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## THE DEVELOPMENT OF TRITIUM TECHNOLOGY FOR THE UNITED STATES MAGNETIC FUSION ENERGY PROGRAM

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### Summary

Tritium technology development for the DOE fusion program is taking place principally at three laboratories, Mound Facility, Argonne National Laboratory and the Los Alamos Scientific Laboratory. This paper will review the major aspects of each of the three programs and look at aspects of the tritium technology being developed at other laboratories within the United States. Facilities and experiments to be discussed include the Tritium Effluent Control Laboratory and the Tritium Storage and Delivery System for the Tokamak Fusion Test Reactor at Mound Facility; the Lithium Processing Test Loop and the solid breeder blanket studies at Argonne; and the Tritium Systems Test Assembly at Los Alamos.

### Introduction

Tritium technology development for the DOE fusion program is taking place principally at three laboratories, Mound Facility, Argonne National Laboratory, and Los Alamos National Scientific Laboratory. This paper will review the major aspects of each of the three programs and look at aspects of the tritium technology being developed at other Laboratories within the United States.

### Current Activities in Tritium Technology

#### Mound Facility (MF)

Mound Facility has been actively involved in tritium technology for over 20 years. Recently Mound has focused most of its tritium technology development on tritium containment and environmental control. The two components of Mound's tritium technology development which are currently most active and most relevant to fusion needs are the Tritium Effluent Control Laboratory (TECL) and the Tritium Storage and Delivery System (TSDS) project for the TFTR.

The TECL was initiated in 1971 for development and demonstration of tritium containment. The goal of TECL is to prevent any tritium release to the environment and to recover for reuse all tritium released within the laboratory. TECL consists of an integrated set of containment

systems and detritiation experiments. Tritium containment is provided by gloveboxes, a glovebox atmosphere detritiation system (GADS), a laboratory which can be isolated from the remainder of the building, and an emergency containment system (ECS). The GADS is a 0.05 m<sup>3</sup>/s helium purifier which continuously cleans the glovebox atmosphere. The ECS is a 0.5 m<sup>3</sup>/s catalytic oxidation, water vapor adsorption system for room air detritiation. In addition, a 7.5 x 10<sup>-3</sup> m<sup>3</sup>/s utility air detritiation system of the oxidation-adsorption type is available for cleaning passbox atmospheres and for non-standard applications.<sup>1</sup>

Four tests have been performed to measure the efficiency of the GADS and ECS in cleaning up after a tritium release in a glovebox or a room. Glovebox cleanup was as predicted; however, ECS tests showed substantial tritium loss through imperfect exhaust duct seals. More tests are planned to permit study of surface absorption and conversion to HTO. This was the first test of such a large ECS system within the United States.

Tritiated water vapor collected by the ECS or by other, similar systems can be detritiated by the combined electrolysis catalytic exchange (CECE) pilot-scale unit which is part of TECL. The CECE incorporates a countercurrent flow of water and hydrogen gas in two 2.5-cm diameter, 7.5-m long columns packed with a hydrophobic, precious metal catalyst which was developed by Atomic Energy of Canada, Limited. Bottom reflux is provided by a solid polymer electrolysis unit, whereas top reflux is provided by a catalytic recombiner. Tritiated water containing 300 Ci/liter has been stripped to 10<sup>-3</sup> Ci/liter in the CECE at feed rates of approximately 4 ml/min of water. Although tests of CECE have not been run long enough to accumulate an equilibrium concentration, values in the range of 1-10 mCi/liter are expected.<sup>2</sup> The goal of the CECE work is to develop a full scale detritiation plant which will be suitable for processing tritiated aqueous waste from fusion or fission reactors or from fuel reprocessing plants.

Part of the hydrogen generated in the electrolysis unit can be withdrawn to be used as

feed for the remaining TECL component, a cryogenic distillation system. This system includes a single 0.6-cm diameter x 50-cm long packed column, operating at approximately 25 K. Bottom and top concentrations of 2500 Ci/m<sup>3</sup> and 10<sup>-3</sup> Ci/m<sup>3</sup>, respectively, were measured during one run with this column, for an enrichment factor of 2.5 x 10<sup>6</sup>. Feed rates of 100 standard cubic centimeter per minute are possible at this enrichment factor. This work is supported with computer simulation studies of the distillation process. In addition, an experimental study of the rate of hydrogen isotope equilibration (e.g. 2DT  $\rightleftharpoons$  T<sub>2</sub> + D<sub>2</sub>) at cryogenic temperatures is being performed. This is of interest both in cryogenic distillation and in cryogenic fuel pellet production.

The Tritium Storage and Delivery System (TSDS) has been designed and constructed for use in the Tokamak Fusion Test Reactor (TFTR) at Princeton Plasma Physics Laboratory. The TSDS will receive, assay, store, and deliver measured quantities of high purity tritium to fuel the TFTR. The TSDS consists of a receiving manifold, uranium tritide storage beds, transfer pumps, and metering volumes, all contained in two stainless steel gloveboxes. In addition, a quadrupole mass analyzer and pressure-volume-temperature measuring equipment permit assay of the tritium as received as well as confirmation that the tritium delivered to the reactor has the required purity. Extensive development was done with the mass analyzer to permit analysis of the required sensitivity and accuracy.<sup>3</sup>

In operation, tritium received at TFTR is introduced to the TSDS through the receiving manifold; and after being assayed, is pumped into a uranium bed where it is stored as uranium tritide for future use. During D-T operation of TFTR, an appropriate tritium storage bed is heated to raise the internal tritium pressure to about 1/2 to 1 atmosphere. Each time the reactor is to be fueled (as often as every 5 minutes), the required amount of tritium is pumped into a metering volume. The gas then passes through a delivery manifold to three calibrated injection volumes near the torus. Finally, injection to the torus is controlled by a specially designed piezo-electric valve at each injection volume. After a burn, the fuel is not recycled, but is recovered by the TFTR vacuum system and stored for later reclamation.

The TSDS is designed to achieve the highest possible levels of reliability, safety, and tritium containment. The two transfer pumps are interchangeable, two-stage, doubly contained, metal bellows pumps, one of which is a spare. The three, interchangeable storage beds are each doubly contained in stainless steel, with a provision for purging the secondary volume of tritium which permeates from the primary container.

Each bed has a rupture disc sealed connection to an evacuated "dump" tank. At any given time, one storage bed is active; one is available for cleaning operations, and one is a spare.

Complete instrumentation is provided for TSDS to permit accurate control and to promote safety. The tritium generators are provided with pressure sensors and redundant thermal control and over-temperature protection.

Normal operations of the TSDS are performed remotely through computer control of pneumatically operated valves. All process valves are bellows sealed with polyimide used in place of metal. These valves have been individually remachined to ensure reliable leak-free operation.

The TSDS will be tested with a load of 10<sup>4</sup> Curies of tritium to ensure reliable operation of the entire system before delivery to Princeton. A dummy manifold and injection chamber will simulate the manifold and torus at Princeton.

#### Argonne National Laboratory (ANL)

The program on fusion reactor research at the Argonne National Laboratory includes a number of applied research topics covering a variety of interrelated areas of fusion tritium technology. The principal focus of this work is on studies of fuel handling, breeder blanket processing and tritium containment. These studies are both experimental and analytical in character and have, in recent years, spearheaded technological advances in a number of important fusion-specific areas which are described below.

Modest advances have been made in recent years in the study of liquid lithium processing. A 200-liter-capacity system, the Lithium Processing Test Loop<sup>4</sup> (LPTL), has been operated for over 9500 h. Cold trapping, reactive-metal getter trapping, and a method based on molten-salt extraction have been tested<sup>4</sup> using the LPTL and related facilities. The ranges of the projected lower-limit impurity control levels for the elements H (D,T), O, N, and C, based on these tests, are shown in Fig. 1. Molten salt extraction offers the best potential for developing a regenerable process capable of recovering tritium (from D-T reactor blankets) and controlling impurities at the 10 appm level. The salt extraction method also appears to be applicable to the processing of liquid lithium-lead alloys.

In the area of instrumentation and hardware for liquid lithium systems, permeation- and resistivity-type meters have been developed to a point where they can be meaningfully employed for monitoring of hydrogen<sup>5</sup> and hydrogen plus nitrogen,<sup>5</sup> respectively. The principal difficulties in operating present day stainless steel-constructed lithium systems have resulted from (1) cracking of special components containing

cold-worked material with high residual stress (EM-pump channels and valve bellows),<sup>4</sup> and (2) mass transfer buildup<sup>4,6</sup> involving iron, chromium, and nickel. A method developed by DeVan et al.,<sup>6</sup> wherein aluminum is added to lithium so that it can react with and stabilize the surfaces of stainless steel components, has shown promise as a means of retarding mass transfer and intergranular penetration in stainless steel systems.

The use of solid lithium compounds as the breeding material for D-T fusion reactors has come under increasingly greater study in the USA in recent years<sup>7</sup>. Three different approaches have been suggested: (1) in-situ tritium recovery, (2) removal and external processing of fuel-pin-type breeder assemblies on a periodic basis (every six to twelve months), and (3) continuous circulation of solid material into and out of the reactor with tritium processing done externally. There is evidence to suggest that the in-situ recovery of tritium is feasible, but a carefully controlled breeder material temperature distribution is required<sup>7</sup> so that (1) tritium does not build up to excessive levels in low-temperature regions and (2) sintering does not occur in high-temperature regions.

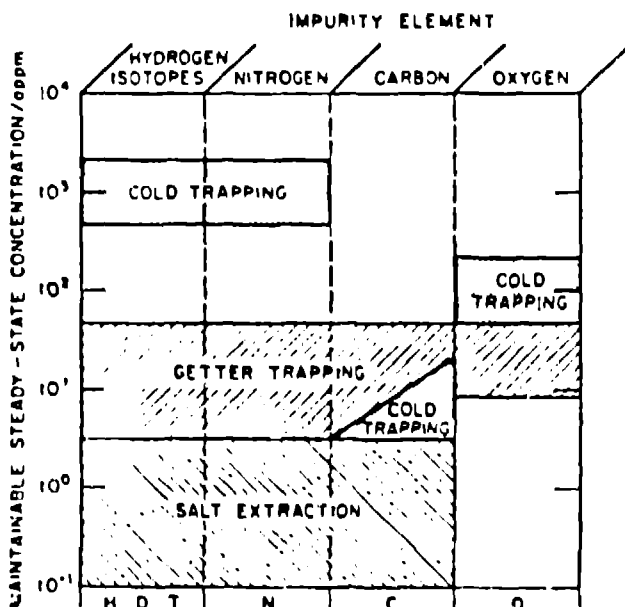


Fig. 1. Summary of projection of achievable lower-limit impurity control levels for selected lithium processing methods.

The fuel-pin approach to tritium breeding is saddled with the problem of large in-blanket tritium inventories (~50 kg per fusion GW for annual pin removal), and the circulating solid approach presents formidable engineering complexities. However, if the in-situ method of

handling solid breeder blankets turns out to be intractable and if liquid metals are eliminated from consideration because of engineering or safety considerations, then the fuel-pin and circulating-solid approaches may prove to be the only remaining recourse for self-sustaining D-T fusion reactors.

In work related to the STARFIRE Commercial Fusion Reactor Study<sup>8</sup> currently ongoing in the USA, a top-down selectivity analysis has been made<sup>9</sup> to identify the most tractable breeder/coolant/structure (B/C/S) combinations for D-T fusion reactors. In a typical analysis, a breeder material (e.g., liquid lithium, liquid lead-lithium alloy, solid Li<sub>7</sub>Pt<sub>2</sub>, Li<sub>2</sub>O or another lithium-containing ceramic) is matched with various coolants (e.g., water, helium, liquid metals, molten salts) and compatibility assessments are performed. The criteria for these assessments are breeding capability, exothermicity of breeder/coolant interaction, operating temperature requirements, and thermodynamic efficiency of the associated power cycle. Breeder/coolant combinations that survive the scrutiny of this first assessment are then matched with likely classes of structural materials (e.g., austenitic-, nickel-, ferritic-, and refractory-base alloys) and a second assessment is made. Here the criteria are based mainly on structural material compatibility (with breeder and coolant) and temperature constraints. In summary of this exercise it can be said that no B/C/S combination from among the choices listed parenthetically above is developed to a point where one could predict with confidence that satisfactory performance in the fusion environment could be achieved. Liquid lithium/refractory metal (Nb- or V-base alloys) and ceramic breeder/water (pressurized or boiling)/austenitic or ferritic combinations appear to be among the least objectionable choices from an engineering and compatibility point of view.

#### Los Alamos Scientific Laboratory (LASL)

In January 1977 the Los Alamos National Scientific Laboratory (LASL) undertook the design of a national Tritium Systems Test Assembly (TSTA)<sup>10,11</sup> with the purpose of developing, interfacing, and demonstrating all the technologies related to the deuterium-tritium (DT) fuel cycle for fusion reactors. The first machine requiring extensive use of these technologies will be either the Engineering Test Facility (ETF) or the International Tokamak Reactor (INTOR). The TSTA will be operating as a full-scale tritium-processing plant in late 1981, years before any DT-burning power reactor is operational. Data on efficiency and reliability of processes and components will thus be available to fusion engineers in time for design of ETF, INTOR, and more advanced reactors.

TSTA objectives are balanced between developing technically feasible processes and

securing public acceptance of the necessary technology. The purely technical objectives include development and continuous demonstration of all aspects of a closed loop DT fuel cycle, Fig. 2, from plasma chamber evacuation through fuel purification and isotopic enrichment to plasma fuel injection. The equally important environmental and public safety objectives are the development and long-term demonstration of environmental and personnel protective systems with no significant releases or personnel exposures, and minimized off-site radioactive waste handling. At TSTA, these objectives will be met with a DT inventory of 150-200 g, processed continuously at the rate of 1.5-2 kg/day. This processing rate equals or exceeds the requirements for ETF or INTOR.

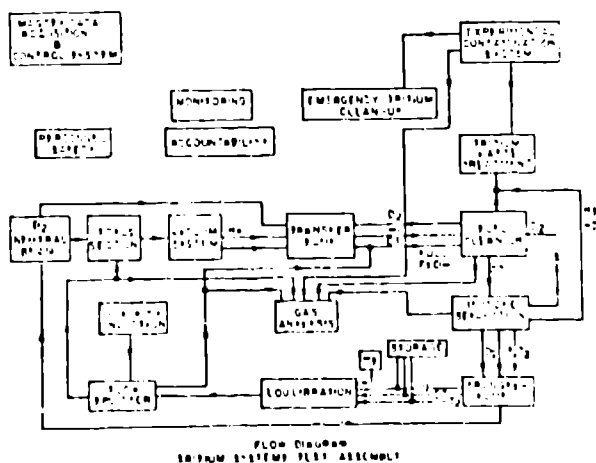


Fig. 2. The TSTA Process loop showing subsystem interactions.

Design work is complete, major components have been purchased, and installation is proceeding on schedule for the following major environmental and DT-processing systems: an emergency tritium cleanup system capable of confining any major release of tritium within the TSTA building; a continuously operating tritium waste treatment system that removes residual tritium from all process waste streams; a plasma-chamber evacuation system based on a compound condensation/sorption cryopump; a fuel cleanup system that uses a combination of catalytic reactors, hot metal getters, and cryogenic sorption/freezout to reduce nonhydrogen impurity levels below 1 ppm; a hydrogen isotopic separation system based on cryogenic distillation; a number of transfer pump assemblies for safe, contaminant-free transfer of DT about the process loop; a computer-based master data acquisition and control system, which monitors and controls all TSTA processes.

Preliminary experimental work and computer simulations were instituted to aid in mechanical and conceptual design work for TSTA, with the

following favorable results to date: a compound cryopump is capable of simultaneously exhausting helium and DT from a plasma chamber; the helium ash can be separated from reusable DT fuel in the compound cryopump during cryopanel regeneration; the design of cryogenic distillation columns can be optimized for any desired total flow, number and purity of product streams, etc., and still provide great operational flexibility and control stability; data on cryogenic sorption of hydrocarbons and  $\text{NH}_3$  have been collected, enabling us to design a fuel cleanup system of adequate performance using a combination of known technologies; new tritium-compatible mechanical vacuum and transfer pumps, useful in several subatmospheric regimes, have been developed by cooperative efforts between the TSTA project and industry.

#### Lawrence Livermore National Laboratory (LLNL)

There are several areas of tritium technology of interest to the fusion energy community being pursued at the Lawrence Livermore Laboratory. One area of concern is the recovery of tritium following an environmental or atmospheric release of tritium. In current designs of systems for recovery of such released tritium the method of recovery is catalytic oxidation with atmospheric oxygen forming tritiated water which is then collected. Sherwood<sup>12</sup> has measured room temperature kinetic data on tritium/air oxidation with three common catalyst/substrate formulations. These were platinum/alumina, palladium/kaolin, and palladium/zirconite. Each of the dispersed-metal catalyst is extremely effective in promoting tritium oxidation in comparison with self-catalyzed atmospheric conversion; equivalent first-order rate constants are higher by roughly nine orders of magnitude. Electron-microprobe scans reveal that the dispersed metal is deposited near the outer surface of the catalyst, with metal concentration decreasing exponentially from the pellet surface. The platinum-based catalyst is more effective than the palladium catalyst on a surface-area basis by about a factor of three.

Miller<sup>13</sup>, et al at LLNL are working on the development of an organic getter which will operate in the presence of air and minimize the formation of the more hazardous tritiated water. The compound of interest, 1,4-diphenylbutadiyne, is a hydrocarbon,  $\text{C}_6\text{H}_5-\text{C} \equiv \text{C}-\text{C} \equiv \text{C}-\text{C}_6\text{H}_5$ . Here the  $\text{T}_2$  adds to the acetylene (triple) bond of the getter in the presence of a metal catalyst. The necessary catalyst will, however, stimulate the  $\text{T}_2 + \text{O}_2$  reaction, so some tritiated water will be formed. Early results indicate that these getters will indeed remove tritium from air, although with the formation of some water. The organic triple bond appears to be a very good candidate for this type getter. Miller states that the best solution would be to bring the metallic catalyst and the triple bond together in a single, fairly simple molecule,

for instance  $(\text{PhC}\equiv\text{CPh})_2\text{Pt}$  where Ph is a phenyl group. To date very little work has been done on methods of disposing of or storing these tritiated organic getters. This work will be done once an optimum getter material is chosen and the gettering process thoroughly understood.

The Rotating Target Neutron Source (RTNS-II) at LLNL<sup>14</sup> is an accelerator-based neutron source used for studying radiation damage to materials. Here energetic deuterons bombard a solid metal tritide target, producing fusion neutrons (over  $10^{13}$  14-MeV neutrons per second). Deuterium continually displaces tritium from the target at rates which go as high as seven curies per hour. The anticipated addition of a second accelerator and increasing the neutron yield of the present accelerator could increase tritium output to twenty to thirty curies per hour. This tritium is released into the accelerator vacuum system. Since it is not acceptable to release this tritium to the environment, a tritium scrubbing system was devised<sup>15</sup> to clean the vacuum system exhaust before it is vented to the atmosphere. This system consists of a catalytic recombiner where tritiated water is formed and molecular sieve drying towers for collection of the water thus formed. When these molecular sieve driers become saturated they are replaced and the saturated beds are buried. The driers contain approximately seven pounds of molecular sieve which is loaded to 14-15% of the dry weight. At current operating levels, water loads of about 0.25 pounds per week are collected. Most of this is  $\text{D}_2\text{O}$  with only about 1% being tritium. Schumacher reports a  $10^5$ - $10^6$  tritium concentration reduction factor through the scrubber system with most of the escaping tritium being in the gaseous form. Small scrubber systems such as this one in place at RTNS-II may find extensive use in the fusion program.

Souers<sup>16</sup> at LLNL is measuring and correlating cryogenic data on  $\text{D}_2$ ,  $\text{T}_2$  and mixtures of these components. Since solid deuterium-tritium (D-T) may be used as a future fusion fuel the measurement of physical and chemical properties of cryogenic D-T in the solid, liquid and gas phases will be an aid to the design engineer and the plasma physicist considering the use of cryogenic D-T.

Some of the properties they are measuring include the D-T reaction rate, that is the rate  $\text{D}_2 + \text{T}_2$  react to form the three component mixture  $\text{D}_2$ -DT- $\text{T}_2$ . At room temperature, this reaction takes place with an exponential 1/e-time on the order of tens of minutes. At 20 K the 1/e-times are on the orders of tens of hours. Other properties being measured by Souers include the thermal conductivity of the solid mixtures and the electrical conductivity of both liquid and gaseous species in the 20-26 K temperature

range. Souers has compiled a very important report<sup>17</sup> in which he has correlated the measured physical and chemical properties of the hydrogens below 30 K. This compilation will benefit fusion engineers who do not have a strong background in cryogenic materials as well as those specialists who will be concerned with the details of the low temperature deuterium-tritium mixtures.

#### Oak Ridge National Laboratory (ORNL)

Bell, et al<sup>18</sup> at ORNL have measured the tritium permeability of structural materials and surface effects on permeation rates. Tritium management in any system will always include containment such that tritium release rates will be less than established limits and that will be as low as practical. The ability of hydrogen to permeate most materials make complete containment of tritium an extremely difficult task. However, tritium release rates from a given system can be minimized by two primary efforts. First is the selection of a compatible containment material which frequently will also be the structural material. This effort would include films and barriers that would be compatible with the structural material. Second and sometimes more important, the material should have surface chemistry in the particular system that continuously impedes tritium permeation. Bell has shown that by exposing the austenitic Incoloy 800 and the ferritic SS 404 to steam oxidation at 0.94 atm and 930 K the permeabilities of these materials can be reduced by several orders of magnitude over the permeability of the clean metal, Figure 3. These studies demonstrate that in situ surface oxidation of construction alloys can produce oxide barriers that reduce tritium permeation by significant factors. However, all such results have limitations when applied by extrapolation to operating systems with high temperatures and corrosive conditions. Therefore, Bell emphasizes<sup>18</sup>, one must consider current conclusions of permeation barrier effects only as indications of effects under actual operating conditions.

#### Conclusions

The work discussed above shows the breadth of the tritium technology development in the United States. This paper cannot begin to cover all of the many research and development programs in the field. What we hope to show is that there is a large, serious effort underway to develop this technology in a timely manner. With the current ongoing programs the tritium technology should not be an obstacle to the design and construction of the Engineering Test Facility (ETF). These continuing programs will develop, in a timely fashion, the data base and experience necessary to proceed from conceptual design to engineering design and construction of ETF.

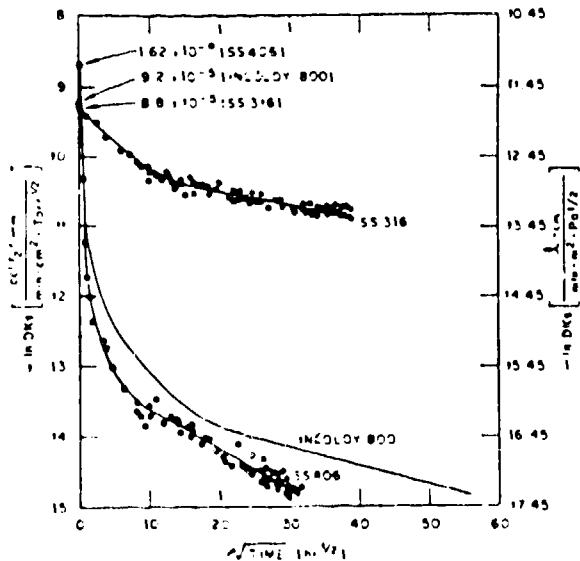


Fig. 3. Tritium permeabilities of three construction alloys at 930 K as the alloys are oxidized by water at 0.94 atm (94 kPa).

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