





The choice of 1000°C for the vacuum annealing temperature in the previous experiments was arbitrary. A series of runs with titanium sponge was carried out to determine the minimum temperature at which the metal could be annealed and still have a rapid hydriding rate. These results are shown in Figure 2B. In each case the initial reaction temperature was 25°C. These data show the minimum temperature for annealing titanium sponge is about 400°C. If the annealing is done at 300°C the subsequent reaction at room temperature is very slow. Annealing temperatures above 400°C have little effect on the rate of hydriding.

Figure 2C shows that within the experimental scatter from run to run there is no difference in the rate of formation of the hydride or deuteride from titanium sponge. From this observation it is expected the reaction with tritium will be similar.

The major difference in the chemistry with tritium compared to hydrogen or deuterium is expected to be the effect of helium-3 from tritium decay. This helium is known to inhibit the hydriding reaction(15). To simulate this effect, samples of titanium sponge were hydrided with hydrogen containing helium using the standard hydriding procedure. The results are shown in Figure 3A. The amount of hydrogen used in these tests was sufficient to yield a titanium-hydrogen ratio of between 0.9 and 1.0 at complete reaction. In the presence of helium there is an initial rapid absorption of hydrogen followed by a very much slower rate of reaction. With as little as 0.5% He only half of the hydrogen is reacted quickly, the remainder reacts at a very slow rate. This effect is thought to be due to a blanketing effect of the metal surface by the helium, preventing access to the surface by the hydrogen(15). To reduce this blanketing effect the procedure and apparatus was modified to allow circulation of the gas over the metal surface. This greatly improved the rate of reaction as shown in Figure 3B where complete reaction of the hydrogen in a mixture containing initially 6% He occurred in about 10 minutes. Without circulation the reaction would have required several hours. The rate with circulation is still slower than with pure hydrogen but is fast enough to be acceptable for the tritium application. An alternative explanation for the observed helium effect has been suggested by Cox(16) who speculates the inhibition is due to small quantities of oxygen contained in the helium poisoning the metal surface. Since the oxygen content of the helium used in these experiments was not measured this possibility cannot be assessed from our data.

The inhibition of the hydriding reaction can be overcome either by purification of the tritium immediately prior to reaction or providing for circulation of tritium-helium mixture

FIGURE 3A: EFFECT OF HELIUM ON FORMATION OF TITANIUM HYDRIDE FROM TITANIUM SPONGE

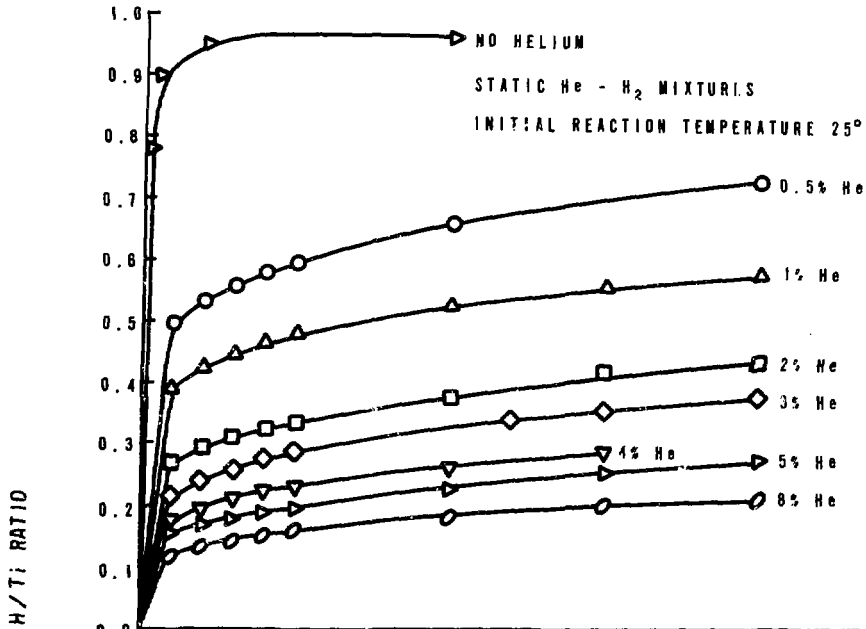
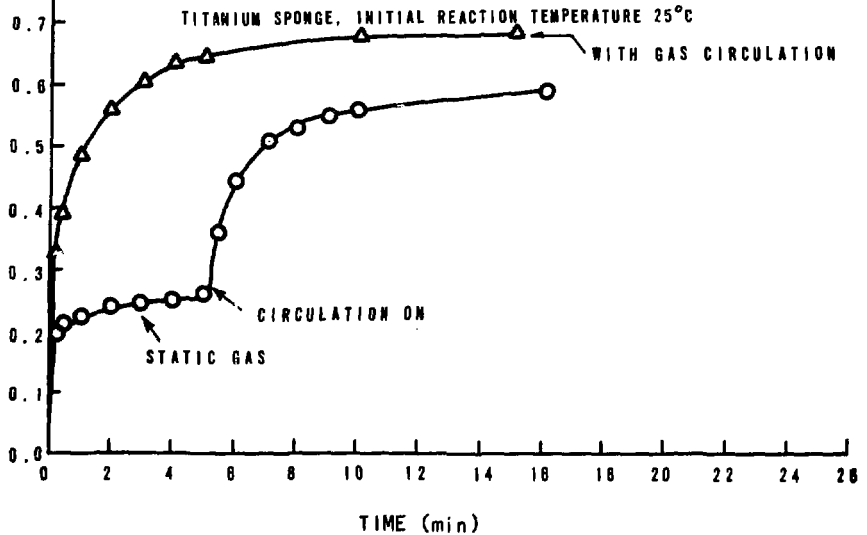


FIGURE 3B: EFFECT OF RECIRCULATION OF GAS



over the metal. The effectiveness of circulation is shown in Figure 3B. Another concern of the helium effect arises if it is required to immobilize additional tritium on a metal bed that already contains partially decayed tritium. To simulate this effect a sample of titanium sponge was activated and partially hydrided, then the container backfilled with helium. After several weeks the helium was pumped off and replaced with hydrogen which reacted quickly. This was repeated several times over a period of months. In each case once the helium was removed the subsequent reaction with hydrogen occurred readily.

3.2 Properties of Metal Hydrides

The property of the hydrides of most interest for tritium storage is their stability at expected storage conditions. The tritium-containing compound should be stable thermally, unreactive in air and resistant to leaching of the tritium by water.

3.2.1 Reactivity in Air

The reactivity of these hydrides in air at elevated temperatures has been investigated in a qualitative way by heating them on a hot stainless steel plate in air and observing when they start to spark and/or burn. Samples of titanium and zirconium metal and their hydrides were observed using various forms such as rods, powders, turnings and sponge. Samples were heated from room temperature to $\sim 800^{\circ}\text{C}$. Temperatures were measured by means of a thermocouple located directly below the sample in the steel plate, and also by an optical pyrometer. The sample sizes were in the range of 2-4 grams. Reproductions of the photographs taken during heating of these samples are shown in Figures 4, 5 and 6*. In Figure 4, sponge samples of Zr metal, ZrH_1 , Ti metal and TiH_1 are shown being heated from 25°C to 810°C over a period of 220 minutes. The color of these samples gradually changed during heating as they slowly oxidized, but at no time up to 800°C was there any evidence of a vigorous reaction. Figure 5 shows samples of ZrH_2 as a very fine powder, $\text{ZrH}_{1.9}$ and $\text{ZrH}_{1.0}$ as large sponge lumps, and $\text{ZrH}_{0.8}$ as rod, heated from room temperature to 620°C in 48 minutes. The fine powder of ZrH_2 started to spark at 470°C and was completely oxidized at 620°C . The large sponge samples of $\text{ZrH}_{1.0}$ and $\text{ZrH}_{1.9}$ showed a gradual color change and some evidence of slow oxidation at 620°C . The only visible change in the samples of $\text{ZrH}_{0.8}$ rod was a slight color change. With the stainless steel block at approximately 620°C , the previous four samples were removed and replaced with samples of $\text{TiH}_{1.9}$ turnings, $\text{TiH}_{1.0}$ small sponge, $\text{TiH}_{1.0}$ large sponge and $\text{TiH}_{1.0}$

*Not all copies of the report will be supplied with colour photographs.

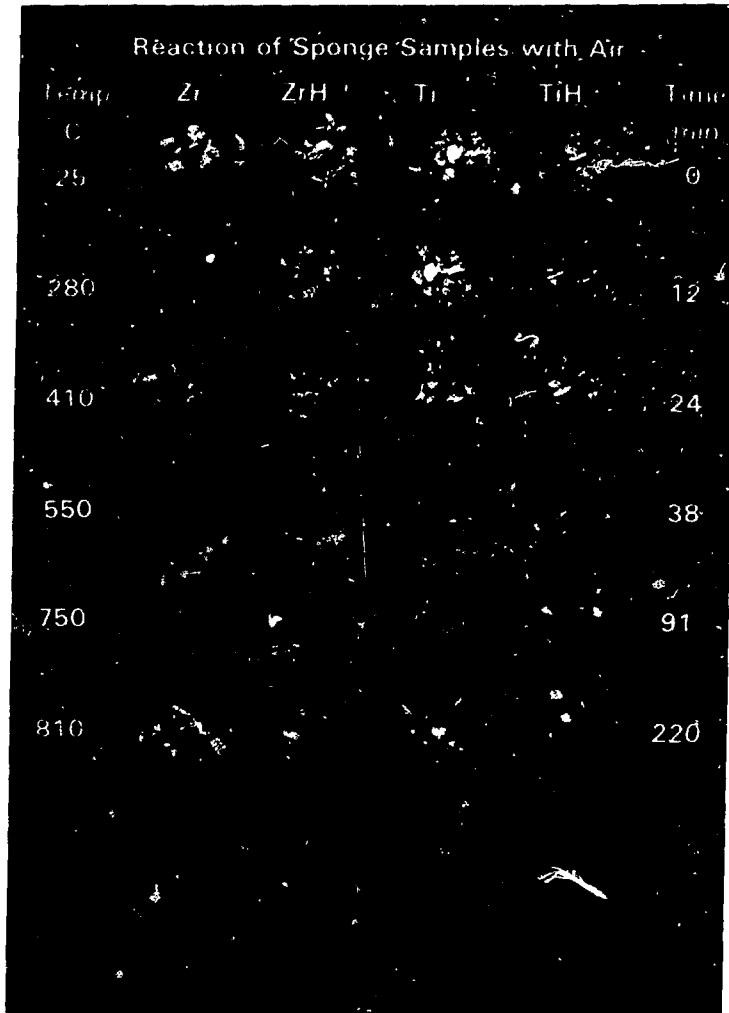


FIGURE 4

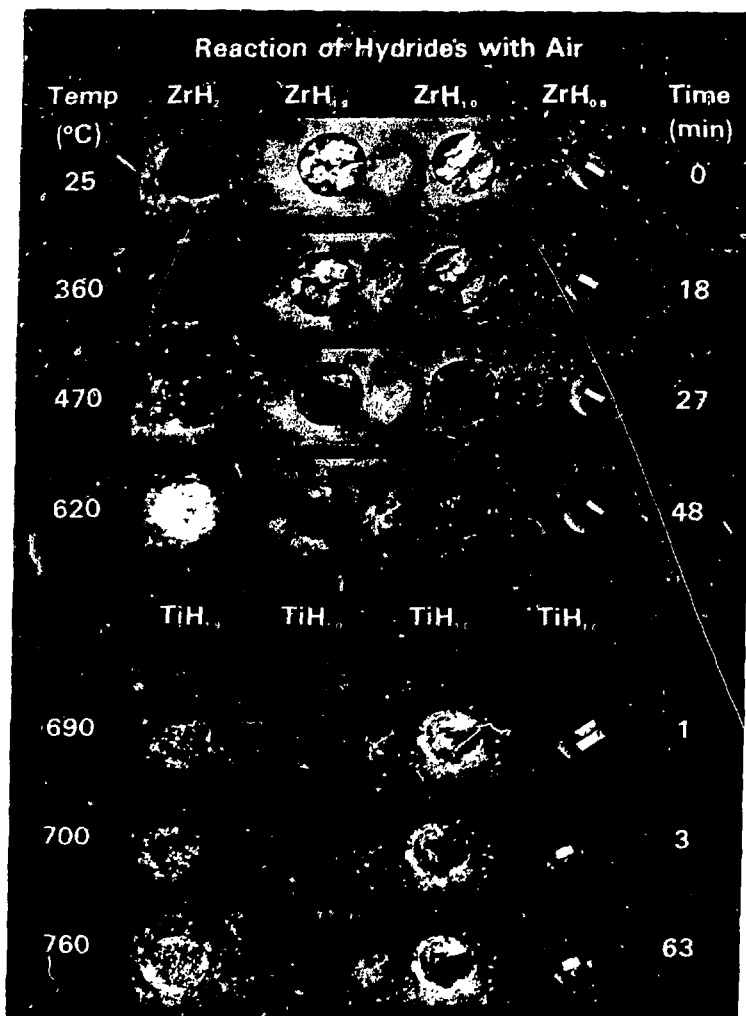


FIGURE 5

plate. None of the samples sparked or burned. During one hour and a temperature rise to 760°C, the turnings slowly oxidized, the sponge samples and the plate gradually darkened in color indicating a gradual oxidation. It is assumed the hydride was dissociated and the hydrogen lost at these temperatures but there was no evidence that the hydrogen burned to form water.

In the tests shown in Figure 6, samples of hydride made from titanium and zirconium sponge were placed on the hot plate at various temperatures from 290°C to 820°C and observed. There was no observable reaction at 290°C and 450°C for any of the samples, at 560°C the smaller particles of $ZrH_{0.8}$ sponge glowed red after one minute, but the whole sample did not ignite. After 4 minutes the glowing particles went out leaving white oxide particles with the bulk of the sample still black. The titanium hydride and deuteride samples did not glow at these temperatures but showed the usual slow oxidation. Similar results for the titanium hydride and deuteride samples were observed at 750°C (with the stainless steel plate glowing red) but the whole sample of the $ZrH_{0.8}$ small sponge glowed red and burned. The largest size lumps of $ZrH_{1.1}$ sponge did not glow red or burn but remained black and only slowly changed in color. Similar observations were found at 810-820°C. Large and small size sponge samples of $TiD_{0.9}$ and $TiH_{0.9}$ when placed on the hot plate at 820°C did not burn but gradually changed to a bluish hue which is the color of the oxide, Ti_2O_3 .

The conclusions from these qualitative observations is the hydrides of titanium and zirconium prepared from metal, sponge and turnings are not very reactive in air at normal temperatures. The reactivity increases with the degree of subdivision of the compound. However the lowest temperature at which a fine powder of ZrH_2 would burn was ~450°C. Samples of zirconium hydride prepared from sponge material did not react until dropped on a hot surface of 560°C. Over the whole range of temperatures up to 820°C samples of titanium hydride did not burn. From this observation it is concluded titanium hydride samples are less reactive than zirconium hydride. Surface areas for these samples have not been measured, however, when this is done an attempt will be made to correlate the reactivity in air at various temperatures with the surface area of the particles. It is evident from these results that the particle sizes will have to be very much smaller than those used here in order to produce a material that will be pyrophoric in air at room temperature.

3.2.2 Tritium Leach Tests on Titanium and Zirconium Deuterides

Samples of titanium and zirconium deuterides were prepared containing trace quantities of tritium. The deuterium-metal ratio ranged from 0.5 to 2.0 with sample sizes from 1 to 7 g. The deuterides were prepared from deuterium gas containing

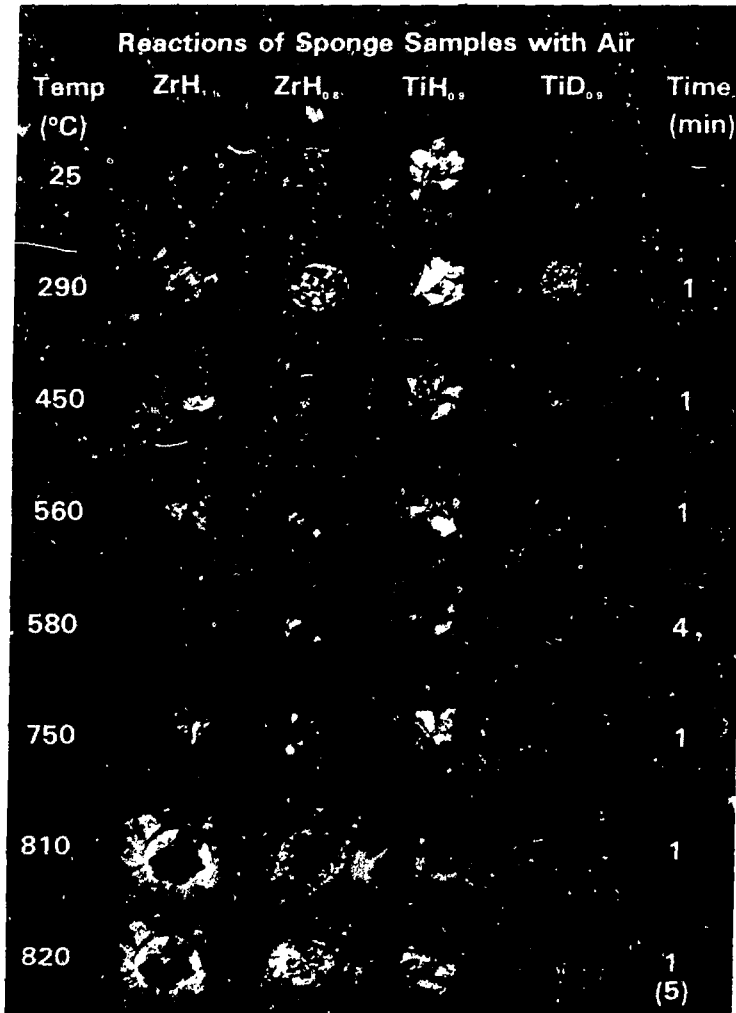


FIGURE 6

approximately 6 mCi/L (0.2 GBq/L) of tritium obtained from the electrolysis of tritiated heavy water. The zirconium and titanium deuterides prepared from both sponge and rod were used in static and dynamic leach tests. In the leach test procedure, the IAEA standard leach test method(17) was followed as closely as possible. The leachants were deionized water, solutions of CaCl_2 , NaCl , and NaHCO_3 (30 meq/L) and a salt brine. The samples were contained in small stainless steel mesh baskets suspended in 50 mL of leachant. For dynamic tests the bottles containing the samples were placed on a tray on an orbital shaker. The leachant was changed daily during the first week and weekly for the following eight weeks followed by monthly changes. Tritium release into the leachant was measured by liquid scintillation counting. The results were expressed as the incremental leaching rate R_n as defined by,

$$R_n = \frac{a_n/A_0}{(F/V)t_n}$$

where a_n = radioactivity leached during the leachant renewal period, n ,
 A_0 = radioactivity initially present in the specimen,
 F = exposed surface area of the specimen (cm^2),
 V = volume of specimen (cm^3),
 t_n = time (days) of leachant renewal period.

The surface area of the sponge samples was estimated from the geometrical shape of the sample, ignoring the porosity of the sponge material. This had the effect of producing a leaching rate higher than would be the case if the actual surface area were used. Even with this assumption the observed leach rates of tritium from these deuterides were very low. For static tests in deionized water the incremental leaching rates were in the range of 10^{-8} - 10^{-9} cm/day. In the dynamic tests the specimens of titanium and zirconium deuteride have higher leaching rates than the same specimens under static conditions but the rates are still in the 10^{-7} - 10^{-8} cm/day range. When actual surface areas are determined and applied these rates will decrease. There has been a slight decline in the leaching rates over the ten months of the test but they are still within the ranges mentioned above. In this time period the cumulative fractional releases range from 10^{-3} to 10^{-6} .

Leaching rates for deuterides prepared from titanium and zirconium rod specimens were found to be in the 10^{-9} - 10^{-10} cm/day range for deuterium-metal ratios of 1.0 and 2.0.

The leaching behaviour of $\text{TiD}_{1.5}$ and $\text{ZrD}_{1.5}$ sponge specimens was also studied in CaCl_2 , NaCl , and NaHCO_3 solutions

(30 meq/L) and in a salt brine solution. In all the above solutions the leach rates were in the range 10^{-8} - 10^{-9} cm/day.

3.3 Yttrium and Erbium Hydrides

A brief examination was made of the hydrides of yttrium and erbium as potential candidates for the non-recoverable storage of tritium. The dissociation pressures of these hydrides is very low even at elevated temperatures, and while recovery would be possible it would not be convenient.

It was found that samples of yttrium and erbium hydrides could be prepared using the same techniques described in the previous sections for titanium and zirconium. The rates of reaction were somewhat slower than for Ti and Zr at room temperatures but at temperatures of 170°C YH_3 was formed in 35 minutes. Somewhat higher temperatures were required for erbium.

There was no vigorous reaction on heating these hydrides in air from 25° to 750°C using the technique described in section 3.2. Slow oxidation was observed. However when samples of $\text{YH}_{2.8}$ and $\text{ErH}_{2.7}$ were placed on the hot stainless steel block at 600°C they immediately ignited.

Leach tests have not been done.

4. ANALYSIS OF TRITIUM

The tritium from the tritium recovery plant will not be 100% T, there will be some D and perhaps some H. In order to know how much tritium is placed in each package it is necessary to know the number of moles of gas put into the hydride, plus the composition of that gas. The number of moles of gas is readily measured through standard pressure, volume and temperature measurements. For the composition measurement, there are a number of choices available: mass spectrometry, gas chromatography, ionization chambers and calorimetry. Of these, gas chromatography appears to be the simplest and most direct method for this analysis. It is not extremely sensitive for hydrogens but it is expected the range of compositions of tritium in deuterium from the recovery plant will be in the range of 1-99%, which will not be a problem for the technique.

The literature reports the separation of hydrogen isotopes by gas chromatography(18). Preliminary work was undertaken using H_2 , HD, D_2 to develop expertise and experience with the method. Good separation between these three hydrogens has been observed using a 3 m column packed with ferric hydroxide treated alumina, operated at 77 K. The separation achieved is shown in Figure 7. On the basis of this separation compared

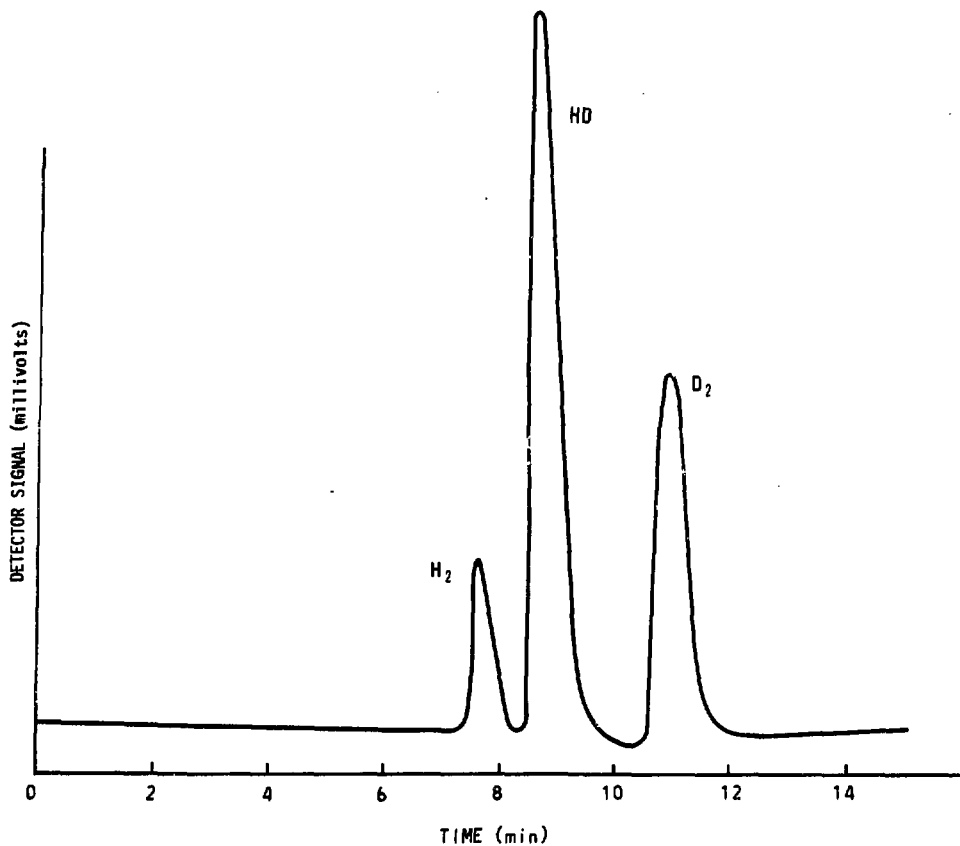


FIGURE 7: SEPARATION OF H₂, HD, D₂ ON AN ALUMINA COLUMN COATED WITH FERRIC HYDROXIDE (80 - 100 MESH, 3 m x 1 mm); 77 K; HELIUM FLOW, 98 mL/min

to those in the literature where the tritium containing hydrogens were also separated, this method appears to be feasible for the necessary composition measurements.

Some work on the other methods of analysis is also planned to provide alternative or back-up techniques.

5. TRITIDE PACKAGE

Even though tritium immobilized as a titanium or zirconium tritide is very stable, and inert, and the β -particles from the tritium are of such a short range that the tritide could be safely handled without further protection, it is certain that packaging of the tritide will be required for permission to transport or store the material. This package must be designed to withstand the postulated accidents that may occur in transportation of radioactive materials and must meet the requirements of licencing authorities.

A conceptual design of a primary container and a shipping package has been made to contain 0.5 MCi (19 PBq) of tritium as the metal tritide(19). The schematic diagrams of the primary vessel and the transportation package are shown in Figures 8A and 8B, respectively.

The primary vessel is a 4.5 L stainless steel container designed to contain the helium generated by the complete decay of the tritium. Austentic stainless steel has been shown to be a suitable material for the long-term storage of tritium at ambient temperature(20). This vessel has been designed to double as the reactor vessel in which the metal tritide is prepared. For this reason it is equipped with two valves to permit circulation of the gas during the hydriding procedure. The inlet and exit of the vessel are protected by sintered steel filters to prevent loss of any metal tritide particles that may be present. Tests with a prototype vessel of this design have shown the hydriding reaction can conveniently be carried out in this vessel.

For transportation it is expected the primary container will be placed in a second stainless steel container with a flange closure and this two-container assembly placed in a suitable sized drum packed with insulation to provide protection from heat in the event of a fire.

FIGURE 8A: CONCEPTUAL DESIGN FOR METAL TRITIDE CONTAINER

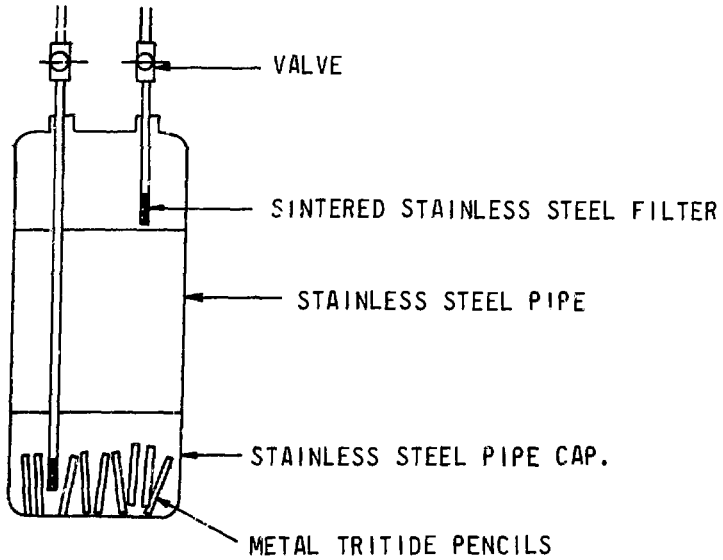
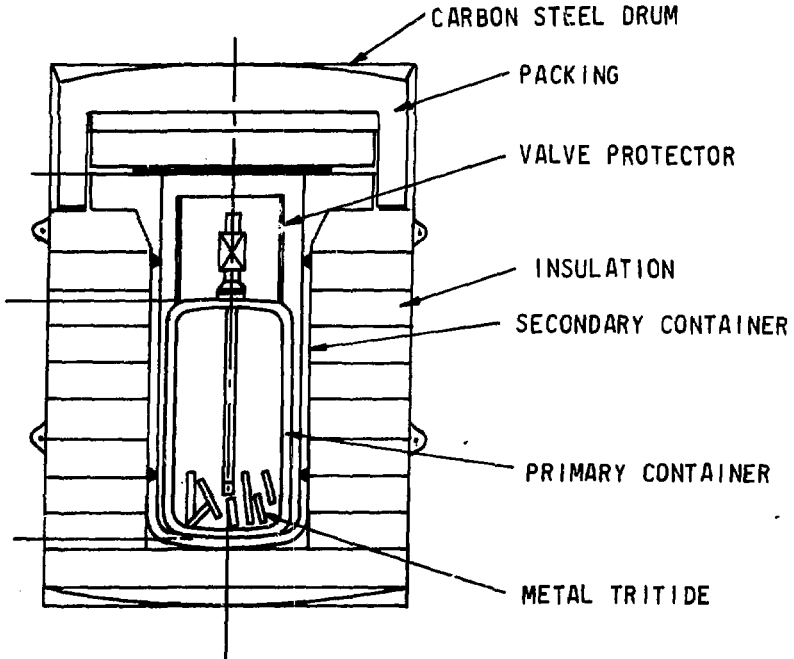


FIGURE 8B: CONCEPTUAL DESIGN OF A TRITIDE TRANSPORTATION PACKAGE



6. FUTURE PLANS

The work covered here, with the exception of the leach tests, has been done with non-radioactive hydrogen and deuterium. To do the same work with high specific activity tritium as T_2 special facilities for handling radioactive materials are required. A laboratory is being modified and equipped to allow work similar to what has already been done to be done with tritium. The hydriding apparatus has been designed for operation in a glove box, with components suitable for tritium service.

The future work will include the modification of procedures developed for the inactive work, evaluation of equipment for tritium service, preparation of high specific activity metal tritides, evaluation of their properties and development of analytical techniques.

7. SUMMARY

It has been shown the hydrides of titanium and zirconium are suitable for the storage of tritium. These hydrides are easily prepared by a simple reaction. If the sponge forms of the metals are used the reaction occurs quickly at room temperature. The metals must be preconditioned by heating in vacuum (0.1 Pa) at 500°C for about one-half hour and the hydrogen must be free of impurities to ensure rapid reaction. Similar reactions were observed using erbium and yttrium sponge. Studies on the reactivity of these hydrides in air and water showed they do not react or decompose at normal temperatures so that tritium will not be released during storage.

A primary tritium storage vessel has been designed and fabricated. It was demonstrated to be practical to use this vessel both as a reactor for the immobilization process and the primary storage vessel. This eliminates the necessity of handling of the tritide outside a container.

Based on the technique developed with hydrogen and deuterium, facilities and equipment have been designed to handle high specific activity tritium and to prepare metal tritides in packages safe for storage or disposal.

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