Tritium

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1 INTRODUCTION

The first generation of controlled fusion devices are being designed to liberate heat from the fusion of a deuteron D and a triton T to form an alpha particle, α , and a neutron, n, in the strongly exothermic nuclear reaction

$$D + T \to \alpha + n + Q, \tag{1}$$

The energy release per reaction is

$$Q = 17.6 \text{ MeV.}$$
 (2)

Using the energy release Q of (1) and neglecting the additional heating due to neutron capture, which can add several MeV per reaction, we find that to produce an average fusion (thermal) power P_F would require a tritum mass consumption rate of

$$\dot{M}_1 = 56 P_F \text{ kg y}^{-1} \text{GWT}^{-1}.$$
 (3)

Here one Gigawatt thermal power has been abbreviated as GWT. A few tens of percent of this fusion (thermal) power can be converted into electrical power, for example, in steam or gas turbines.

Radioactive tritium spontaneously decays to ³He, an electron e^- and an electron antineutrino $\tilde{\nu}_e$ with a half life of 12.3 years in the process of beta-decay

$$T \to {}^{3}He + e^{-} + \bar{\nu}_{e} + 18.6 \text{ keV}.$$
 (4)

The stable isotope ³He, produced when tritium decays, has many uses already, for example, in neutron detectors, in ³He-⁴He dilution refrigeratators to produce millikelvin temperatures, and in magnetic-resonance imaging. Naturally occurring ³He is so rare that virtually all of the current demand for ³He is met with gas collected from the radioactive decay of tritium, either manufactured for use in nuclear weapons by the United States and the Russian Federation or recovered from the heavy-water of Candu fission reactors. A fusion economy would provide a substantial new resource of 3 He, and would encourage new applications.

Unlike the stable isotope deuterium, which makes up 156 ppm of hydrogen on earth, tritium has a relatively short "shelf life" because of the radioactive decay (4), so tritium is most efficiently used within a few years after its manufacture. The ITER experiment will make use of the 20 kg of T that has been accumulated from Candu reactors. A single, 1 GW(E) fusion power plant, at 30% conversion efficiency from thermal to electrical energy, would use 20 kg of tritium in less than two months.

With these facts in mind, the United States Department of Energy asked JASON to respond to the following charge during the JASON Summer Study of 2011:

2 STATEMENT OF WORK

The MITRE Corporation will contract with DOE with funding supplied through the United States Army (CECOM) to provide the necessary personnel, services, and facilities to support JASON until 31 January 2012. All activities and deliverables called for in this paragraph shall be provided in accordance with the schedule in paragraphs 5.1. Study funding will support the annual integrated program across multiple agencies. All products will be shared subject to classification requirements.

Large-Quantity Tritium Production: Tritium breeding is an essential component of potential future GWE sources of electrical energy based on nuclear fusion. Such reactors require kg quantities of tritium per year of operation which must be bred as part of the overall reactor cycle. Traditionally, tritium is assumed to be bred from neutrons involved in fusion energy-production processes of the particular type of reactor using a lithium (Li) "blanket" or related alloys such as the Pb-17Li eutectic. As such, tritium breeding is intimately connected with energy production, thermal management, radioactivity management, materials properties, and mechanical structures of any plausible future large-scale fusion power reactor. JASON is asked to examine the current state of scientific knowledge and engineering practice on the physical and chemical bases for large-scale tritium breeding. In particular, JASON's report should address the following questions and provide answers with supporting analysis:

Specific questions:

1. How realistic are 1GWE-scale tritium breeding blankets from today's perspective?

- 2. What are the most important development issues from today's perspective?
- 3. Do tritium breeding considerations favor certain classes of fusion reactors over other possible choices?

3 BRIEF ANSWERS TO SPECIFIC QUES-TIONS

Brief answers to the specific questions are given here. Reasons for these answers will be outlined in the remainder of the report.

1. How realistic are 1GWE-scale tritium breeding blankets from today's perspective?

Blankets are in the advanced conceptual design stage. Some detailed modeling and experimental tests have been done. This work shows that in principle it is possible for a well-designed reactor to breed enough tritium to replace what it burns, to make up for losses in reprocessing, and to produce enough surplus every few years to fuel a new reactor. Many important engineering issues need to be solved, for example, possible corrosion of the pipes by circulating blanket material like molten Li or PbLi metals, extraction and purification of tritium from the blanket, tritium leakage, etc.

2. What are the most important development issues from today's perspective?

Since the detailed fate of 14 MeV neutrons from the fusion reaction (1) must be precisely known to predict tritium breeding, a sufficiently accurate data base of neutron cross sections must be certified. In assembling this data base, we recommend using methods similar to those used in the stockpile stewardship program, "Quantification of Measurements and Uncertainties." Cross sections deemed to be of insufficient accuracy should be measured more carefully. Additional experiments should be done to more accurately quantify the difference between predictions of the tritium-breeding codes and experimental measurements. Requirements for tritium-breeding should be one of the key tradeoffs in reactor design. For example, maximizing the burnup fraction f_b (a plasma-physics issue) and minimizing the reprocessing time t_p of unburnt tritium (a chemical-engineering issue) are essential to minimize both the required tritium inventory and the required tritium breeding ratios.

3. Do tritium breeding considerations favor certain classes of fusion reactors over other possible choices?

Blankets of magnetic confinement fusion (MCF) reactors are constrained in space by magnetic field coils, conduits for initial plasma heating power, divertor placement, etc. Many of these elements also contain neutron-absorbing structural materials. Inertial confinement fusion (ICF) designs, which are less mature than those of MCF, also have constraints, for example, paths for target injection and for driver beams like lasers or heavy ions, the need for walls capable of withstanding the periodic impulsive loads from exploding pellets, and openings to allow rapid clearing of the chamber debris after each pellet implosion. All of these factors limit the fraction of neutrons that can participate in breeding. At the present level of understanding, blanket design appears somewhat less constrained for ICF than for MCF machines. For both MCF and ICF, adequate tritium breeding is feasible, and issues other than tritium breeding are likely to be much more important in assessing relative advantages of MCF and ICF.

4 CONSTRAINTS OF BASIC PHYSICS

The feasibility of tritium breeding depends on both basic physics and engineering issues. In the two-body reaction (1), about 4/5 of the 17.6 MeV energy release, or 14.1 MeV is carried by the neutron and 1/5 or 3.5 MeV is carried by the alpha particle, which heats the magnetically or inertially confined plasma. Most of the neutrons are absorbed in a breeding blanket, where they produce more than one triton per neutron to make up for neutrons that escape through gaps in blanket coverage or are absorbed by structural material, to make up for losses in the tritium handling system, and to provide excess tritium for new reactors. The tritium breeding ratio (TBR),

$$TBR = \frac{\text{tritium bred}}{\text{tritium burnt}} \tag{5}$$

is defined as the average number of tritium atoms bred per tritium atom burnt in the reaction (1). We must have TBR > 1, for a self-sustained fusion economy.

Blankets with Pure Lithium. With proper design, it is possible to obtain TBR > 1 in lithium-containing blankets with some combination of the reactions

$$n + {}^{6}Li \rightarrow T + \alpha + 4.8 \text{ MeV},$$
 (6)

$$n + {}^{7}Li + 2.5 \text{ MeV} \rightarrow T + \alpha + n'.$$
 (7)

The product neutron n' of (7) has a smaller energy than the incident neutron n. The per cent atomic abundances of naturally occurring Li are

The exothermic reaction (6) has a very large cross section, 940 b, for neutrons with a velocity of 2200 m/s [1]. The cross section scales very accurately as

1/v for energies up to about 60 keV. Consequently, with careful selection of structural materials and geometry to minimize the loss of neutrons by absorption or escape from the blanket, nearly all of the neutrons that slow down to thermal energies from their initial 14.1 MeV can be absorbed by ⁶Li and can generate tritium. The reaction (6) also makes a substantial contribution to the thermal power output of the fusion reactor since the energy release per reaction, 4.8 MeV, is 27% of the 17.6 MeV energy release of the primary fusion reaction (1).

The cross section of the second, endothermic reaction, (7) is heavily suppressed because of the Coulomb barrier for charged product particles at energies just above the 2.5 MeV threshold, but it has a cross section of order 0.3 b for neutrons with energies above 5 MeV. It can be readily driven by the 14 MeV neutrons from the primary fusion reaction (1), and it allows a single neutron to make two or more tritons. For example a 14 MeV neutron can break up a ⁷Li into a T and an α according to (7). If the slower, product neutron, n', happens to retain more than 2.5 MeV after breaking up a ⁷Li nucleus, it can break up a second ⁷Li and release another T before falling below the energy threshold for the reaction (7), when it can still be captured on ⁶Li, in accordance with the reaction (6), to form a final T.

Blanket with neutron multipliers. A second way to get relatively large tritium breeding ratios is to include neutron multipliers, notably beryllium (Be) and lead (Pb), in lithium blankets. The most promising neutron multiplication reactions are

$$n + {}^{9}Be + 3 MeV \rightarrow 2\alpha + n' + n'',$$
 (9)

$$n + Pb + 10 \text{ MeV} \rightarrow Pb + n' + n''.$$
 (10)

One of the product neutrons n' from the low-threshold, (n,2n) reaction (9) may occasionally retain enough kinetic energy to drive another cycle of the reaction (9) or (7), but in most cases the product neutrons will be below



Figure 1: Cross sections for the reactions (6), (7), (8) and (10). Other important cross sections include elastic and inelastic scattering cross sections for Li, Be and Pb, needed to model the slowing down (moderation) of the 14 MeV primary neutrons, and neutron absorption cross sections for structural materials (Figure from M. Sawan).

thereshold and can make tritium only through capture on ⁶Li in accordance with (6). Therefore, when using the (n,2n) reactions of (9) and (10), one can almost always increase the TBR by using lithium that has been enriched in the isotope ⁶Li. The energy-dependence of cross sections for the most important reactions for tritium breeding are shown in Figure 1.

4.1 Neutronics

As discussed above, under ideal circumstances it is possible to produce nearly two tritons for every 14 MeV neutron. In pure lithium of natural isotopic composition this would give an ideal tritium breeding ratio, TBR ≈ 2 . But many practical engineering issues will limit the TBR to smaller values. With sufficiently reliable values for the cross sections of key reactions like (6), (7), (9), (10), etc., and with numerical methods that fully account for the heterogeneous geometry and structural materials of the reactor, it should be possible to calculate values of the TBR that will be within a per cent of those that are observed. Analogous calculations for fission reactors predict the multiplication factor and its dependence on control rod positions and other reactor parameters to a small fraction of a per cent. However, many decades of work have gone into perfecting neutronic computational methods for fission reactors and in "tuning" the codes by comparing their predictions with observations. Analogous codes for tritium breeding have had less time for perfection and less measured data to use for tuning.

Much progress is being made on tritium-breeding neutronics, notably by University of Wisconsin Professors Mohammed Sawan, Laila El -Guebaly and their collaborators in Madison, and by University of California Professor Mohammed Abdou and his collaborators in Los Angeles. Work in this area should be continued to identify any neutronics issues that differ substantially from those of fission reactors. Since tritium breeding depends critically on reactions with much faster neutrons than those normally encountered in fission reactors, key fast-neutron cross sections of breeding and structural materials need to be evaluated with particular care.

4.2 Tritium Burnup Fraction

The probability that a triton injected into a reactor is burned in the reaction (1) before it escapes the confinement region in the case of MFE, or before the capsule disassembles in the case if ICF, is called the tritium burnup fraction, f_b . Unburned tritium is collected and used again as fuel. As we will discuss in more detail below, f_b should be as close to unity as possible to

minimize the required inventory of tritium and to reduce the required values of the tritium breeding ratio.

Let the number densities of tritons and deuterons in the plasma be [T] and [D] respectively. The reaction (1) will cause [T] and [D] to decrease at equal rates

$$\frac{d[\mathbf{T}]}{dt} = \frac{d[\mathbf{D}]}{dt} = -[\mathbf{T}][\mathbf{D}]\langle v\sigma\rangle.$$
(11)

where $\langle v\sigma \rangle$ is the product of the velocity-dependent cross section σ for the reaction (1) and the relative velocity v of the colliding pairs, averaged over a Maxwellian distribution of velocities at an ion temperature T_i , assumed to be the same for D and T. The rate coefficient, in m³ s⁻¹, for ion temperatures T_i in keV can be well approximated by [2]

$$\langle v\sigma \rangle = \frac{3.68 \times 10^{-18}}{T_i^{2/3}} \exp\left(-\frac{19.94}{T_i^{1/3}}\right).$$
 (12)

For a representative MFE ion temperature

$$T_i = 20 \text{ keV}, \tag{13}$$

we find from (12) that

$$\langle v\sigma \rangle = 3.22 \times 10^{-22} \text{ m}^3 \text{ s}^{-1}.$$
 (14)

Over the range of validity of (12), the reaction rate increases rapidly with temperature since

$$\frac{d}{dT_i}\ln\langle v\sigma\rangle = \left(\frac{19.94}{3T_i^{1/3}} - \frac{2}{3}\right)\frac{d}{dT_i}\ln T_i \tag{15}$$

At the the temperature $T_i = 20$ keV, we find

$$\frac{d}{dT_i}\ln\langle v\sigma\rangle = 1.8\frac{d}{dT_i}\ln T_i \tag{16}$$

So at a nominal plasma temperature of 20 keV, a 1% increase in ion temperature will increase the reaction rate by 1.8%.

Suppose the reactor is stoichiometrically fueled so

$$[N] = [T] = [D],$$
 (17)

where [N] is the common number density. Then one can readily solve (11) to find that the number density $[N_{\tau^*}]$ at time $\tau^* > 0$ is related to the number density $[N_0]$ at the time of fuel injection by

$$[\mathbf{N}_{\tau^*}] = \frac{[\mathbf{N}_0]}{1 + \langle v\sigma \rangle [\mathbf{N}_0]\tau^*}.$$
(18)

The burnup fraction is

$$f_b = 1 - \frac{[\mathbf{N}_{\tau^*}]}{[\mathbf{N}_0]} = \frac{\langle v\sigma\rangle[\mathbf{N}_0]\tau^*}{1 + \langle v\sigma\rangle[\mathbf{N}_0]\tau^*}.$$
(19)

For magnetic confinement fusion, the effective time for burning, τ^* , can be bigger than the ion confinement time τ_c of the plasma, because some fraction R of the ions that escape the plasma can recombine and reenter the plasma as neutral atoms. Then the effective confinement time τ^* is

$$\tau^* = \tau_c + \tau_c R + \tau_c R^2 + \dots = \frac{\tau_c}{1 - R}.$$
 (20)

Experimental evidence so far suggests that $R \ll 1$ so τ^* will not be much bigger than τ_c . Using (14) and (19) and representative numerical values for magnetic confinement machines,

$$[N_0] = 10^{20} \text{ m}^{-3}, \quad T_i = 20 \text{ keV} \text{ and } \tau^* = 2 \text{ s},$$
 (21)

we find

$$f_b = \frac{0.064}{1.064} = 6.02\%. \tag{22}$$

The burnup fraction anticipated for the International Thermonuclear Experimental Reactor (ITER) is considerably smaller, less than 1%. Proponents of inertial confinement fusion hope to achieve burnup fractions as high at 30%. Large burnup fractions help to keep the required tritium inventory low, as we will discuss in the next section.

4.3 Tritium Inventory

Unburnt fuel from the reactor is sent to a tritium reprocessing system that takes a mean time t_p to clean up and recycle the tritium. The continually reprocessed tritium is injected into the plasma with an efficiency η . The efficiency can be relatively high, several tens of per cent, for frozen pellet injection, but it appears to be relatively low, perhaps only a few percent, for gas injection at the edge of the plasma. Then to provide the burn rate (3), and to make up for radioactive decay at the rate

$$\gamma_s = \frac{\ln 2}{12.3} \, \mathrm{y}^{-1},\tag{23}$$

and the loss rate γ_r in the reprocessing of unburnt T, we must have

$$\frac{\eta f_b M_0}{t_p} * TBR = \dot{M}_1 + (\gamma_s + \gamma_r) M_0,$$
(24)

where \dot{M}_1 , the burn rate needed to power the plant, was given by (3) and M_0 is the time-independent, recirculating tritium inventory. As we discuss in the next section, in addition to the recirculating inventory M_0 , there will normally be a growing inventory m of tritium being bred for additional reactors, with a separate processing and storage system. From (24) we see that the inventory is

$$M_0 = \frac{\dot{M_1}}{TBR * \eta f_b/t_p - \gamma_s - \gamma_r}.$$
(25)

From (25) we see that the relative tritium production, $TBR * \eta f_b/t_p$ must at least exceed the relative loss rate $\gamma_r + \gamma_s$ from the reprocessing loop and from radioactive decay. If the breeding rate is large compared to the loss rate, and $TBR \approx 1$ we can approximate the mass inventory (25) by

$$M_0 \approx \frac{t_p M_1}{\eta f_b}.$$
(26)

For example, if

$$f_b = 0.05,$$

$$\eta = 0.5,$$

$$t_p = 1 \text{ day},$$

$$P_F = 1 \text{ GWT},$$
(27)

Equation (26) gives

$$M_0 = 6.14 \text{ kg.}$$
 (28)

This is a large amount of tritium, but much less than the 56 kg that must be burnt per year to produce 1 GW of heat. We see from (26) that the recirculating inventory can be minimized by minimizing the reprocessing time t_p and by increasing the injection efficiency η and burn fraction f_b with respect to the representative values of (27).

The estimates above assumed no breeding of additional tritium, a topic we address in the next section.

4.4 Tritium Breeding Ratio

The blanket is designed to breed enough tritium to at least make up for that which is burnt, and what is lost in reprocessing and radioactive decay. Moreover, after a desired doubling time t_d , typically a few years, enough extra tritium should be produced to provide the initial inventory for an identical reactor.

Denote the mass of the tritium inventory by

$$M = M_0 + m, (29)$$

where the time-independent circulating mass, M_0 , needed to run the reactor was given approximately by (26) and m is the growing, time-dependent mass that is being bred for fueling the next reactor. Radioactive decay at the rate γ_s , losses at the rate γ_r in the reprocessing loop, burning and breeding will cause the refueling mass to change at the rate

$$\frac{dm}{dt} = -\gamma_s m - (\gamma_s + \gamma_r) M_0 - \dot{M}_1 + \dot{M}_1 * \text{TBR}$$
$$= -\gamma_s m + \kappa M_0. \tag{30}$$

where we can use (25) with (30) to write the net production rate coefficient as

$$\kappa = \left(\frac{\eta f_b}{t_p} - \gamma_s - \gamma_r\right) (\text{ TBR} - 1) - \gamma_s - \gamma_r.$$
(31)

For no breeding, m = 0 and dm/dt = 0, (30) gives (24). Solving (30) under the assumption of a positive tritium production rate $\kappa > 0$, and assuming m = 0 at time t = 0 and $m = M_0$ at the doubling time, $t_d > 0$, we find

$$\kappa = \frac{\gamma_s}{1 - e^{-\gamma_s t_d}}.\tag{32}$$

For certain ranges of the parameters η , f_b , t_p , t_d , γ_s and γ_r we can equate the expressions (31) and (32) for κ to find the required tritium breeding ratio TBR. For simplicity, consider sufficiently short doubling times t_d (a few years) that there is negligible radioactive decay of the tritium and $\gamma_s t_d \ll 1$. Then we can approximate (32) by $\kappa \approx 1/t_d$ and neglect γ_s and γ_r in (31) to find that the required tritium breeding ratio is

$$TBR - 1 \approx \frac{t_p}{\eta f_b t_d}.$$
(33)

For example, if we use the parameters of (27) in (33) with a doubling time, $t_d = 3$ y, we find a required tritium breeding ratio

$$TBR \approx 1.04. \tag{34}$$

From (33) we see that to have the least demanding requirements on the tritium breeding ratio it is necessary to use the longest possible doubling times, t_d , the largest tritium burn fraction f_b , the largest injection efficiency η and the smallest recycling time t_p .

5 ADDITIONAL ISSUES

In this section we discuss issues related to tritium breeding that go beyond the explicit questions of our charge.

5.1 Deuterium-Rich Fueling

The fusion community may have already considered deuterium-rich fuel mixtures, but we think this is worth serious consideration since deuteriumrich mixtures could be more favorable for tritium breeding than stoichiometric mixtures with equal atomic fractions of of D and T. For DT fueling, three other fusion reactions that occur at the same time as (1) are

$$D + D \to T + p + 4.04 \text{ MeV}, \tag{35}$$

$$D + D \rightarrow^{3} He + n + 3.27 MeV,$$
 (36)

$$T + T \to \alpha + 2n + 11.4 \text{ MeV.}$$
(37)

The rate coefficients, $\langle v\sigma \rangle$, corresponding to the reactions (35) and (36) are about 200 times smaller than for the main reaction (14) and the energy release per reaction is substantially less[3]. The rate coefficient for the reaction (37) is about 90 times smaller than (14). Using non-stoichiometric, deuteriumrich fuel, which would enhance the contribution of the reactions (35) and (36) and supress the contribution of (37) to the burning, could be advantageous to the breeding cycle because:

• Deuterium-rich fuel could substantially increase the burnup fraction f_b of the tritium. The larger the burnup fraction, the smaller the required inventory of tritium and the smaller the required tritium breeding ratio.

• The reaction (35) makes tritium directly, and the reaction (36) makes the valuable isotope ³He and a neutron which can convert ⁶Li to T in the blanket.

For example, suppose that the number density $[N_0]$ of (21) is sufficient to provide the desired thermal output of the reactor with stoichiometric fuel. Then we would get the same thermal output from a non-stoichiometric fuel with triton and deutron number densities give by

$$[D][T] = [N_0]^2. (38)$$

Call the relative deuterium enrichment

$$\epsilon = \frac{[\mathbf{D}]}{[\mathbf{N}_0]}.\tag{39}$$

The relative burn rate, γ_{ϵ} , of tritium will then be

$$\gamma_{\epsilon} = -\frac{1}{[T]} \frac{d[T]}{dt} = \epsilon \gamma_1.$$
(40)

where the stoichiometric burn rate is

$$\gamma_1 = [N_0] \langle v\sigma \rangle. \tag{41}$$

Other things being equal, the plasma pressure will scale as the sum of the deuterium and tritium number densities, so enhancing the deuterium content by ϵ will cause the ion pressure p_0 for a stoichiometric fuel mix to increase to

$$p = \frac{p_0}{2} \left(\epsilon + \frac{1}{\epsilon}\right). \tag{42}$$

or

$$p - p_0 \approx \frac{p_0}{2} (\epsilon - 1)^2$$
, if $|\epsilon - 1| \ll 1$. (43)

For example, consider a modest, 30% D enrichment of

$$\epsilon = 1.3. \tag{44}$$

We see from (40) that the tritium burn rate will increase by a factor of 30%. If the required 4.5% ion pressure increase from (43) has a negligible effect on the ion confinement time, we would also get an approximately 30% increase in burn fraction. According to (25) and (33) this would decrease both the required tritium inventory M_0 and the breeding-ratio excess, TBR-1, by about 30%. The contribution of the reactions (35) and (36) to the burning would be increased by a factor of $1.3^2 = 1.69$. But the rates (35) – (36) are so small compared to the rate of the main DT fusion reaction (1), that their increased contribution from deuterium-rich fueling will not be a very important factor compared to the substantially larger burn fraction f_b , and the resulting decreases of the required tritium inventory and tritium breeding ratio.

5.2 Lithium and Lead-Lithium Blanket

Pure lithium and lithium-lead (Pb-Li) mixtures have been investigated in some detail as potentially useful blanket materials, that could also provide some or all of the heat transfer to the balance of the plant for partial conversion to electrical energy.

Pure lithium has a very high electric conductivity, and rapid pumping of liquid lithium against the large magnetohydrodynamic (MHD) forces of MCF reactors will be very difficult. Pure lithium metal is often envisaged as a combined breeding and heat transfer medium for ICF reactors, which do not require such large magnetic fields.

A currently interesting MCF design envisages using a homogeneous mixture of the liquid metals, Pb and Li, as the tritium breeding blanket and to provide some heat transfer. The bulk of the heat transfer would be provided by circulating helium gas for which there are no MHD issues. The phase diagram of the Pb-Li system, shown in Figure 2. There is a eutectic compo-



Figure 2: Phase diagram of the PbLi system[5]. Many blanket designs envisage using the eutectic mixture of about 15.8% Li, with a melting point of about 235 C.

sition, 15.8% (atomic) Li and 84.2% (atomic) Pb, with a minimum freezing temperature of 235 °C. In most of the briefings we received, the breeding blanket was assumed to have the eutectic composition. A NaK eutectic is used in experimental fast-neutron fission reactors to facilitate frequent refueling with a liquid-metal coolant that remains liquid at room temperature or below. The Pb-Li eutectic melting point is well above room temperature so it is not clear that the eutectic mixture would have any advantages over a mixture with a larger Li fraction and better tritium breeding potential. We recommend that whatever considerations went into selecting the eutectic composition for the Pb-Li blanket be reviewed.

Solid eutectics are easy to make in a reproducible manner. If a melt that does not have the eutectic composition is cooled, the phase rich in whichever component is more abundant than the eutectic composition will freeze first. If Li-rich it will float to the surface; if Pb-rich it will sink to the bottom. When the temperature reaches the eutectic point the remaining material, of eutectic composition, will freeze homogeneously. It is only necessary to cut off the non-eutectic layer at the top or bottom of the solid ingot, and the remaining material will have the eutectic composition and can be repeatedly remelted and refrozen as a homogeneous solid.

Liquid mixtures of non-eutectic composition can be easily made by melting weighed quantities of their components. In fusion breeders the temperature of the blanket material is supposed to range from about 500°C to 700°C, and for this range one can see from Figure 2 that compositions with Li fractions up to 40 % (atomic) should remain liquid, and could have better tritium breeding potential.

Initial melting and mixing is not trivial because of the large density mismatch between Pb and Li, but once a liquid is mixed it remains mixed until frozen. Once frozen, careful mixing is necessary to homogenize the remelted material if the eutectic composition is not used. Magnetohydrodynamic mixers have been developed [5] and could be useful for the initial mixing of non-eutectic compositions.

For relatively inexpensive and non-hazardous tests of tritium breeding in Pb-Li, it would be useful to use solid, room temperature samples. Maintaining such samples above their melting point would introduce significant additional costs and hazards. A suitable proxy material could consist of interleaved sheets of eutectic and pure materials (or of the two pure materials) in the appropriate thickness ratios. The probing neutron beam should enter the stack of sheets perpendicularly (or obliquely). Because the fast-neutron interaction length in any solid-density material is several cm or more, sheets of 1 cm thickness could provide a satisfactory approximation to homogeneity.

It is often remarked that the relative amount of the isotopes ⁶Li and ⁷Li in Pb-Li blanket material can be chosen to optimize tritium breeding. It is also true that the relative amounts of Pb and Li can be chosen to optimize breeding and the eutectic composition probably has too little Li for optimum performance. As discussed above, there is no obvious reason to be limited to the eutectic composition of a melt.

5.3 More Tritium is Better

There has been some consideration of deliberately decreasing the TBR, for example, but adjusting the relative fraction of ⁶Li and ⁷Li in blanket material, to avoid producing more tritium than needed for fueling existing reactors, providing fuel for additional reactors, and making up for losses from processing and radioactive decay.

We doubt that overproduction of tritium is a real problem. Tritium is readily stored as metal tritides, at modest cost and with no significant hazard. Tritium is a valuable not only for fusion energy, but because it is the only currently practical source of ³He. Demand for ³He far outstrips the supply. The largest projected demand at present is for slow-neutron detectors, which DHS wants to proliferate in great numbers to counter nuclear terrorism. An additional growing use for ³He is in medical imaging of the lung. There is no substitute.

5.4 Tritium Handling and Leakage

In steady-state operation relative small quantities of tritium must be extracted from a much larger mass of blanket material. Although the quantity of tritium produced is relatively small, there will be stringent regulatory constraints on its emission to the environment in any chemical form. Gaseous HT and the vapor HTO are of most concern, the latter because of its ready substitution for H_2O in most chemical and biological systems. A plausible regulatory limit is 1–3 g year⁻¹ of tritium loss. This is about 10⁻⁴ of the tritium burnt in one year to make 1GW of thermal energy. Material control at this level can be achieved with careful chemical engineering, but tritium presents special problem because of its volatility, diffusivity in many materials, and the ready formation of mobile and reactive compounds like HTO.

Tritium extraction from the breeding material is complicated because large amounts of heat may also need to be extracted through heat exchangers with significant tritium permeability. Thus tritium could contaminate the heat transfer medium (for example helium) in the secondary loops of heat exchangers. Materials with low-permeability to tritium at elevated temperatures are needed. A representative material is tungsten with a permeability of 1×10^{-7} in units of cm³(STP) [m s kPa^{1/2}]⁻¹ at 700 C. This extrapolates to 10^{-23} in the same units at room temperature. To get the permeability current in cm^3 of T_2 gas (at standard temperature and pressure) per second, one must multiply the permeability by the difference in the square root of the pressure in kPa times the ratio of the area of the barrier to its width. The dependence on the square root of the pressure is called Sievert's law, and it comes from the dissociation of T_2 to atomic tritium in the metal. The diffusion coefficient of T_2 in alumina, where there is no dissociation of the T_2 molecule, is 1×10^{-15} m² s⁻¹ at 700 °C. The room-temperature value is extrapolated to be 10^{-31} m² s⁻¹. Such extrapolations are necessarily uncertain because they assume the temperature dependence of permeabilities follows an Arrhenius law into temperature ranges far from those for which direct measurements are available. Unrecognized transport mechanisms with small prefactors but lower activation energies could give much larger permeation coefficients at low temperature. Direct measurements extend to 1×10^{-10} cm³(STP) [m s kPa^{1/2}]⁻¹ for tungsten and to 10^{-20} m² s⁻¹ for alumina, with no sign of deviation from Arrhenius temperature dependence. Some of these coefficients were measured for protium or deuterium, and the values for tritium are expected to be smaller because tritium's greater mass reduces the tunneling that can play an important role diffusion and permeation of hydrogen isotopes.

In principle, it is possible to reduce tritium leakage to negligible levels with multiple barriers of small, well-characterized permeabilities. However, in practice, things may go wrong. For example, material corrosion by tritiated vapors (HTO, HT) may lead to leaks. Because of their ionizing radiation, tritiated gases are known to be especially corrosive. There are bound to be unanticipated problems, as has been true for every new technology.

Detriviation of hydrogen-containing (such as water vapor) plant exhaust is, in principle, feasible to any desired degree of completeness with enough stages of electrolytic or other isotopic separation. The basic performance (physical chemistry) parameters are known; this is not a new technology, and can be engineered to any desired limits. As for any highly engineered system, the issue is unanticipated failure, not the nominal calculated leakage rate.

In summary, the chemical engineering issues connected to tritium breeding will be a very challenging. While these are hard problems, we do not see them as show-stoppers. Judging from the briefings we heard, the world fusion energy program is well aware of tritium handing issues and has effective research and development programs underway to address uncertainties.

5.5 ⁶Li, Tritium and ³He Economics

The cost of 60% enriched ⁶Li has been estimated to be \$800/kg, or $$1,333 \text{ kg}^{-1}$ of ⁶Li in the enriched product [6]. As a first approximation, we assume the cost of further enrichment is proportional to the additional separative work units (SWU's) required. Assuming a tails fraction of 0.0375 (half the natural ⁶Li abundance of 7 %) for both the enrichment to 60% and the subsequent enrichment from 60% to 90%, we find that enrichment to 60% requires 16.5 SWU per unit of ⁶Li, while further enrichment to 90% requires an additional 3.7 SWU per unit of ⁶Li. Hence we estimate a cost of 90%

enriched ⁶Li of $1,600 \text{ kg}^{-1}$ of ⁶Li in the enriched product. Of course this is not a very refined estimate since the cost should include many other factors, for example, the capital cost of equipment, which is larger at low enrichment because more material must be handled.

With efficient conversion, one ⁶Li makes one triton. So to make 1 kg of tritium requires 2 kg of ⁶Li, and if the ⁶Li is from material enriched to 90 %, the contribution to the cost from enriched lithium is $3,200 \text{ kg}^{-1}$. The cost of the lithium is definitely the "tail of the dog", with tritium prices in the range $10,000-30,000 \text{ g}^{-1}$ (g, not kg!), and a speculative future cost (Willms: fire.pppl.gov/fesac_dp_ts_willms.pdf) of $100,000-200,000 \text{ g}^{-1}$.

Fusion reactors may be a significant source of tritium for purposes other than fusion fuel. A 2.5 GWth, 1 GWe fusion plant with TBR = 1.10 and 33% efficiency for converting heat to electrical energy, would produce about 17 kg y ⁻¹ of tritium beyond the 170 kg needed to replace what must be burnt. If the electric production of 3.16×10^7 GJ y⁻¹ = 8.8×10^3 GWhr y⁻¹ is valued at \$0.10 kWhr⁻¹ the electric power produced (at 100% duty cycle) is worth \$ 8.8×10^8 y⁻¹. At \$100,000 g⁻¹ for 17 kg y⁻¹ of tritium, the value of the tritium produced would be \$ 1.7×10^9 y⁻¹, more than the value value of the electrical energy. Although the price of \$ 100,000 g⁻¹ for tritium is completely unrealistic, the point is that overproduction of tritium could be a significant contribution to the economics of the reactor. Before priorities at the Department of Homeland Security made it difficult to purchase ³He at any price, it was available commercially at around \$100 per liter (at standard temperature and pressure), or \$ 2,240 per mole, or about \$ 750 g⁻¹. We have see prices quoted as high as \$ 4,000 g⁻¹ [7].

Of course, the prices mentioned above are artificial and not free market prices. Should fusion power provide sources of tritium and ³He that are orders of magnitude greater than present production, a real market for both tritium and ³He could develop, with prices completely different from those

prevailing today. Abundant new supplies of tritium and ³He will create new applications. For example, escape path lighting (as on airplanes) could be put in every building, just as public buildings now have exit signs, often powered by tritum decay. We regularly hear of people, including firemen, dying in fires because the become disoriented in the smoke and cannot find their way out.

5.6 Quantification of Margins and Uncertainties

Although it is our judgement that adequate tritium breeding is possible for both MCF and ICF, we recommend that the key uncertainties of breeding be quantified. A well-established formal method for dealing with uncertainties in predictions of untested complex systems is called Quantification of Margins and Uncertainties [8, 9, 10]. The reader is referred to these references for details. This methodology was developed to understand and quantify the confidence that can be placed in performance margins of untested systems in the presence of uncertainties. In the case of nuclear weapons there is a moratorium on testing for the indefinite future; in the case of fusion power reactors, there will be no reactors for full-scale tests for many years, probably several decades. So quantification margins and uncertainties will helpful to both the fusion energy and nuclear weapons programs to point research and development toward problems of highest priority.

5.7 Tritium Breeding Issues for ICF

Research on inertial confinement fusion (ICF) has been focussed on achieving ignition, and little work has been done on the breeding issue beyond some conceptual paper studies. Nevertheless, the overall neutron economy of an ICF power plant would be broadly similar to a MCF plant, and much of the technical discussion already given in this report carries over to ICF. This subsection will explain these similarities and highlight the differences.

The most expensive capital cost in an ICF plant is likely to be the driver, whether it be a laser, a heavy-ion accelerator, or (more speculatively) a pulsed power machine. The driver must deliver energy to the fusion target, which contains a capsule of DT fuel. The drive causes the fuel to implode, ignite, and deliver yield in the form of neutrons, alpha particles, x-rays, and debris. Yield per pellet might be in the range 100 MJ - 1000 MJ, and a power plant of capacity 1000MWe would therefore need to inject and burn between 2 and 25 targets per second.

A fusion chamber several meters in diameter is required to contain the fuel capsule explosions. As with MCF, the first wall is a crucial component, but in ICF it must tolerate pulsed energy deposition on sub-microsecond timescales. In some conceptual designs, a buffer gas at low pressure (perhaps xenon at 10 torr) helps protect the first wall from x-rays.

Behind the first wall, coolant flows and breeder blankets must operate, just as in MCF. The coolant and breeder may well be the same material, such as liquid Li or Li-Pb mixtures. In some conceptual designs, the first wall is a "waterfall" of liquid lithium that serves both as a blanket and heat transfer medium while helping to protect the innermost solid wall.

In any case, tritium is bred in the blanket material, and must be recovered. Moreover, unburned tritium must be recovered from the fusion chamber. The recovered tritium must be purified and used to manufacture new targets, at high efficiency and low loss. Target manufacture would probably take place right at the plant, to minimize transport and delay, and is likely to represent the single largest operational cost for the plant. Some of the principal differences in tritium breeding for MCF and ICF are:

- 1. If ICF is to work at all as a power plant, the target must burn fuel at fairly high efficiency (30% has been projected), in contrast to MCF, where the burnup of injected fuel may be only a few percent; thus the quantities of tritium recovered from ICF chamber would be smaller than for MCF.
- 2. Since an ICF chamber needs no strong magnetic fields, flow of conducting coolant/breeder would not be impeded by magnetohydrodynamic effects. (The exception might be pulsed-power ICF plants, where strong transient fields might exist.)
- 3. Manufacture of targets for ICF is enormously more challenging than manufacture of fuel pellets for MCF. An ICF target is a precision part, which must survive injection at high speed (100s of m/s) into the hot fusion chamber. In the case of indirect drive, the fuel capsule is enclosed in a cm-sized hohlraum.
- 4. An ICF chamber will not need a central post and will have different penetrations in its first wall compared to MCF, changing the geometrical neutron losses. Some ICF conceptual designs envisage relatively small geometrical losses compared to those of MCF.

5.8 Sources of Tritium

To indicate the scale of the problem, recall that a full-scale fusion reactor generating 3 gigawatts thermal burns about 150 kilograms of tritium per year. We note the following issues:

1. Fission reactors. A basic disadvantage of fusion when compared with fission is that even the most energetic fusion reaction (1) produces

about 20 MeV energy per neutron while fission produces about 200 MeV per neutron. At present, tritium is produced in fission reactors, but if fission is used to breed fuel for fusion, the tail would be wagging the dog.

- If a fission reactor is dedicated to producing tritium, the maximum yield is about one atom T per fission. 10 watts of thermal fission energy will produce tritium fuel for 1 watt of fusion energy. This makes no economic sense, unless the tritium is used as a temporary input to get a fusion reactor started. The fusion reactor must be a breeder with a breeding ratio greater than unity.
- CANDU reactors. The only source of non-military tritium is the CANDU reactors, 21 operating in Canada, 4 in Korea and one or two in India, China, Rumania, Pakistan, and Argentina. From all of these, about 1.8 kilograms tritium per year is recovered from the heavy water that is used for cooling and moderation. About 100 grams of tritium per year are sold at a price of \$ 30K per gram. The CANDU are mostly scheduled to retire around the year 2025. The stockpile remaining in Canada is about 20 kilograms. This will be just sufficient to supply the ITER machine, which is not a breeder, with tritium for 10 years of experiments. There will not be enough left over to make a significant contribution to future fusion reactors.
- Other commercial fission reactors, using light rather than heavy water, require substantial modification if they are to be used for tritium production. For example, much of the boron and gadolinium, currently used burnable neutron poisons to stabilize the reactivity over the lifetime of the fuel load, could be replaced by lithium. The modifications would be expensive and would require a lengthy process of development and regulatory certification.
 - In 2003 a commercial TVA reactor at Watts Bar began irradiat-

ing Li-containing rods for tritium production for the U.S. nuclear weapons stockpile. However, an October 2010 GAO report[11] notes that NNSA has not been able to irradiate as many tritiumproducing burnable absorber rods (TPBAR) as it had scheduled because of problems with tritium permeation— an indication of the difficulty of controlling the leakage of tritium.

- 2. An earlier JASON study, (JSR-95-310, unclassified), studied in detail a number of schemes for producing tritium with accelerators. In 1995 we recommended one such scheme to DOE as a practical solution of a DOE supply problem concerned with tritium for nuclear weapons. In that context, the quantity of tritium required was small and predictable, and a high cost per gram was acceptable. If tritium is to be produced for fusion energy, the quantities are much larger and the affordable cost per gram is much smaller. Accelerator schemes suffer from the same circumstances that make tritium production in fission reactors uneconomic. If the accelerator is used to provide a spallation source of neutrons which are absorbed directly to make tritium, the input of energy required to run the accelerator is greater than the output of energy produced by the tritium. If the spallation source is amplified by surrounding it with a sub-critical fission reactor, then the fission reactor might as well be operated as an ordinary critical reactor, and the accelerator becomes superfluous.
- 3. The migmatron promoted by Bogdan Maglich. The migratron is a proposal to run a fusion reactor with ordinary hydrogen and boron as fuel, so that all the energy goes into charged ions and none into neutrons. This scheme would have many advantages as an energy source, if it could be made to work. It would incidentally produce small amounts of tritium as a by-product of its operation. Unfortunately, it has never come close to a practical demonstration. In event that a migma reactor could actually work, it would probably make DT reactors obsolete and

would enormously reduce the demand for tritium. The migma reactor would then be used directly as an energy source and not as a breeder of tritium.

- 4. Requirements for lithium and beryllium. A fusion power reactor generating 3 gigawatts thermal will require about 500 tons of lithium for a breeding blanket using lithium 7 as the source of secondary neutrons. A breeder with the same power using beryllium as neutron source will require about 250 tons of lithium and about 100 tons of beryllium. These materials must be reprocessed periodically but will not be used up.
 - Lithium resources. World production of lithium in 2010 was about 20,000 tons, mostly used for ceramics, but increasingly for lithium batteries. Production is expected to rise rapidly if the population of electric automobiles expands. Proven reserves in 2010 were about 4,000,000 tons, mostly in Argentina, Chile and Bolivia. Estimated resources were about 16,000,000 tons. Compared with these resources, the requirements for tritium breeding are small. Blankets for 100 full-scale fusion breeders would require less than one percent of the resources. If the producers of lithium should try to raise the price dramatically, the electric automobile industry would be priced out of the market before the fusion industry would be seriously affected.
 - Beryllium resources. World production of beryllium in 2010 was about 300 tons. Proven reserves were 16,000 tons, mostly in the USA, and estimated world resources were about 80,000 tons. If all fusion reactors were using beryllium blankets, the proven reserves would be sufficient for 160 full-scale reactors each using 100 tons. If the beryllium production capacity was insufficient or if the price of beryllium was too high, fusion reactors could use alternative blankets using lithium-lead or liquid lithium. In any

case, it is unlikely that shortage of beryllium would be a factor limiting the growth of fusion energy.

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