Tritium Issues in the HAPL IFE Reactor (U)

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Abstract

The High Average Power Laser (HAPL) Inertial Fusion Energy (IFE) Reactor (abbreviated as HAPL herein) is a direct-drive inertial confinement fusion reactor, which will utilize a deuterium-tritium (D-T) fuel mixture. This reactor will process approximately 25 kilograms of tritium (T) per year when operating at nominal design levels. This document discusses the tritium related issues involved with the entire HAPL facility arising from the need to prepare fuel pellets for the reactor, recover the unburnt tritium for recycle, and breed additional tritium for subsequent use. Tritium will be found in the HAPL facility in four phases (liquid, solid, gas, and plasma) and will be located in free ions, free molecules, and bound in solids and liquids. All of this tritium will either be fused, recovered for further use, or disposed of as radioactive waste. In this document we discuss the conceptual tritium process designed by a team headed by PPPL, and propose additional processes for tritium recovery and handling for tritium located in the breeder blanket and bound in structural parts of the reactor. We also discuss certain materials issues associated with materials currently designated for use in HAPL arising from tritium effects on those materials. We also present some recommendations for needed research into these processes and materials.

Introduction

The HAPL IFE Reactor has been under design for some time and periodic workshops to discuss design issues and plan research responses to them have been held under the auspices of the Naval Research Laboratory (NRL) since early 2001. The majority of the information presented at these workshops is archived at the HAPL Website (http://aries.ucsd.edu/HAPL/) and will not be discussed here. At this point, major physics issues have been resolved and the HAPL community is ready to begin consideration of the research and development issues involving the reactor's support facilities. These support facilities will be required to prepare and introduce fuel to the reactor, to recover unburnt fuel from the reactor, to recover new tritium produced in the breeder blanket, and to recover tritium from reactor parts. It is most likely that technology to meet these requirements exists today, but the unique combinations needed for the specific HAPL facilities may require additional R&D to work out details, especially in the situations where one technology will serve two or more functions, in order to keep the HAPL facility operating at or below regulatory and release limits. In the event that problems are discovered, new technologies may have to be developed and applied.

The Savannah River Site (SRS) has designed and started up several major tritium handling processes in the recent past. Some of these are: the Tritium Extraction Facility, a facility designed to extract T2 from irradiated fission reactor fuel rods; the Tritium Facility Modernization and Consolidation Project, a project designed to move several existing processes from the now-closed old tritium processing building to the H-Area New Manufacturing Facility; and the New Manufacturing Facility itself, a metal hydride based tritium purification, storage, and handling process. Normal operations of the SRS Tritium Facility, which is collection of the above listed facilities plus support facilities, has included a wide range of chemical processes over the years: cryogenic distillation, high pressure and low pressure tritium handling, handling of leaks and low T content gas streams, etc. Additionally, several large and small processes have been shut down and in some cases disassembled, which has provided SRS with large-part disposal experience. In all these examples, tritium inventory had to be determined or tracked and appropriately handled.

As well, SRS started up the Defense Waste Processing Facility to handle radioactive slurry waste accumulated from the many years of plutonium Plant operation. That facility is relevant to the HAPL because a) it is a remotely operated facility, and b) it has the capability to handle large contaminated pieces (10 foot tall stainless steel containers of radioactive contamination containing glass) and to store them for a cool-down period, prior to shipping to a final waste repository. Also, SRS for several years operated Separations Facilities that used extensive remote handling operations and robotics to provide materials for the national defense. Besides the tritium expertise found at SRS, this robotics and remote operations experience should be useful to HAPL as well. These skills are now being employed in the recently started up Tritium Extraction Facility, and will certainly be used in the HAPL.

The Savannah River National Laboratory (SRNL) has several R&D groups that routinely address issues in the tritium handling facilities at SRS in new process design and current process support R&D, shutdown, and startup activities. In addition, the Tritium Facility (TF) itself has an associated engineering organization that supervises the TF processes on a daily basis, addressing normal problems that arise, and calling on the SRNL R&D groups for support when needed. These two large entities provide a solid resource on tritium handling and have been extensively probed in the writing of this document.

This paper's intent is to provide a distilled list of issues and typical solutions that routinely impact the TF here at SRS, and to point out where and how these issues impact HAPL. The collective SRS experience allows the construction of a short list of typical tritium issues (Table 1). They can be broken down into general groups as shown (alternative methods can also be used). We feel that SRNL and SRS have considerable practical experience and R&D capabilities that can be brought to bear on HAPL design issues, and we discuss these issues more fully below.

Table 1. General Tritium-Related Issues in HAPL Tritium Handling Facilities

Regulatory/Release requirements

Tritium confinement
Tritium safety
Tritium packaging, handling, shipping, and receiving
Tritium storage
Tritium processing/purification
Tritium security/accountability
Procedures for Tritium operations & maintenance

HAPL Processes

Fueling - D/T target fabrication, charging, & injection
Exhaust processing- separation of D/T/He/contaminants
Tritium breeding targets/tritium breeding test modules
Tritium extraction from targets/test modules
Isotopic separation
Instrumentation/diagnostics for D/T
Environmental monitoring for tritium

Materials Effects/Properties

Tritium effects on materials (T2 retention, He embrittlement, welding irradiated materials, etc.)
Tritiated waste handling and disposal
Tritium offgasing during startup & operation
Tritium extraction from irradiated components
Tritium permeation and permeation barriers

General Issues

In any chemical process and/or reactor operation, there are four general operating conditions (modes) that must be considered during design; startup, shutdown, nominal operation, and maintenance modes. Shutdown (and possibly maintenance) mode needs to be subdivided into planned and unplanned. (The unplanned shutdown mode is the most problematic, as this tends to occur due to unforeseen events.) Tritium issues relating to these modes are numerous. For example, during startup it is presumed that all or some of the reactor now consists of newly installed parts. These parts will typically have fresh oxide/sulfide coating from air exposure which will interact with gas phase tritium (of either the elemental or oxide form). Thus, a significant T inventory can be consumed in

these reactions, which will normally decrease as the parts see normal use. Likewise, an unplanned shutdown may well be due to new leaks into the reactor chamber (maintained under low vacuum), which will lead to nitrogen and oxygen introduction into the system. Oxygen will react with tritium to form tritiated water which will have to be removed from the system and probably recovered (or else discarded). The HAPL tritium handling processes will all have to be protected and designed for as many off-normal conditions as can be deemed reasonably possible.

The HAPL Tritium Facilities will have to perform several process functions. These include facilities to prepare fuel pellets for the reactor, recover the unburnt tritium for recycle, and breed additional tritium for subsequent use. As well, facilities to handle the inevitable radioactive waste (unrecoverable tritium and tritiated/activated parts) will have to be built. Each of these functionalities will have to be carefully designed, with considerable R&D invested to prevent unpleasant surprises during startup and normal operations. The key to a successful design is knowing what you are designing for, i.e. the input streams to each facility and sub-facility must be well defined for a successful design effort.

The initial step of conceptually designing the fuel recovery system (FRS) has already been taken, with a joint design being issued for consideration by a team headed by PPPL, with contributions from SRNL, SRS Tritium Engineering, and others, under funding supplied by the HAPL Program. Several minor issues remain that suggest some needed R&D efforts to further quantify the behavior of selected components of the conceptual process.

The tritium recovery process from the HAPL breeder blanket has not been completely defined. This is primarily because the actual breeder blanket material is not yet chosen. The leading candidates for this material are a Pb-17%Li blanket material or a “Flibe” (BeF2-(LiF)2) breeder material contained in either a W on stainless steel tube or a SiC tube. However, also under some consideration at this time are moveable graphite tiles which will contain Be for breeding T. These would not be contained in a tube, but would be supported by rails in the reactor core. Pending the final decision on the actual materials to be used, a conceptual process design would be premature. However, some facts can be ascertained at this point, and thus it is possible to make some generic statements about a putative tritium recovery system. Clearly an extraction process will be required to remove the tritium from the breeder material. The tritium will primarily be in the elemental form and may be processed in bulk in fashions similar to other existing tritium extraction facilities. This generic Tritium Extraction System (TES) and related issues will be discussed more below.

The process of imploding plastic shells filled with a D-T (deuterium-tritium) mix via high power laser energy produces a plasma wherein the D and T fuse, releasing energy that will then be converted to electrical power. This process produces copious ions that are expelled from the fusion zone and strike the various parts of the reactor, usually with

enough energy to be significantly implanted in those materials (i.e. ion implantation). These ions include T ions, and thus the reactor parts become tritium reservoirs by virtue of ion implantation. This T inventory can potentially become quite high, and thus will have to be handled, either by a recovery process or by a waste disposal process, or both. The objective of such a system, which could be called a "Tritium in Solids Recovery System" (TSRS), would be to minimize waste and maximize recovery. This process will have to handle both large reactor parts and residual powdery material, produced by erosion and exfoliation of the first wall material and armor, removed from the reactor. Both types of materials will have to be heated to extract T in a similar fashion as the TES, however the details of this process may vary considerable depending on the shape and size of the materials presented to the TSRS.

Besides the tritium processing aspects in HAPL there are also tritium-induced materials properties changes that need to be addressed. One working rule of thumb with tritium is that it gets everywhere. Tritium migration to unexpected areas should be carefully considered, as at the elevated temperatures of the HAPL reactor core, tritium will be very mobile. T should be expected to reach and permeate into portions of the reactor besides the first wall, and it is likely that that T will be very difficult to recover. The majority of parts permeated with T will most likely have to be disposed of as radioactive waste. Since considerable amounts of T will most likely be lost to these unrecoverable materials, the impact on the required minimum breeder ratio will need to be carefully estimated. As well, T permeation, which typically is significant even at as low as 200 ^{o}C, will require the designed facilities to have secondary containment on most heated parts (which the FRS Conceptual Design (CD) does).

A Tritium Loading Facility (TLF), which will load the plastic shells with a D-T mix that will freeze on the inside walls, is required and as yet undesigned. An annealing process designed to smooth the inner surface of the D-T ice will also be employed. A unique feature of this process will be the use of cryogenic temperatures to keep the D-T mix in a solid form inside the plastic shells. This will necessarily place thermal stresses on various points of the process vessels and piping, which brings up its own issues, e.g. cracking. But since the systems must be cooled, a heat transfer fluid of some sort will be used, and tritium permeation into that medium must also be considered. The issue would arise if tritium is held in TLF vessels and pipes for any extended time, allowing permeation to occur.

A separate potential issue is tritium exchange with protium in the plastic shell material itself. This would reduce the purity of the D-T ice by introducing H, and negatively impact the fusion process. The basis for this concern is discussed further below.

Tritium permeation into metallic parts invariably has deleterious effects on those metal parts' performance. This is primarily due to T decay which produces He in the bulk, but H-embrittlement is another distinct possibility. The use of ferritic steel in the blanket and armor support system is a case in point. Research has shown that ferritic steels are more

susceptible to tritium damage than austenitic steels, but the higher nickel content of austenitic steels raises a neutron activation products issue. The T impact on ferritic steel part's lifetimes need to be included in the operating cost estimates for HAPL, as replacing embrittled parts might be a significant maintenance operation requiring considerable downtime, and would generate more solid radioactive waste than otherwise expected. Also, the embrittlement may focus in stressed regions of the steel as opposed to being distributed evenly. This would accelerate the damage in those regions, and such regions are often critical to the mechanical integrity of the part.

Tritium may also interact with constituents of the reactor parts to form other chemicals that may introduce new issues as well. One classic case would be the interaction of dissolved H (or D or T) with C to form methanes. These methanes are formed in the solid and agglomerate into bubbles just as He does, sometimes causing blistering of the metal surfaces. Some of these blisters can vent, potentially producing more particulates in the reactor in a process known as methane embrittlement. (Whether higher hydrocarbons are formed, i.e. ethanes, is an open question.) Likewise, high O contents can react this way in a process known as steam embrittlement. As well, most steel parts have a dissolved H content which will come out as the part's temperature is increased, producing H-containing species in the gas stream in addition to the H from the plastic fuel shells. Thus early reactor gas streams may have a different isotopic distribution that later ones, where the H has been driven off and replaced by D and T during cooldowns. Note that every new part placed in the reactor will go through this 'seasoning' process. And finally the question of how much tritium ends up further back in the reactor, i.e. in the laser ports and on the optics, remains an open question.

Specific Issues

Gas Phase Tritium

Tritium recovery from the reactor effluent gas phase is addressed quite well in the Conceptual Design (CD) produced by the PPPL-headed team. Only a few minor residual issues have been identified in this process, some of which may cross the boundary into items that will be handled at the next design stage. They are:

- Are the Ni on kieselguhr (Ni/k) beds adequate to meet the design goals specified? Will they handle all input stream conditions anticipated in all operation modes? Will the CO/CO2 removal be adequate?

- Is adequate time allowed in the nominal scheduling for In-Bed Accountability?

- Is a Thermal Cycling Absorption Process (TCAP)-variant process really excluded from consideration or would such be feasible? (TCAP is the chromatographic process used for isotope separation at SRS.)

- Is the plan for handling tritiated water collected on the molecular sieves adequate?

- Will charcoal filters make methanes (and is the CD adequate to handle the implied load above that from the plastic shell material)?

- Will the cycle life of the ZrFe getter beds be adequate?

- Does O2 removal need to be considered more thoroughly in the CD?

- Should we use a Pd-based material in the Room Detritiation System (RDS) beds to avoid the requirement for hot standby status 24/7?

- What happens to the RDS cleanup capability for low level leaks if a prior large leak has been processed. i.e. will the residual contamination in the RDS from a large leak back-contaminate a room?

- Generically, have all dual-use subsystems been examined for this kind of 'high-then-low-T" issue?

- Is a RDS needed if the facility could be sited where minor releases were not important?

- Is a Glovebox purge system specified and/or needed?

- Are the heaters placed close enough to the beds in the Bed Regeneration System to avoid significant heat loss in the gas lines prior to the bed? (The CD seems to show them separated from the beds.)

- Do we need a H2O/T monitor at the start of the Glovebox Cleanup System as well?

- Have the special electrical requirements for operation in Ar-inerted environments been considered adequately?

- Will RTDs be used?

- How well will the FRS pumps survive T and/or particulate exposure?

Solid Phase Tritium

Tritium will exist in several solid phases in the HAPL. The most obvious is in the breeder material. The next most obvious is in the ion-implanted zones of materials that face the plasma/fusion region. A less obvious place would be in or on particulate matter produced by the various chemical/physical processes in the reactor, such as exfoliation and erosion. Perhaps the least obvious is in tritium-permeated reactor parts. Because no CD exists for facilities to handle these materials, the identified issues are much more

generic. Certainly, the largest issue is how the facilities developed to handle each of these T-containing solids will be designed. Some specific, high-level concerns are:

- Can the FRS process be modified to handle T from the breeder material T extraction processes, or is a separate TES needed?

- Will breeder material contaminant of the TES streams, e.g. lead, adversely impact the typical hydride bed materials used in anticipated T processing streams?

- What is the estimated rate of particulate production, and what is their T content? (Note that this question is applicable to each base system, i.e. FRS, TES, TSRS, etc.) (This also has significant impacts on contamination control procedures, which can impact downtimes for maintenance and repair.)

- Should particulate filters in the FRS be placed closer to the exit of the reactor to protect downstream parts and processes and concentrate the particulates for easier handling?

- An ancillary issue is: Will particulates collect in the ion dumps proposed for magnetic intervention and significantly alter the dumps' performance?

- Will a 'cool-off' facility be required for tritiated metal parts from the reactor? Can those parts be processed immediately to remove/recover tritium?

- Will there be significant T offgassing from reactor parts upon shutdown, and what does this imply for downtime extents?

- During shutdown, will the chamber be backfilled and what with (presumably not air due to possible tritiated ammonia and water formation)? How will that gas be processed after a return to normal operations reduces chamber pressure again? Is an emergency shutdown blanket facility needed to rapidly fill the chamber with inert gas?

Tritium Compatibility/Effects

Tritium compatibility issues with metals are well known. Tritium will permeate through most metals to some extent, and then its decay will produce an internal He atom. These atoms are highly insoluble and will aggregate into bubbles in the metal which can significantly alter structural characteristics, especially since initial aggregation typically occurs around grain boundaries and defects. However, hydrogen itself can cause other compatibility problems, and can aggravate other compatibility problems that may be present. Even for non-metallic materials, issues exist. Tritium can stimulate H exchange in plastics, and can cause plastics to begin releasing large amounts of hydrogen. Plastics containing aromatic moieties are more resistant to the excessive release problem, but recent research at SRS has shown that even Vespel, a polyimide polymer, will exchange.

This is shown in Figure 1, where the %H in T is plotted as a function of exposure time for three plastics. Figure 2 shows the test cell pressure, which for Vespel and Teflon remained nearly constant, while for UHMW-PE, increased dramatically.

In general these processes lead to material property changes that are usually detrimental. Thus any plastics potentially exposed to tritium need to be assessed for compatibility issues as well. As a side issue, the question of how much exchange into the plastic fuel pellet material will be seen, and whether it is significant to fusion yield, needs to at least be considered. The SRNL studies on T exchange into plastics (Fig. 1) were not conducted on a short enough time frame to directly assess this problem. But the observed rapid initial rate of exchange could potentially have an impact on fuel purity if it is fast enough, or if it stimulates excessive H release.

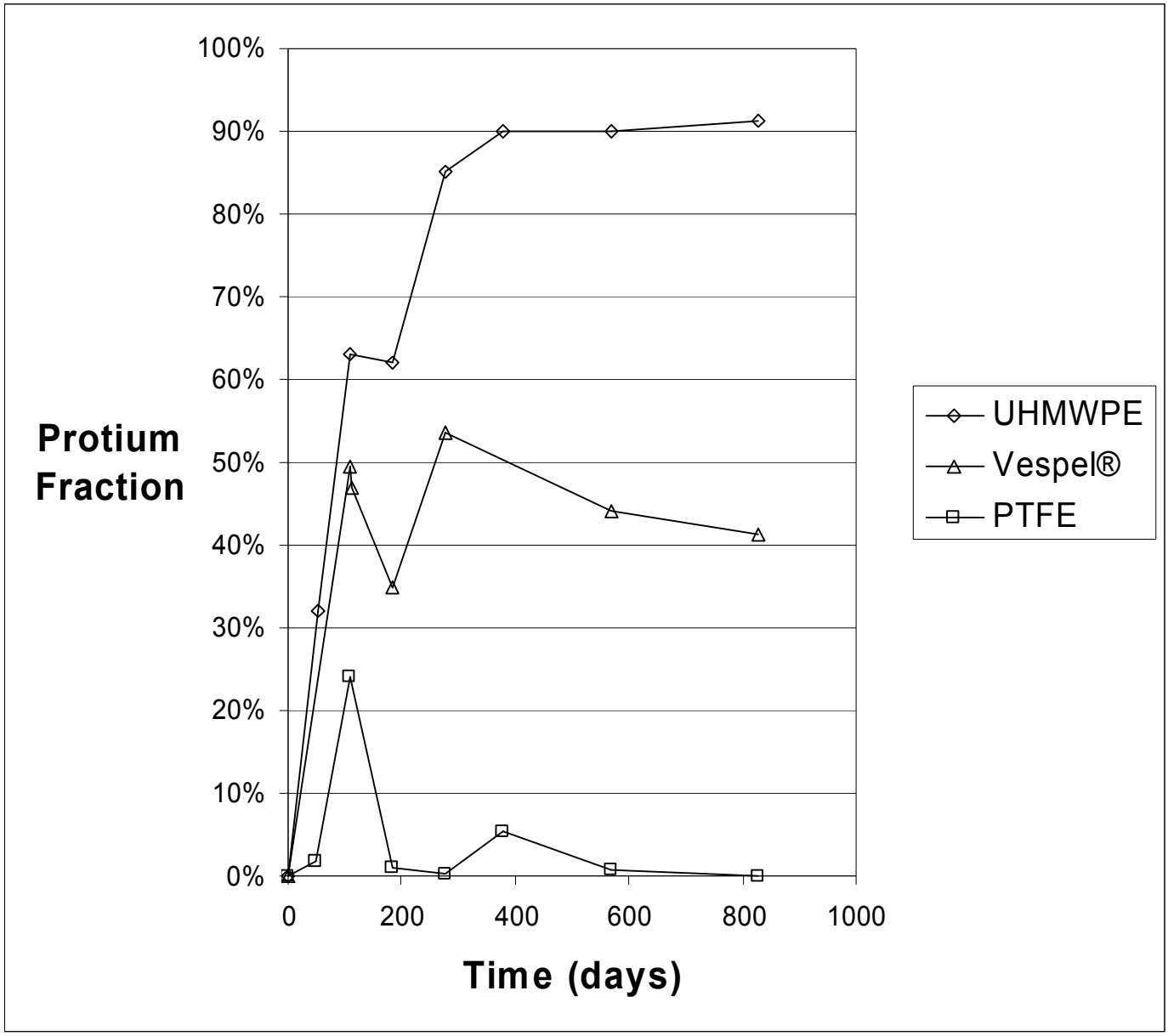

Fig. 1. Fraction of gas detected as protium, as function of exposure time. UHMW-PE, Vespel®, PTFE as marked.

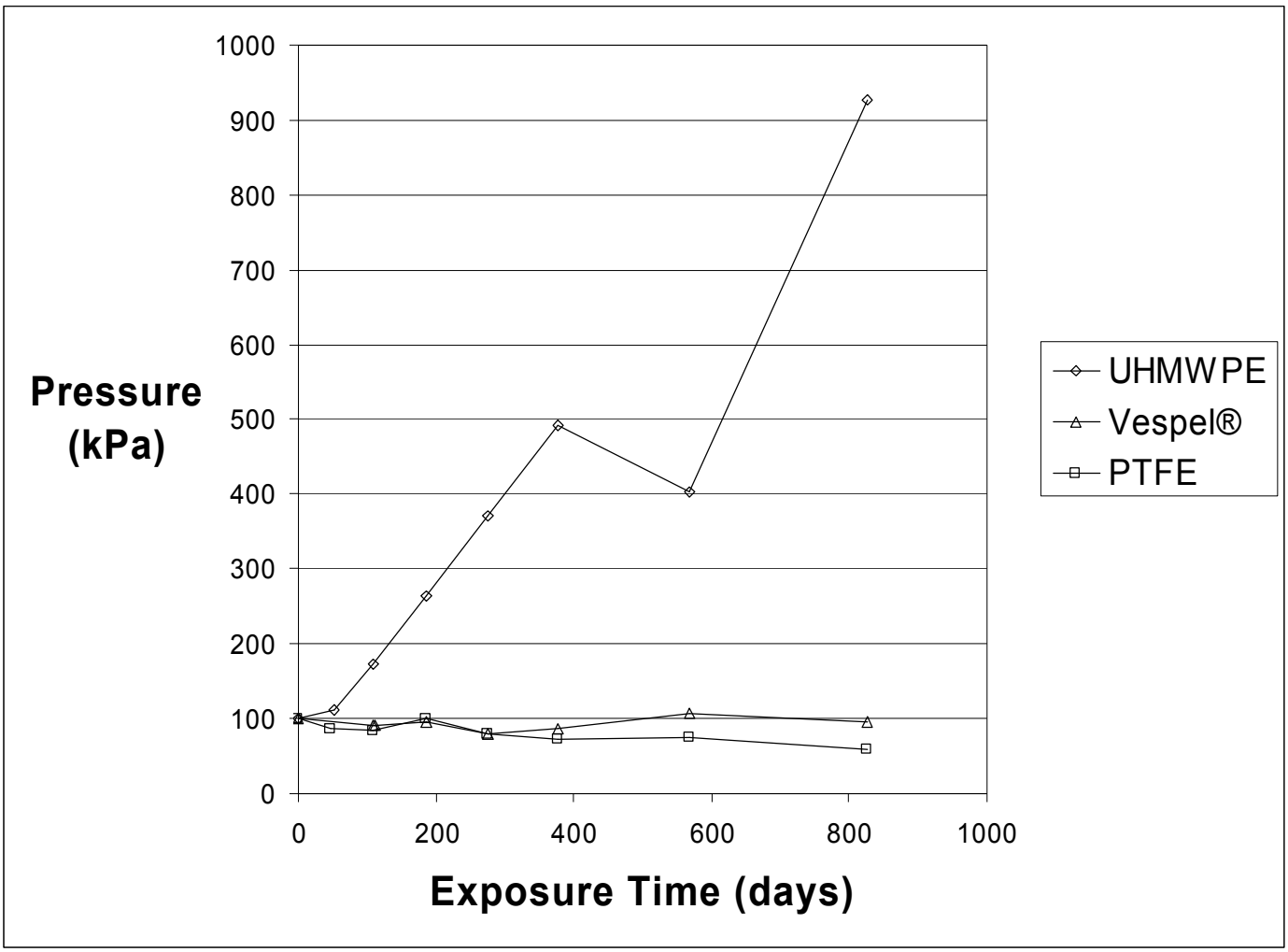

Fig. 2. Total pressure in exposure container at end of exposure, as function of exposure time. UHMW-PE, Vespel®, PTFE as marked.

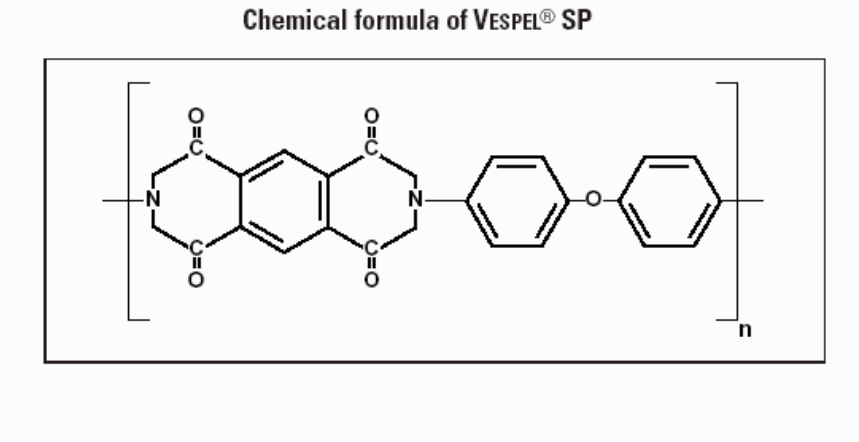

Figure 3. Chemical monomer of Vespel SP.

In Appendix A, a material selection guide written by the SRNL Materials Science and Technology Section is included. One key point therein is the warning to avoid ferritic steels in tritium applications, yet HAPL is currently considering extensive use of these materials. Figure 4 compares the hydrogen permeability of ferritic versus stainless steels. As can be seen, the permeability is much higher in ferritic steels. The SRNL Materials Science and Technology Section has conducted extensive work on various kinds of austenitic steels, and could easily adapt existing facilities to the study of tritium effects on ferritic steels.

An examination of tritium's impact on those materials in HAPL needs to be conducted with the intent of clarifying its impact on the service life various system parts, and therefore HAPL operating costs. Costs would be impacted from the point of view of downtime for maintenance as well as in the impact on loading of the facility built to handle used reactor parts.

One significant technology that may be applicable to resolving these issues is the use of permeation barriers. SRNL has also conducted considerable research in this arena on austenitic steels, developing both instrumentation to study the problem and barrier materials. These methods can easily be expanded to the ferritic steels.

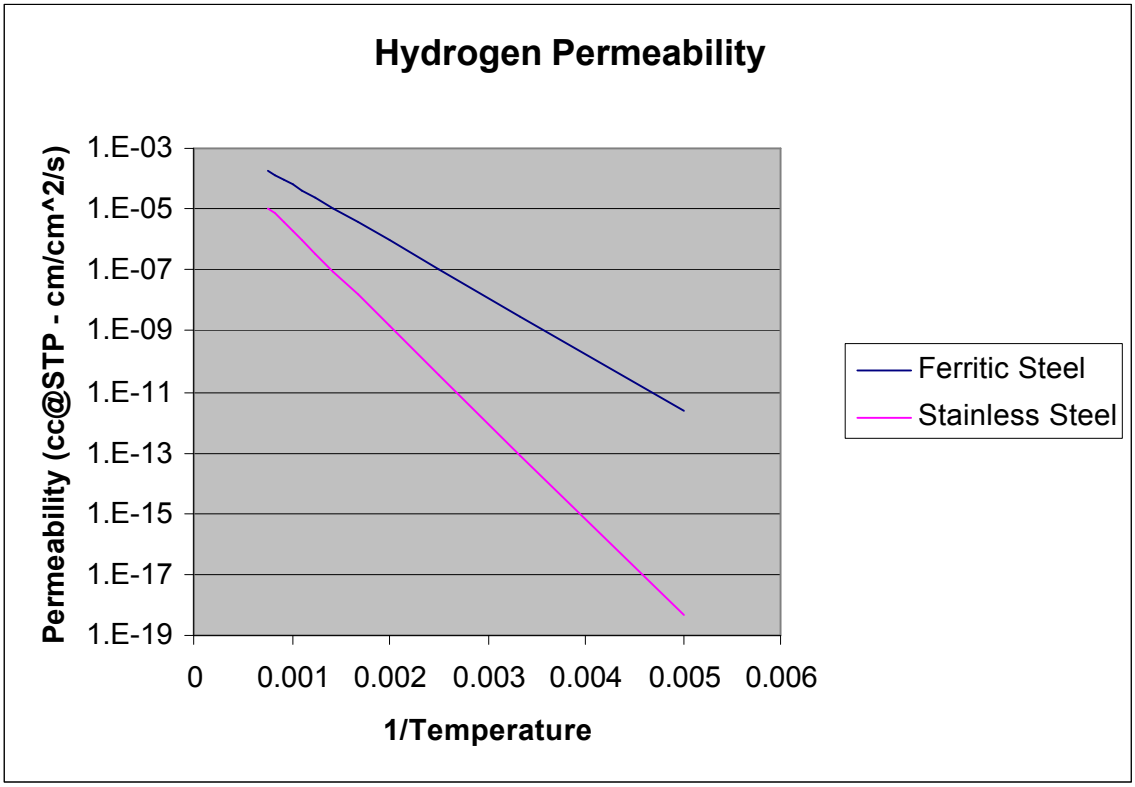

Figure 4. Comparison of Hydrogen Permeability in Steels

Others

Problems will arise during normal operations, and facilities to address these problems need to be available. This requires an analytical facility designed to study potentially highly tritiated materials to help in identifying and solving any such problems. This will require gloveboxes and possibly shielded cells to handle the samples due to the highly radioactive nature of 'fresh' process samples. It is also frequently useful to have a tritium-capable gas handling manifold to use to study process/material problems, which would also be located in this facility.

By careful siting of the HAPL facility, it is possible that existing tritium handling facilities can be easily brought to bear, but in lieu of that, a full analytical facility needs to be built at the HAPL site. This facility should be as carefully designed as any other integral part of the HAPL facility, as it will play a crucial role in meeting duty cycles and minimizing downtimes (especially unexpected ones).

In addition, the proposed analytical facility would also have the duty to characterize radioactive waste for proper disposal and shipping. This would include determining

tritium content of highly tritiated items, such as reactor parts, through sampling, and lightly tritiated materials such as Radioactive Contamination Control waste. Such characterization is normally required for controlled waste disposal per federal regulations.

Although not specifically a tritium issue, it also needs to be determined if the HAPL facility will be required to meet reactor training guidelines, which usually require training simulators. All the proposed processes for tritium handling may have to be included in such simulators. The relevant NRC regulations will need to be determined.

Finally, in a separate whitepaper, SRNL has examined the use of the HAPL facility to produce hydrogen for vehicular fuel usage, and determined it as a feasible addition to the HAPL reactor. The excess reactor heat would be used to drive this production reaction. The hydrogen produced from this process will have to be maintained nearly tritium free if it is to be used in personal and commercial transportation applications. This will necessitate a very close watch on the heat exchange fluids used to drive the production process. If tritium significantly permeates into the produced hydrogen, it would become unusable for its intended application.

Conclusions

Several potential tritium issues have been identified. They clearly lead to the need to develop Conceptual Designs of two to four additional facilities besides the Fuel Recovery System for a fully functional HAPL IFE reactor complex. These new facilities include: a Tritium Loading facility to load the target shells, a Tritium Extraction System designed to recover bred tritium from the breeder blanket material; a Tritium in Solids Recovery System designed to process particulate matter to recovery tritium and possibly to extract tritium from larger parts as well; possibly a Tritiated Waste Handling Facility, designed to handle and dispose both low level and high level tritium contaminated materials from which the remaining T cannot be recovered; and possibly a full analytical/R&D facility to serve as a process support center. A qualified shipping and receiving organization is also likely to be required to handle sample and part movements around the country, especially if tritiated samples are to be shipped off-site. (Note that ‘tritiated samples’ may include large parts as well as small chemical samples.) This group would probably be part of the Waste Handling Facility.

The PPPL Conceptual Design of the Fuel Recovery System is quite well thought out. Nevertheless a variety of small issues have been identified that need to be dealt with, potentially through some bench scale testing, during the anticipated upcoming continuing development of the facility design.

Some questions have also been raised regarding potential H-T exchange in the fuel pellets that could also be addressed through some short-term tritium exchange

experiments. As well, concerns about ferritic steel hydrogen embrittlement have been raised.

HAPL Facility planners have made an excellent start in designing tritium handling facilities for HAPL. Clearly there is a need to continue and expand this effort. SRNL and the SRS Tritium Facility Engineering organization has been involved in three major Tritium Facility startup projects in the last 15 years, all of which have been deemed successful, and we feel we have some significant contributions to make to the design, construction, and startup of a HAPL IFE reactor tritium handling facility. In addition, SRS' Waste Handling divisions, especially those at the DWPF, have expertise in the handling of highly radioactive large parts that can be brought to bear. But clearly, the most efficient way to develop a HAPL Facility will be a team effort that utilizes existing tritium handling expertise from all relevant sources, including PPPL, the National Laboratories, and the SRS Tritium Facilities.

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Appendix A.

Tritium Issues in Materials Selection for Tritium Handling Processes

The following is a discussion of tritium issues in materials prepared by the SRNL Materials Science and Technology Section as guidance in materials selection for tritium handling processes. It is used as such by the SRS Tritium Facilities.

MATERIALS SELECTION FOR TRITIUM SYSTEMS

Overview

This guideline first discusses how tritium systems have been successfully designed, and then presents information about materials that are commonly employed in tritium systems. Austenitic stainless steels, copper, aluminum, welding and other forms of joining, surface treating and coatings, alloys to avoid, and polymers are discussed.

System Design Methodology

Tritium handling systems are gas handling systems that must have an exceptionally low probability of leaking. The consequences of tritium leaks can include personnel uptake, release to the environment, ignition (if mixed with oxygen) and violation of operating permits. Normally, tritium systems are designed and constructed using codes and standards applicable to boiler and pressure vessels, or to nuclear reactors. In the United States, the American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code can be used. Systems constructed using this code have an accepted rigor in design, construction, and inspection that facilitate approval and acceptance of the system by regulators. Other design standards, for example resistance to seismic events, also have to be followed, depending on the location and regulators of the facility.

Proper materials selection and rigorous design have led to tritium handling systems that are extremely safe for long periods of time. Materials exposed to tritium under certain conditions can be susceptible to hydrogen embrittlement. The chances of embrittlement are significantly reduced by proper material selection. No additional thickness of components, such as the “corrosion allowance” used to mitigate uniform corrosion, is added to components to reduce or eliminate the chance of hydrogen induced cracking and subsequent failure. In the past, tritium systems were located in glovebox hood systems having a single pass air flow, to protect workers should a component fail and tritium be released; the released tritium would be removed from the facility into the atmosphere by the hood exhaust system. Current system design normally consists of a closed glovebox, with nominally pure inert gas such as nitrogen or argon, and a stripper or getter system to trap tritium that permeates or is released into the glovebox. This arrangement protects workers and minimizes releases to the environment.

Tritium can permeate vessel barriers, especially in components operating at elevated temperature. Currently available tritium permeation data is normally sufficient to estimate the order of magnitude of tritium permeation through barriers. These estimates can be used during system design or to determine whether additional purging, confinement and stripping systems are required to "clean up" permeated tritium or whether other design changes (such as wall thickness, material, or coating) are required to reduce permeation. Tritium permeation can not only lead to contamination outside the barrier, it also results significant quantities of tritium dissolved in parts, which can lead to hydrogen embrittlement. Over time, this tritium decays to helium-3, which has been found to accentuate hydrogen embrittlement and to cause weld cracking (see below).

After fabrication, thoroughly leak testing tritium systems is extremely important. Normally, leak testing employs commercial helium mass spectrometer based systems, and is performed after any other required proof, pressure, or vacuum performance tests. Other leak detection methods, such as rate-of-rise, can also be employed in addition to helium mass spectrometry. A dilute solution of tritium in an inert gas can also be used to detect small leaks. Tritium is a highly effective leak detection species, since it travels rapidly through cracks and can be easily detected at very low levels.

Structural Metals

Recommended Materials

Austenitic Stainless Steels

The recommended materials of construction for tritium handling systems are from the class of wrought AISI 3XX series of austenitic (face-centered-cubic) stainless steels, including Types 304L, 316L, and 347. Types 304L and 316L are most often used in tritium processing systems in the United States. These steels provide good strength, weldability, and resistance to hydrogen embrittlement. Components fabricated from these materials are routinely procured. Many commercially available vacuum system components that are used in tritium systems, such as valves, piping, pumps, and analytical instrument sensors, are fabricated from these types of austenitic stainless steel. Wrought materials are preferred to cast, because wrought materials normally have a much more homogenous microstructure. Tritium has leaked through parts having poorly oriented stringers and inclusions in the past. The forging direction of some wrought components has been specified, so that the orientation of any inclusions is not in a direction that could result in a tritium leak path. Low carbon grades (such as 304L and

316L) are preferred, to avoid weld sensitization and to reduce the number of inclusions (impurity particles such as oxides, carbides, etc.). Modern vacuum-arc-remelted steels are a good choice, because they have lower impurity levels, resulting in fewer inclusions that could aid hydrogen induced cracking or provide leak paths. Typically, tritium system components employ seamless pipe and tube where practical.

Stabilized grades, such as Type 347, have been employed in applications where post-weld heat treatment is not possible. This usually occurs when a process vessel contains a working material (such as hydride or getter) that will degrade when exposed to the post-weld heat treatment, which is typically performed at about 1100° C for austenitic stainless steels.

High carbon grades, such as Type 347H or Type 316H (Type 316 H has 0.04% carbon minimum), have been successfully employed for tritium confinement if high temperature strength is required. Type 310 stainless steel has good oxidation resistance and can be considered for elevated temperature applications if oxidation is a concern. Type 316 stainless has superior creep resistance but inferior oxidation resistance to Type 310.

Some tritium storage and isotope separation process beds are exposed to continual temperature cycling. Type 316L may be preferred to Type 304L for the container vessels of these beds, because Type 316L is more stable with respect to forming stress induced martensite than Type 304L. Also, if process beds are exposed to sub-ambient temperatures, selecting Type 316L is important to minimize the chance of forming thermally induced martensite at low temperature. This would reduce the already small chance of forming small amounts of undesirable martensite phase, which is significantly more susceptible to hydrogen embrittlement than austenite.

Some types of higher strength austenitic stainless steels not generally employed for tritium containment may be required for fasteners (such as nuts and bolts) in mechanical joints in high-temperature regions. This may be acceptable if the bolts are exposed to only residual amounts of tritium. These materials also may be used to contact 3XX stainless steels to avoid galling of mating screw contact surfaces. Typical materials used in these applications include Nitronic 60, Nitronic 50 (also called 21-13-9), and Nitronic 40 (also called 21-6-9); these are all nitrogen strengthened austenitic stainless steels.

Copper and Copper Alloys

In principle, copper should be a suitable material in tritium systems. Copper has several tritium compatible properties: tritium has a low permeability in copper, and copper is a ductile, stable, face-centered-cubic metal and so is resistant to hydrogen embrittlement. The high thermal conductivity of copper is a desirable property for process vessels requiring heat flow or constant temperature. Copper can be easily joined in a number of ways such as soldering, brazing, or welding. In spite of these advantageous properties, copper and copper alloys are not commonly used for confinement in tritium systems. Several factors may account for this. The ASME allowable design strength of copper falls rapidly at temperatures above 200 deg. C, making it difficult to use copper for process beds that operate at elevated temperature. Also, hydrogen isotopes can react at elevated temperature with oxygen, a common impurity in copper, whether the oxygen exists in solid solution or as copper oxide precipitates. In either case, water is formed when hydrogen isotopes interact with oxygen, and over time water vapor agglomerates at grain boundaries, which eventually results in intergranular cavitation, cracking, and failure. This failure mechanism is sometimes termed “steam embrittlement”. Also, a transition junction (normally nickel) is required to join copper and the stainless steel components of the remainder of the system.

Aluminum and aluminum Alloys

Aluminum has properties making it potentially desirable for tritium systems. It has a low density and a high thermal conductivity. Hydrogen isotope permeability is very low in aluminum, compared to stainless steel. Aluminum is used in applications where light weight is important, such as in containers that must be lifted by personnel in gloveboxes. However, aluminum is not commonly used in tritium systems. Stainless steel has much higher strength, at room and elevated temperature. Welding aluminum requires more precautions, because aluminum reacts with atmospheric water vapor, which can cause porosity due to hydrogen in the weld fusion zone.

Joining

Components of tritium systems are commonly joined by welding. Welds are normally designed so they can be non-destructively inspected by a suitable method such

as radiography or ultrasonic testing (UT). Also, the weld design should minimize so-called "virtual leaks" on the interior of tritium containing volumes. Examples of weld practice to minimize virtual leaks include i) using full penetration welds where possible, and ii) welding feed-throughs to the interior wall surface (not just on the outside, which would leave a gap on the inside around the feed-through that is difficult to outgas). Standard weld rod filler materials are chosen, depending on the base alloy. Every effort should be made to reduce or eliminate welding residual stresses.

There is less experience using other joining methods such as brazing or high temperature soldering in tritium systems. Dissimilar materials may have to be joined by a transition junction, using an intermediate material to enable proper welding and accommodate differences in thermal expansion and other properties. Palladium tubes in hydrogen isotope purifying systems are typically connected to stainless steel by a braze joint.

Tritium gas permeates austenitic stainless steels, and over time helium-3 is created by beta decay of tritium in solution in the material. Welding stainless steel containing solute helium is extremely difficult because intergranular cracking can occur. During welding, the solute helium agglomerates at grain boundaries and forms both intergranular cracks in the heat affected zone and pores in the fusion zone. Low-heat-input weld techniques have been shown to mitigate this problem to some degree, however weld repair of helium-containing stainless steel is normally difficult to perform without some cracking.

All-metal mechanical joints are a sound way to join components in tritium systems. Typically copper, silver-plated nickel, or silver-plated stainless steel have been used as gaskets. Commercially available high- and ultrahigh- vacuum fittings are normally compatible with tritium.

Surface Coatings and Treatments

Aluminide coatings have been successfully employed on stainless steel to reduce permeation into and through the steel. These coatings can be applied on large items using a proprietary fluidized bed furnace, having a controlled atmosphere (in the so-called "calorization" process). Gold has also been used as a permeation barrier in some applications, and is often applied over a thin nickel buffer layer ("strike") which has been applied to the bulk metal (e.g. stainless steel), after proper surface preparation.

Several companies treat stainless steel surfaces using various proprietary electrochemical processes to "passivate" the surface. These processes probably enrich the chrome content of the surface oxide, polish the surface (thereby reducing the effective surface area), and remove carbon and hydrogen from near the surface. All of these microstructural changes may be desirable for tritium systems in which the process gas must remain at high purity. The inlet systems of mass spectrometers are commonly passivated, to reduce changes of gas composition by isotope exchange while the gas flows from the sample location to the mass spectrometer. Passivated surfaces reduce the rate of isotope exchange in hydrogen isotope mixtures, probably by reducing the catalytic effect of the surface decomposing hydrogen isotope molecules to atoms, which enables isotope exchange. Vacuum systems having surfaces treated in this way evacuate faster. This type of surface passivation can be expensive, so many parts of tritium systems are not passivated; normally only parts requiring the special properties of passivated surfaces are treated.

Metallic Materials to Avoid

Plain carbon steels and alloy steels must not be used for tritium confinement. These steels have high strength and (normally) a body-centered-cubic crystal structure, both of which make the material less ductile and much more susceptible to hydrogen embrittlement than austenitic stainless steels. Ferritic stainless steels (such as Type 430), martensitic stainless steels (both quench-and-tempered (such as Type 410) and precipitation hardening (such as 17-4 PH and PH 13-8 MO)), and precipitation hardened austenitic stainless steels (such as AM-350) should not be used for general tritium containment; they are all more susceptible to hydrogen embrittlement than the austenitic stainless steels. Additionally, free-machining grades of austenitic stainless steel (such as Type 303) should not be used- these materials contain large concentration of sulfur, which can result in H_2S gas in the process stream.

Other materials that must not be used for tritium confinement are any materials that form hydrides. Examples include titanium, zirconium, tantalum, niobium, and many alloys of these materials.

Polymers

General

All polymers degrade when exposed to radiation. Both tritium and tritiated water permeate all polymers, and permeated tritium deposits the beta decay energy throughout the polymer bulk. Although the tritium beta energy is very low and has a small penetration depth in matter, permeation allows tritium atoms to be near enough to polymer chains throughout the bulk to cause radiation damage throughout the polymer. Types of radiation induced changes in polymer properties include either softening (degradation) or hardening, ductility loss, color change, dimensional change, and gas evolution (radiolysis). Because of these effects, polymers should only be used in tritium systems where no metal alternatives exist. Normally, only non-halogenated polymers are used, because products of degradation can be species such as HF and HCl that can both pollute the tritium system and form corrosive hydrofluoric and hydrochloric acids. Polymer parts must be easily replaceable as a part of normal operations, and a program of regular testing, inspection, and replacement must be established. The system should be designed to expose any polymers to as little tritium as possible. Typical uses of polymers in tritium systems include gaskets, O-rings, electrical cable insulation, and valve parts, including seats, stem tips, and packing.

Many deleterious effects of radiation on polymers are aggravated by dissolved oxygen. Protecting polymers from oxygen or air will likely lengthen the lifetime of polymers exposed to tritium. Also, temperatures above about 120 degrees C accelerate radiation effects in polymers, so the temperature of any polymer parts should be kept as low as possible. Inert additives such as glass or graphite generally enhance the resistance of polymers to radiation. Addition of antioxidants or “anti-rads” may also enhance radiation resistance.

Plastics in Tritium Service

Vespel™, a polyimide, has been successfully used for valve stem tips in some tritium laboratories. It is quite resistant to radiation damage compared with other polymers, however it’s high stiffness makes it problematic to install in sealing applications- several new tips may fail leak tests until one fits. Ultra-High-Molecular-Weight polyethylene (UHMWPE) has been used for valve stem tips in larger automatic valves. It becomes brittle, dark, and shrinks with time when exposed to tritium gas, but is considered to be

an acceptable material selection, as is High Density Polyethylene (HDPE). Low-Density polyethylene (LDPE) is very permeable by tritium and tritiated water, and should not be considered for use in tritium systems.

Polytetrafluoroethylene (PTFE, a trade name is Teflon™) degrades and decomposes in tritium, resulting in HF. If water or water vapor is present, highly corrosive hydrofluoric acid forms from HF. Chlorofluorocarbon polymers in general should not be used in tritium systems, except where no alternatives exist (such as the packing of some valves, partly made of glass-filled Teflon™). Polyvinyl chloride (PVC) and vinylidene chloride (Saran) are among several polymers used in tritium protective clothing, but should not be used for process equipment because they contain chlorine.

Elastomers in Tritium Service

Ethylene propylene diene monomer (EPDM) elastomer has been employed for low pressure process flange gaskets and other applications, because of its relatively good performance in tritium service. In some cases, Buna-N (NBR) process flange gaskets are being replaced by EPDM when the gaskets are changed, however in other applications Buna-N remains in use. Butyl rubber has low permeation for both tritium and tritiated water, but is not as resistant to radiation damage as EPDM. Butyl rubber is used for glovebox gloves. Water vapor in the air outside gloveboxes permeates glovebox gloves, and can lead to a significant portion of the residual tritiated water vapor in tritium confinement gloveboxes.

Kel-F™ is a common chloroflurocarbon polymer, and is incompatible with tritium. It (like Teflon™, above) degrades in tritium gas and should not be used. Viton™, another common fluorine containing O-ring material, is used occasionally, but can embrittle in months in tritium service and should be avoided if possible.

Summary

The most successful class of materials used for tritium systems is one of the AISI Type 3XX series of austenitic stainless steels, especially Types 304L and 316L. Copper and aluminum (and their alloys) can be used for specific purposes in tritium systems. Materials in these systems are most commonly joined by welding, and modern all-metal vacuum joints are also common. Surface treatments and coatings can be used to reduce permeation, hinder isotope exchange, increase purity, and enhance the outgassing and pump down speeds in systems. Ferritic (carbon and alloy) steels and non-austenitic

(ferritic, martensitic, and precipitation hardened) stainless steels must not be used for confining tritium, because they are susceptible to hydrogen embrittlement. Polymers are susceptible to radiation damage when exposed to tritium, and should be used in tritium systems only if no other reasonable alternatives exist.