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**Tritium Interactions of Potential
Importance to Fusion Reactor Systems -
Technology Requirements**

William R. Wilkes

January 15, 1976

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Research and Development Report

MOUND LABORATORY

Miamisburg, Ohio

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Abstract

The tritium technology requirements created by the controlled thermonuclear research program to develop a demonstration fusion power reactor by the year 2000 are reviewed. It is found that the majority of the technological advances which are needed to ensure adequate tritium containment in a tritium breeding power reactor need to be demonstrated on a pilot scale by approximately 1983, so that they may be incorporated into EPR-II, the second of two planned experimental power reactors. The most important advances include development of containment materials with permeabilities to tritium well below measured values for stainless steel; large scale, low inventory deuterium-tritium separation systems; and improved monitoring and assay systems. There are less critical requirements for information about the effects of tritium and helium on the mechanical properties of materials, the effects of tritium on biological systems, and data on physical and chemical properties of tritium. Substantial progress needs to be made on these problems early enough to permit possible solutions to be tested on EPR-I. In addition, major improvements in tritium handling equipment are required for EPR-I. Those technological problems for which solutions have not yet been demonstrated by EPR-II must be solved by 1989 if they are to be assured successful application in the demonstration reactor.

Introduction

The goal of extracting usable power from the energy released by a controlled thermonuclear reaction has been actively pursued for many years. Until recently, the major effort of this pursuit was concentrated on plasma physics research in order to understand the conditions which are necessary to initiate and sustain a fusion reaction. Within the past few years, it has become evident that the necessary physical conditions for fusion power may soon be realized, and as a result, there is now an increasing emphasis on the many engineering and materials problems which must be solved in order to be able to construct a practical fusion power reactor.

Some of these problems are related to the use of tritium in fusion reactor systems. For at least the first generation of fusion power reactors, the fuel will be a mixture of deuterium and tritium, as the temperature required for the D-T fusion reaction is lower than that for any other fuel and because the energy released in a single reaction is large compared to that for most other fuels.

The presence of tritium in a fusion reactor poses a number of unique and difficult problems, principally because, as an isotope of hydrogen, tritium readily diffuses through most materials at reactor temperatures, and because tritium is radioactive. Solutions to these problems are

not readily available, however, for many of the properties of tritium and of its interactions with other materials which are necessary to solve these problems are incompletely understood. As a result, considerable technology will need to be developed to provide means of containing and handling tritium under reactor conditions and to solve the many other problems created by the presence of tritium in a fusion reactor.

Some information on tritium and its interactions with materials does exist, as tritium has been used and studied extensively within the AEC (now ERDA) for many years in connection with a variety of AEC programs. Much of this work, however, has been described only in classified reports and is, therefore, inaccessible to the general scientific community. In the first phase of this project, the classified literature pertaining to tritium was surveyed by the author, and a critical review of this literature was performed.

There is also a substantial body of unclassified tritium research. This includes some work which was once considered sensitive or related to sensitive work but which has since been declassified or published. This work, in combination with that which is in the classified literature comprises the

available store of information on tritium and its interaction with matter.*

The second phase of this project, which is the subject of this report, consists of a comparison of the tritium technology required for fusion power with that information which is available, either in the open literature or which can be

obtained from classified sources. This was done in order to outline future tritium research which will be required to ensure successful operation of deuterium-tritium fueled fusion machines. A timetable for future tritium research has been developed which is based upon this comparison and upon current plans for fusion research.

CTR Timetable

The requirements for tritium-related research in the controlled thermonuclear program must be considered in relation to a timetable for major fusion program milestones. These milestones have been set by the Division of Controlled Thermonuclear Research of the U. S. Energy Research and Development Administration, and they are scheduled so as to achieve the operation of a commercial scale demonstration power plant during the period 1995-2000. There are four major milestones which involve increasingly larger and more complicated tritium processing systems.

The first is the Tokamak Fusion Test Reactor (TFTR),¹ for which the conceptual design has been completed. This machine, which will begin operating in 1980 will be the first in the world to use deuterium and tritium in a reactor-like plasma. Power will be produced in the plasma at a rate of several thermal megawatts, but there will be no power-recovery system or breeding system. The tritium fuel will be stored as a solid compound until it is needed, when a few milligrams of gas will be generated and injected into the plasma. After a test shot is performed, the unburned plasma

will be pumped out, absorbed on a second, more stable hydride-forming material, and eventually disposed of. The lifetime of the machine will be short, and the duty cycle will be low, so that current technology should largely be adequate to ensure the success of the experiment.

The second milestone is the Experimental Power Reactor (EPR-I).² This machine will produce a few tens of megawatts of electrical power, but will probably not breed tritium. If tritium breeding is attempted, it will be as a supplementary experiment, added on or inserted as a module after other aspects of EPR-I have been successfully tested. It is expected that large extrapolations of current technology will not be necessary to the success of EPR-I. This machine should begin operation in 1985.

Following EPR-I is EPR-II, which will be a breeder, although perhaps the breeding ratio will be less than unity, and which will produce approximately 100 MW of electrical power. It will probably be the first machine to generate electricity at rated power for extended periods of time. It will follow EPR-I by 4 or 5 yr and thus can take advantage of some more advanced technologies than can EPR-I.

The final milestone is a commercial scale reactor capable of producing 500 MW of electrical power in the year 2000. It will operate continuously,* with a design lifetime of 30 yr, and, of course, will be capable of breeding tritium faster than it is consumed. Major technological advances will be incorporated in this machine provided that they have first been successfully tested elsewhere (EPR-I or -II, for example).

All these machines are tokamaks, because at this time the tokamak is the most

*A thorough review of unclassified tritium-related research is beyond the scope of this project. Therefore, it has been necessary to assume in many cases that other investigators have surveyed those parts of the open literature relevant to their work. For example, no attempt was made to survey the open literature for information on the permeability of niobium to tritium. No such information was discovered in the survey of the classified literature, and discussions of the permeation problem in fusion systems by various authors indicated that knowledge of hydrogen (to say nothing of tritium) permeation through niobium is woefully incomplete. It was therefore concluded that there is a need for research on the permeation of tritium through niobium.

*The plasma will probably be pulsed, but electrical power delivery to the network will be continuous.

promising magnetic confinement fusion device available. Both θ -pinch and mirror machines are also being explored, and although there are no firm plans for specific machines of either type in the FTR-EPR-DPR sequence, they are available as backup systems. In the event either device should replace the tokamak as the primary fusion machine, the timetable will not be drastically altered, nor will the character of the tritium-related problems change much.

In addition to these milestone experiments, all of which involve D-T fuel, there are supporting fusion experiments which are planned to provide information about specific parts of the overall problem. For example, an intense neutron source (INS)³

and a fusion engineering research facility (FERF)⁴ are contemplated. The INS will provide ~ 14 -MeV neutrons ($10^{14}/\text{cm}^2 \cdot \text{sec}$) to test radiation damage effects in materials. The neutrons will be produced in a supersonic deuterium jet into which is injected a beam of tritium ions. The FERG is a D-T fueled, mirror reactor which will provide a source of 14-MeV neutrons at a flux of $\sim 10^{15}/\text{cm}^2 \cdot \text{sec}$, for more advanced engineering and materials tests than are possible in either the INS or EPR-I. The INS will operate around 1980-81, and the FERG is expected to operate in 1985. Other, less well-established experiments include a θ -pinch FERG and a second fusion test reactor, TFTR-II. The research plan for magnetically confined fusion is described in greater detail elsewhere.⁵

Subject Categories

At the outset of this study, several general categories of possible tritium interactions were outlined as an aid to the organization of the work involved. The definitions have shifted during the study with improved understanding of the problems involved, and there are many overlapping areas, so the categories are considered only as guidelines. They are in approximate order of importance:

1. Kinetics (diffusion, permeation through materials).
2. Mechanical properties (effects of hydriding, embrittlement, corrosion).
3. Breeding and recovery (removal of tritium from breeding material). This principally involves tritium as tritide; its solubility, vapor pressure over solutions, etc.
4. Interactions resulting from decay of tritium (generally involves creation of helium-3 and attendant phenomena).
5. Environmental, health, and safety considerations.
6. Techniques for handling and transporting tritium (including equipment, machinery, etc.).
7. Physical and chemical properties (vapor pressure, reaction rates, etc.).
8. Instrumentation, assay, and monitoring.

During the search and review of the classified literature on tritium it was discovered that the great majority of the research falls into one of the first three

categories. There are, in addition, a limited number of reports describing work on the remaining subjects. Finally, there are many reports on topics which have so little relevance to foreseeable fusion reactor problems that they are eliminated entirely.

Even in the work that fits one of the above categories, a large part of the information that has been reported is only marginally or indirectly relevant to fusion reactor requirements. There is, for example, a substantial body of information on tritium breeding in lithium-aluminum alloy, which is, of course, a possible breeding blanket material. None of the reports, however, discusses the behavior of the tritium-alloy system in the low tritium concentration ranges (e.g., <100 ppm) and high temperatures that will be required in a practical reactor situation.

There are numerous permeation data as well, but the temperature ranges covered are usually below those where information is required for fusion applications. In addition, the pressure ranges are generally above an atmosphere, whereas fusion applications may require knowledge of permeation processes at near vacuum (e.g., $<10^{-4}$ torr) conditions.

In summary, although there is a considerable amount of information available in classified sources on tritium and its interactions with materials, few relevant data exist on the properties which must be known to solve the problems most crucial to the success of fusion power. It is these problems which will be discussed in the remainder of this report.

In discussing the areas of research which are considered to be required, I examined two principal criteria. The first is an assessment of the information in terms of its proximity to the critical path for fusion power. As an example, information on the diffusion of tritium through lithium aluminate has a very low priority for any nonbreeding reactor, because it is not on the critical path, but a very

high priority for studies of reactors employing solid blankets. The second assessment is in terms of the time by which the information is required. Hence, refractory metal permeation data have a lower time priority, for example, than do stainless steel permeation data, because current technology does not yet allow construction of a refractory metal reactor.

Required Research On Tritium Interactions

The first category generally covers the permeation, diffusion, and solubility of tritium in solid structural materials. It also includes kinetic effects in breeding materials, which may be either liquid or solid.

Probably the single most crucial question relating to the use of tritium in a fusion reactor is that of containing the tritium within vessels whose walls are more or less permeable to tritium. Tritium leakage rates as low as 1-10 Ci/MW·yr of electrical power capacity may be required. At pressures above a few tens of millimeters of mercury, the permeation of tritium through the best available structural material, stainless steel, is well known; and at temperatures high enough to permit efficient power generation, the rate of permeation, when extrapolated to pressures that are typical of a reactor blanket, is excessive. Hence, if these extrapolations are valid, a major problem is the development of effective barriers to tritium permeation. There is evidence that the extrapolations are not valid, however, in which case accurate low pressure permeation data are still required. We shall discuss permeation barriers first.

KINETIC EFFECTS - BARRIERS TO PERMEATION

The first category of permeation barriers includes those which will be used where there is not a high neutron flux, for example the heat exchangers used to cool the deuterium jet in the INS or the heat exchangers in the power cycle of an EPR. These barriers will probably be the primary defense against tritium loss in such devices, for the tritium concentration inside the confined volume will be high, while outside it must be very low.

Primary candidates for these barrier materials include beryllium-copper, aluminum, composite or sandwich type materials,⁶ and metal oxides. Beryllium-copper and aluminum have permeabilities two or more orders of magnitude below that of stainless steel in the temperature range below about 500°C. For those applications

where these materials may be utilized, the advantages are great, but the experience of the fission reactor industry suggests that large-scale testing of these materials under reactor conditions is necessary to ensure against unexpected failures in actual use.

Composites permit low permeability materials such as aluminum or tungsten to be used in conjunction with corrosive media by means of a corrosion-resistant material such as stainless steel which is bonded to one or both sides of the barrier material. There is evidence that the permeability of such composites is actually even lower than would be predicted theoretically from the individual permeabilities. This is attributed to the formation of a very stable oxide film at the interfacial surfaces. The fabrication technology for these composites is still being developed, however, and extensive testing on at least a pilot scale is required to ensure their successful utilization.

Metal oxides (e.g. Al_2O_3) generally have extremely low permeabilities, but even a few widely spaced, minute cracks can destroy the effectiveness of such a barrier. Furthermore, the thermal expansion coefficients of metal oxides are generally quite different from those of metals, so that the bond between an oxide and a metal may fail with temperature cycling. Self healing barriers, if they can be developed, would be a major improvement. Other surface treatments need further investigation, as well. Electropolishing the surface⁷ and calorizing an aluminum layer onto the surface⁸ have both been found to reduce the rate of permeation through stainless steel. Again, however, these techniques have been tried only on a laboratory scale; there is no information about the effectiveness of these barriers in corrosive environments or in large-scale applications.

A second category of tritium barriers which will probably be required includes those which will be exposed to high neutron fluxes. These barriers may be

required in such locations as the blanket walls of a reactor. The blanket inner walls are exposed to very high (14 MeV) neutron fluxes, and it may be desirable to have some part of these walls impermeable to tritium. For example, water cooling is used for part of one conceptual design of a low activity blanket.⁹ Tritium contamination of water is very undesirable, so an effective barrier here is a necessity. Aluminum is proposed, but it has yet to be demonstrated that it can withstand a high (14 MeV) neutron flux and retain its low permeability to tritium.

Blanket outer walls may also be exposed to high neutron flux, although the neutron energy will be moderated by the blanket. Here the problem is similar to that of the heat exchangers mentioned earlier. The wall surface area may be quite large and the wall may serve as the principal defense against tritium leakage. The need for this kind of barrier is not so urgent, as the extreme neutron fluences of interest here will not be encountered in large scale until the demonstration reactor, at least.

All of these possible barriers will potentially need to be capable of withstanding the effects of flowing water, steam, liquid metals (principally Li), or salts [e.g., LiFBeF_2 (Flibe)]; high temperatures (up to 1000°C for some applications); and possibly high neutron fluxes (10^{14} – 10^{15} /cm²·sec). In addition, impurities, such as oxygen or water in a helium-cooled blanket or fluorine compounds in Flibe, may attack exposed surfaces. Such impurities are not only difficult to control, they may even be added deliberately, as in the case of oxygen in helium, to facilitate tritium recovery.

The barrier must maintain its integrity for at least a year, but preferably for 10 yr or more. It must be capable of large-scale fabrication on a production basis and must permit connections and penetrations to be made without breaching the containment. For the near term (up to 1985), the chief structural material for fusion devices will be stainless steel, so any barrier should be compatible with stainless steel or other materials which may be used in heat-exchangers.

There is an almost certain need, although at a later time, for detailed data on permeation of tritium through refractory metals at temperatures up to 1000°C. Niobium in particular is not well understood. No data exist on various refractory metal alloys, e.g., Nb-1% Zr, V-10% Ti; and at

the very low tritium pressures which are expected to be encountered, there is very little information for any material. There is no satisfactory current technology for fabrication of refractory metal devices on the scale of a fusion power reactor, so that it is certain that through the EPR-I stage of fusion machines, major refractory metal structures will not be used. The earliest machine which might reasonably use refractory metal structures is EPR-II or the demonstration plant.

LOW PRESSURE PERMEATION

There is need for tritium permeation data at low pressures, for example below one torr. Stickney¹⁰ has pointed out that there should be a deviation from the dependence of hydrogen permeation on the square root of the pressure when pressures are very low or temperatures are very high. Permeation measurements at pressures below 10^{-2} torr support this view.¹¹ The effect arises as a result of the fact that the solubility of hydrogen (at least in equilibrium conditions) is directly related to the partial pressure of monatomic hydrogen at the surface of the material in question. The permeability in turn depends upon the solubility and the diffusivity. The partial pressure of monatomic hydrogen at low temperatures and moderate pressures depends upon the square root of the total pressure of hydrogen and exponentially on the inverse of the temperature. Hence at high temperatures and/or very low pressures, the fraction of hydrogen that dissociates approaches one. At this point, the partial pressure of monatomic hydrogen becomes linearly dependent on total pressure and roughly independent of temperature. Both of these effects tend to reduce the solubility of hydrogen and hence its permeability below values extrapolated from lower temperatures or higher pressures.

As this effect may be large enough to reduce tritium permeation by several orders of magnitude at pressures below about 10^{-3} torr, it is important that permeation experiments be extended to low enough pressures to permit accurate predictions of permeation rates under reactor conditions. In addition, the effect of blanket materials on low-pressure permeation needs to be investigated. It is known, for example, that the presence of lithium deuteride can substantially increase the permeability of stainless steel to deuterium.¹²

HELIUM EFFECTS

A closely related problem is that of the effect of helium on the properties of structural materials. Helium in structural materials may be implanted by high energy ion bombardment or it may be created by the decay of dissolved tritium or by n, α reactions. The solubility and the diffusivity of helium in metals are both lower than for hydrogen in metal. As a result, the helium becomes trapped in the metal, creating bubbles or generating cracks. These effects can both weaken the metal and provide tritium leakage paths. Research is required on mechanisms to improve helium mobility and to reduce helium bubble formation. Physical barriers (for example, a carbon curtain) are good candidates to prevent ion implantation in first wall materials, but they must themselves be able to withstand the fast particle flux.

WINDOWS AND GETTERS

At the other end of the spectrum from permeation barriers are permeation windows. Some tritium recovery schemes depend upon a high permeability barrier between the liquid blanket and a tritium recovery device.^{13,14} The same low pressure effect on permeation, i.e., a dependence on pressure to the first power, that is beneficial in a barrier material would be a hindrance in this application; hence investigation of low-pressure, high-temperature permeation rates through candidate window materials is indicated. Here again composite materials may be required to prevent corrosion. Palladium, palladium-silver, and niobium are attractive window candidate materials.

In a permeation window application it is necessary that the tritium transport to the window be as fast as that through the window. The static film of liquid metal (or vapor) adjacent to the window may be a significant barrier to tritium flow because the tritium must diffuse across the film, while the bulk fluid is expected to be turbulent to promote heat and tritium transfer. An estimate of this effect in liquid lithium indicates that it is minor,¹³ but this estimate is based on extrapolations which could easily be in error by a factor of two. Errors of this size are sufficient to invalidate the conclusion. Data on the diffusion of tritium in liquid metals (Li, K), the high temperature viscosity of the same metals, and the low-concentration, high-temperature solubility of tritium in these metals are critical to the success of the concept of permeation windows.

In addition to windows, high permeability or diffusion rates are advantageous in materials which getter tritium, as a getter in which tritium can diffuse through the bulk of the material has much more capacity than one in which the tritium is trapped only at the surface; further work is required on mechanisms (e.g., fluidized beds) and materials which permit this rapid transport of tritium through the bulk.

Breeding materials other than liquid lithium also require more investigation. Flibe (LiFBeF_2), LiAlO_2 , LiAl , and other materials are essentially unstudied in the low-concentration, low-pressure regions required by inventory, safety, or environmental constraints. Both solubility and permeation/diffusion data are required for these and other breeding and gettering materials, particularly for pressures below 1 torr.

MECHANICAL PROPERTIES

The success of future fusion reactors will depend not only upon tritium containment, but also upon the ability of structural materials to withstand the extreme demands placed upon them. One of the well recognized sources of mechanical failure of metals is hydrogen embrittlement. Since hydrogen as tritium and deuterium is the fuel for a fusion reactor and since it must be stored, transported, heated, cooled, purified, and otherwise processed, there are many opportunities for the deleterious effects of hydrogen in metals to occur.

Three criteria have been suggested as influencing an upper limit to the quantity and concentration of tritium that can be allowed in a fusion reactor. The first is that of cost and inventory. Although this is very important now, it becomes the weakest criterion once fusion reactors are available to breed the tritium for new reactors. The second is the question of embrittlement of structural materials exposed to tritium, particularly in the blanket first-wall region. This is a somewhat more restrictive criterion. Fraas¹⁵ suggests that perhaps 100 ppm of tritium could be allowed in a liquid lithium blanket. The third is the necessity for close containment of tritium to ensure environmental safety. At present, the tritium concentrations dictated by this requirement (i.e., ~1-10 ppm) are low enough that the other two requirements are automatically met.

If, however, more effective barrier materials, as were discussed earlier, are developed, it is possible that inventories could rise and that embrittlement of the

blanket and first-wall structure could become a significant problem. It has also been suggested that, as the diffusivity of hydrogen in metals is high at the reactor operating temperature, tritium will diffuse out of the structure during cooling thus preventing embrittlement. The possibility of a rapid shutdown with the structure cooling too fast to permit the escape of the tritium should be considered, however, before breeding blankets are used. Because of the much smaller quantities and concentrations of tritium in nonbreeding machines, this does not appear to be a significant concern in EPR.

Outside of the blanket region, the temperatures and pressures of tritium-containing systems are not extreme except in the cryogenic processing systems, so that there is already available a substantial base of tritium compatibility information which is relevant to these systems.

Tritium is known to degrade glass through a process in which the ionizing effect of the tritium promotes the creation of hydroxyl ions in silica.¹⁶ This results in the conversion of silica to elemental silicon. This effect has not been studied at very low or very high temperatures. Since insulators - usually glass or ceramic - are frequently required to carry electrical signals into or out of tritium systems or, as in the θ -pinch, are part of the reactor, additional information is definitely required on this effect. For high radiation flux applications such as the θ -pinch machine, the additional damage caused by the radiation should be studied as well.

Embrittlement of structural materials by helium has been discussed earlier in connection with permeation. Embrittlement by helium is expected to be a problem wherever (1) the solubility of tritium in a metal is high, or (2) where the helium may be implanted in the metal or created in an (n,α) reaction. These are high-temperature, high-flux areas of the reactor, particularly the first wall, but they also include the structure and containment of the blanket and primary heat exchangers. Further work is required on embrittlement or other damaging effects caused by helium in stainless steel, refractory metals, and possible composite or oxide barrier materials. Particular attention is needed to prevent the possible creation of leakage paths in low permeability materials.

Theoretical studies of the effects of high neutron fluences on structural

materials indicate that after extended service in a high intensity 14-MeV neutron flux, the character of a metal, e.g., stainless steel, can be considerably altered. There are few if any data on the effects of these changes on the mechanical properties of these metals, although high displacement rates are reported to enhance the diffusion of hydrogen through materials.¹⁷ In particular, with regard to tritium, information is required on the effects of transmutations and high displacement rates on the permeability of stainless steel and refractory alloys to tritium and on their resistance to hydrogen embrittlement.

BREEDING AND RECOVERY

Some of the principal problems associated with tritium breeding or tritium recovery have already been mentioned in relation to permeation. For example, it was noted earlier that at low pressures there is expected to be a deviation from a dependence of the solubility of tritium in metals on the square root of the pressure. There is a certain need for experimental data of the low-pressure (less than 1 torr) solubility of tritium in breeding materials. Such data are necessary in order to predict inventory requirements, tritium permeation loss rates, and recovery rates.

A closely related question is that of determining equilibrium constants and rates of reaction between tritium and breeding or gettering materials. In a liquid-lithium blanket, for example, the tritium can be in the form of T_2 or LiT , and it may be in the vapor or dissolved in the liquid. Knowledge of the appropriate equilibrium constants is essential for determining the distribution of tritium within the blanket. The tritium distribution in turn determines how and whether the tritium may be recovered from the blanket.

Reaction rates are required in order to determine the scale of recovery processes, and also whether recovery will occur in the presence of competing processes. Many tritium-recovery schemes depend upon the chemical reaction of tritium with another material so that the tritium is fixed (e.g., yttrium getters) or its equilibrium pressure is reduced (e.g., oxidation in a helium-cooled blanket).¹³ In the former case, small amounts of impurities may coat the surface of the getter, causing the gettering reaction to be too slow to be effective. In the latter case, the oxygen necessary to convert the tritium to water may also attack structural materials (e.g., niobium) so

that either the oxygen is lost or the structure is damaged. The kinetics of these processes are not well known in the temperature and pressure ranges of interest, so that accurate predictions of the behavior of these systems cannot be made.

INTERACTIONS RESULTING FROM TRITIUM DECAY

This has already been covered in the section on mechanical properties, where the effects of helium generation in metals and of tritium in insulators are discussed.

ENVIRONMENTAL, HEALTH, AND SAFETY CONSIDERATIONS

There is already a large source of low-level tritium waste in fission reactors, where tritium is bred in cooling water and is released during dissolution of fuel elements in fuel reprocessing plants. In a fusion reactor, as well, low level tritium waste, particularly in aqueous streams, constitutes a source of tritium release to the atmosphere which is extremely difficult to control. Tritium in gaseous streams is relatively simple to control, as either condensation or oxidation followed by absorption of the tritium is effective in separating the tritium from other gases. Tritium (principally HTO) in water, however, is very difficult to remove as it is in the same chemical form as the water. At the very low concentration levels which are characteristic of aqueous tritium waste ($T/H < 10^{-7}$), the energy cost of separating the tritium is large relative to the amount of tritium extracted. This is true of even the most effective available separation processes - water distillation, electrolysis/cryogenic distillation, and the GS process.¹⁸ Further research is required on advanced isotope separation processes for hydrogen, particularly laser separation, catalytic exchange, and cryogenic distillation. Each of these processes offers the potential of considerably lower cost per unit of tritium recovered than do conventional methods; the first two are particularly suited to recovery of tritium from water, whereas the latter requires hydrogen gas.

It has been observed that there is considerable variation among humans in the ability to absorb and retain tritium, but no model exists to explain these effects beyond the obvious correspondence between biological half-life and body water intake. Further work on this

subject would be of benefit in protecting tritium workers in the event of large exposures as well as possibly providing insight to mechanisms for tritium concentration or dilution in other organisms.

A related question is that of the genetic and/or somatic effects of long-term exposure to tritium. Little is known with certainty about the effects of chronic tritium exposure. Tritium is known to appear preferentially in certain positions of complex molecules, such as DNA for example, but the transmutations caused by the decay of tritium to helium-3 are said to be either efficiently repaired or extremely rare.¹⁹ On the other hand, it has been estimated that even in a fission reactor tritium represents the principal health hazard of all the radionuclides.²⁰ Further research on the biological effects of chronic tritium exposure is required to properly assess the degree of risk involved in the release of tritium to the biosphere.

HANDLING AND TRANSPORTATION

The construction of tritium-burning reactors will require tritium-handling and transportation equipment on a much larger scale than presently exists. There is a great need for the development of equipment for circulating and pumping tritium and tritiated substances. In the vacuum range, e.g., below ~1 torr, mercury distillation pumps and sorption pumps are both effective in pumping tritium. Mercury pumps, however, must be carefully trapped to prevent mercury migration to other parts of the vacuum system. Organic diffusion pump fluids (with the possible exception of polyphenylether) are degraded by exposure to tritium and turn to a gum, which fouls the pump. Sorption pumps are fast and very clean, but they must be operated intermittently, steadily accumulating pumped gas until they saturate or are regenerated. The accumulated gas represents a major contribution to the tritium inventory in a reactor, so it is important to develop new pumps or new pump cycles that permit the ratio of pump speed to inventory to be increased.

At intermediate pressures, e.g., 10^{-2} to 10 atm, there are few mechanical pumps available with which to pump tritium. Specially constructed vane pumps for circulation and vacuum-pressure pumps have been developed for tritium use, but no reliable well-tested pump exists. Pumps are required which will reliably handle kilogram per day quantities of nearly

pure tritium for periods of at least a year without contaminating the tritium or requiring replacement. Improved radiation-resistant pump fluids, both for mechanical and diffusion pumps, would be extremely valuable.

Sorption pumps may be used in the intermediate pressure range as well. These may operate by physical adsorption, generally as cryogenic pumps, or by chemical absorption in hydride-forming materials such as uranium or palladium. The possible variety of pumping materials is large, but, as in the case of vacuum sorption pumps, intermittent operation and large tritium inventories are a weakness of these pumps. In addition, it has been noted that the presence of helium-3 or other nonreactive gases in a tritium stream interferes with the ability of some materials to absorb tritium.²¹ These effects need to be better understood and, where possible, eliminated.

The enormous size of a fusion reactor and its containment structure requires development of atmosphere detritiation systems many times larger than any currently in existence. An orderly program of development is required to ensure that these larger systems are properly scaled. Standards must also be established with regard to the relative scale of these containment and recovery systems compared to that of the contained process. Decontamination level standards must be established with an eye to permissible activity levels for both routine and accidental releases.

At present there are few standards for the selection of materials for use in tritium systems. Even among tritium workers, there is disagreement about preferred materials for a given application. Such standards must be developed and refined as improved knowledge of the effects of tritium on materials is obtained.

There is no agreed-upon means for storing or transporting tritium. Three forms have obvious appeal because they minimize the volume of tritium - high pressure gas, liquid tritium, and tritium compounds, particularly metal hydrides. Each has a major disadvantage. Gas storage involves high pressure and a much larger volume devoted to tritium than do the other two. Liquid tritium requires cryogenic processing and storage facilities and involves a risk of tritium loss in the event of a refrigeration failure. Metal hydrides are quite massive and frequently involve a risk of pyrophoricity. Further work is required to better define the relative

benefits and risks of various storage media, and standards must be developed for storage and transportation of tritium.

PHYSICAL AND CHEMICAL PROPERTIES

Little is known of the behavior of tritium or of heteromolecular hydrogen containing tritium, i.e., HT and DT, in the liquid or solid state. Knowledge of the vapor pressures of these species and their mixtures is needed for the design of cryogenic distillation systems and for an understanding of fractionation effects associated with the fabrication of liquid or solid droplets of D-T mixtures for plasma fueling.

In addition, the exchange of hydrogen isotopes between the various homonuclear (H_2 , D_2 , T_2) and heteronuclear (HD, HT, DT) molecular forms requires further investigation. Both the rate of exchange and the equilibrium constant for such reactions are known to be affected by the concentration of tritium and by pressure, temperature, and impurities. Better knowledge of these effects and of possible catalysts which are effective in promoting molecular exchange at low temperatures (<77 K) would permit improved efficiencies in isotope separation processes.

Better knowledge of isotopic exchange in compounds is required as well. One laser isotope separation scheme, for example, requires the use of a scavenger to remove or react with laser-excited tritium to prevent its recombination with the bulk of the material from which it is being separated.²² Thus, information on chemical reaction rates of tritium as HT, T_2 , T, or T^+ with other substances such as H_2 , H_2O , HCl would be of interest. Further information is also needed on molecular spectra of tritiated compounds which might be used in isotope separation processes.

Tritium-hydrogen exchange rates and the effects of the low energy beta radiation or of decay to helium-3, in organic materials, are needed. Particular emphasis is required for lubricating and/or sealing materials.

As was discussed earlier under environmental considerations, improved isotope separation methods are required, particularly for recovery of very low concentrations of tritium.

It has been observed that a common impurity in tritium is methane (CT_4), which is apparently produced by reaction of the

tritium with carbon present in organics or in small amounts in structural materials, e.g., stainless steel. High-Z impurities in fusion reactor fuel cause large power losses in the plasma through line radiation during the heating period. These losses may prevent the plasma from reaching ignition, hence it is important to keep fuel impurities at a very low level. More information is needed on the process by which methane or other low-level impurities are created in the presence of tritium and on ways in which they can be inhibited.

INSTRUMENTATION, MONITORING, AND ASSAY

In order to measure and maintain the very low levels of tritium required in the blanket and cooling systems of a fusion reactor, accurate, highly sensitive tritium monitors must be developed. For gas streams, such as helium coolant streams, adequately sensitive tritium monitors exist, but their use at the high pressures and in the presence of large radiation levels has not been demonstrated.

In liquid metals, the situation is not as good. Prototype tritium monitors, which depend upon tritium diffusion across a metal membrane, have been developed for use in molten sodium, but no commercial instruments are available. For liquid lithium, however, because of its very great affinity for tritium, diffusion through a membrane is inadequate at the desired concentration and no satisfactory monitor exists. Research is needed to develop a tritium monitor which will detect tritium at the 1 - 100 ppm level in liquid lithium at 500°C.

Tritium monitoring is also required in high-level streams in order to monitor isotope separation processes, for example. There are no real-time high-accuracy tritium monitors available for this purpose. Ionization chambers have been developed with accuracies of $\pm 2\%$,²³ but these are not commercially available.

The effects of impurities and temperature or pressure variations on the accuracy of these instruments need to be understood well enough to permit accurate monitoring in the event of process fluctuations which may affect these variables.

Timetable for Research

All the various technological developments which I have discussed may be related to the CTR proposed timetable for fusion power development. There are, as I mentioned earlier, four major milestones and two secondary milestone experiments which relate to specific fusion problems. In the order of their initial operating dates, they are TFTR (1980), INS (1981), FERF (1985), EPR-I (1985), EPR-II (1990), and DPR (2000). I shall summarize here the technological developments which will be required for each of these fusion experiments. A major assumption in what follows is that new tritium technologies must be successfully demonstrated on a pilot scale before they are incorporated into any of the reactors in the EPR-I - EPR-II - DPR sequence.

The TFTR is intended to be a machine that can be built and operated successfully using currently available technology. Thus no research or development is required for the TFTR. The INS, likewise, represents current technology almost entirely. The heat exchanger which cools the deuterium jet in the INS may represent an opportunity to test permeation barrier materials on a process scale; but the success of the machine does not depend upon advances in this area. The same is true of the cryogenic isotope separation which will be employed; im-

provements here are desirable, but not necessary.

The FERF has a much larger tritium inventory than INS, and the wall temperatures and radiation loadings are higher as well. As a result, there are several areas where technological improvements are desirable. Further work on barrier materials and isotope separation is particularly desirable, as large quantities of tritium will be implanted in the reactor wall, and the D-T separation system will be the first system of its size equipped to handle tritium. Many of the concepts discussed under handling and transportation and under physical and chemical aspects could contribute significantly to FERF. Particularly important are circulating and vacuum pumps in the former category and isotope exchange effects in the latter. The nature of the FERF is such that although the permeation barrier problem is quite important, there are no critical technological hurdles that must be crossed.

EPR-I requires much of the same technology as the FERF. In addition, because breeding modules may be tested on EPR-I, information on breeding and recovery technology and on low pressure permeation through stainless steel is needed in time to test new concepts on EPR-I. The large inventory and tritium consumption rate will

require improvements in tritium monitoring and assay. Because of the publicity which will undoubtedly surround EPR-I, it is important to have made substantial progress in determining the biological effects of tritium on large populations.

EPR-II appears to be the logical focal point for most of the tritium technology requirements discussed in this report. There is a critical need to develop an adequate tritium containment technology before EPR-II construction can begin. Hence permeation barrier technology and/or low pressure permeation data for stainless steel are critically important here. Similar advances for refractory metals will be quite valuable. Tritium inventories and flow rates will be large enough to make the isotope separation and the instrumentation, monitoring, and assay technologies critically important. Mechanical properties data will be necessary, though not critical, as will data on high neutron flux resistant permeation barriers, helium effects in materials, window and getter materials, breeding materials, and recovery technology. Continued progress in understanding the environmental effects of tritium is important.

By the start of the final conceptual design of the demonstration reactor, there will be a critical need for demonstrably successful, large-scale solutions to the problems of containment in both stainless

steel and refractory metals (either by barriers or better low pressure permeation rates); effects of tritium and helium on the mechanical properties of materials; the breeding and recovery of tritium; isotope separation; and instrumentation, monitoring, and assay of tritium. Handling and transportation problems may possibly be postponed a few more years until the Title I stage of DPR, but these problems, too, are critical. All major environmental questions must be settled, and although it is not expected to be critical to the success of the reactor, information on tritium barriers for high neutron flux applications and on permeation windows is probably quite important.

The various tritium technology requirements are summarized in Figure 1, which shows their relationship to the CTR milestone experiments. The requirements are ranked according to their importance and according to time. A rank of 1 indicates that there is a critical need for new information or new technological development. Failure to meet this requirement could prevent a given device from operating. A rank of 2 indicates the need is significant; substantial economic or environmental costs may be associated with failure to meet this requirement. A third rank, indicating desirable information, is not explicitly noted as information on any of the needs I have discussed would be of benefit to the CTR program.

Conclusions

Most of the tritium-related problems that must be solved to ensure the success of a demonstration fusion power reactor in the year 2000 need to be approached with the goal of major progress by the early 1980's in mind. This approach is based upon the assumption that EPR-II, scheduled to operate in 1990, will be required to simultaneously demonstrate tritium breeding; reliable, if not economic, power generation; and adequate tritium containment. The critical problems are those of creating effective barriers to tritium permeation at high temperature and low pressure, large-scale isotope separation systems, and accurate monitoring and assay systems. Substantial progress should have been made on these problems in time to permit preliminary testing on EPR-I.

In addition, significant progress by that time is required in understanding the effects of helium and tritium on mechanical properties of likely reactor materials, principally stainless steel, and on biological systems. New or more accurate

data are required on physical and chemical properties relating to isotope separation, fueling, and tritium recovery processes.

Progress in these areas and in tritium handling and control technology is important to EPR-I as well. These are not critical in EPR-I, but lack of progress could cause substantial environmental or economic penalties. All of the areas discussed are critical to the demonstration power reactor, either because the machine will not operate or will not be capable of producing power at a competitive cost without improved technology, or because otherwise its environmental impact will be unacceptably large.

Because fusion design concepts are still in a rapid state of flux and the majority of the work is needed seven or more years hence, these technology requirements will need to encompass nearly everything that has ever been mentioned about tritium technology for fusion reactor systems. They will need constant revision as new fusion reactor concepts are created.

FIGURE 1 - Technology requirements for fusion reactor developments.

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