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THE TRITIUM HANDLING PROBLEM IN CONTROLLED THERMONUCLEAR REACTORS*

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Introduction

The difficult plasma physics problems have almost completely dominated the thermonuclear reactor program for the past 20 years. These problems have been so formidable that there has been a serious question as to whether they could ever be solved, but in the past few years progress has been sufficiently encouraging so that people have begun to ask, "If you are able to achieve a controlled fusion reaction, what will it be good for?" Thus, although we have not as yet been able to achieve the required conditions for sustained controlled thermonuclear reactions, we have attempted to envision what a full-scale plant might look like assuming the favorable resolution of the outstanding plasma physics problems. As a consequence, several conceptual designs have been evolved, and each of these has revealed a number of difficult materials problems. Of these, perhaps the most difficult and pervasive is the tritium handling problem, for it affects the design of virtually every component in the plant.

Typical Reactor

About a half-dozen different approaches to problems of obtaining a controlled thermonuclear reactor are under active development. Rather than attempt to describe each of these, it seems best for the purposes of this paper to consider one typical example. Most of the effort in the thermonuclear reactor program of the U.S., Britain, and the USSR is going into magnetically confined plasmas. While there are many variations of this approach, a favorite is the use of a large solenoid magnet wrapped into a torus as in Fig. 1. The toroidal magnetic field prevents ions in the high temperature plasma from escaping; instead they swirl about in the magnetic field following elements of helical paths.¹

Toroidal Fusion Device.

The bulk of the effort is being directed toward the use of the deuterium-tritium fusion reaction because this can be ignited at a temperature roughly $1/10$ that for the deuterium-deuterium reaction, and it will be difficult indeed to ignite the D-T reaction. The D-T reaction has the disadvantages that it requires fueling with tritium and yields a very penetrating 14 MeV neutron, but both the D-D and D- 3 H reactions are also accompanied by side reactions that generate both tritium and neutrons. Thus, in any case the fuel cycle will entail handling material containing a substantial fraction of tritium and the rate of emission of 14 MeV neutrons will be at least $1/10$ that for a D-T plasma.² Further, D-D and D-T reactions will lead to radiation damage effects from fast neutrons and an amount of structural activation within a factor of roughly 2 of the corresponding values for the D-T fuel cycle.²

Figure 2 gives some insight into the design requirements of a fusion reactor other than plasma physics considerations and shows a cross section through the magnet and plasma region of a toroidal fusion reactor of the type shown in Fig. 1. Note that in the center we have a plasma at a temperature of hundreds of millions of degrees centigrade, a vacuum wall that encloses a liquid lithium blanket region which operates at somewhere between 500 and 1000°C (i.e., at or above the fuel temperature of an LMFBR), then some thermal insulation, a water-cooled shield region which operates at perhaps 100 or 150°C , and finally a cryogenically-cooled superconducting magnet which would operate at 4°K . Thus we have nearly as extreme a set of temperature conditions as man can produce. This drawing also indicates something of the whole complex of difficult design conditions that must be satisfied if one is to have a commercially viable fusion reactor. One of these, for example, is the requirement that we interpose about 2 m of blanket and shielding material between the plasma and the cryogenic magnets in order to protect them from radiation heating.³ This in turn means that, to keep the magnet costs in dollars per kilowatt to a reasonable level, the useful volume in the plasma region should be a large fraction of the total volume enclosed by the magnets. Thus we must go to a large machine, that is, a plasma region with a minor diameter of perhaps 5 m and a cryogenic magnet diameter of around 10 m. This in turn means that, if we build a toroidal type of machine,

Fig. 2. Section through the minor diameter of a toroidal fusion reactor showing the plasma, blanket, magnet shield, and magnet regions.

the overall diameter of the doughnut will have to be over 30 m. As a consequence, the minimum size of an economically attractive machine of this type appears to be quite large with a power output something like 500 to 1000 MW(e). A large diameter for the plasma region is beneficial from the standpoint of plasma physics. A diameter of only 2 or 3 m might prove acceptable, but a larger diameter will reduce losses to the walls.

Basic Requirements

Once tractable solutions to the plasma physics requirements have been worked out, the next most important requirement will be that any fusion power reactor must breed tritium. There is no significant natural supply of tritium, and the cost of making it in a fission reactor would be exorbitant. Thus, the first fusion reactor to deliver any significant amount of power must breed its own tritium. There is only one neutron released per fusion reaction, hence at first thought it may appear to be extremely difficult to do this. However, the 14 MeV neutron from the D-T reaction will induce a substantial number of $n, 2n$ reactions in the course of slowing down in either beryllium or lithium. As a consequence, it turns out that if one supplies a fairly thick blanket of lithium or beryllium around the plasma of a fusion reactor, it is quite easy to breed a surplus of tritium. Unfortunately, the amount of lithium or beryllium required will be very large — of the order of hundreds of tons per reactor. Fortunately, a check of the world's resources discloses that there is plenty of lithium available to support a world-wide fusion reactor economy based on lithium, but the resources of beryllium, including low grade ore, appear to run only about 30,000 tons, or far less than the amount required for a full-blown fusion reactor economy.⁴ Thus, the designer has no choice but to provide a thick blanket of lithium surrounding the plasma. All that he can do is to reduce the amount of lithium required somewhat by including a thick layer of graphite as in Fig. 2.

As discussed in a companion paper, the use of a lithium blanket raises some difficult materials problems. To obtain a high thermal efficiency, it is desirable to operate the blanket at as high a temperature as possible. Stainless steel could be used, but it is subject to corrosion by lithium if the temperature is raised above about 500°C. With niobium it would be possible to operate at much higher temperatures, and this should make it possible to reduce the waste heat to less than half the amount currently rejected from the best fossil fuel-fired plants. Fortunately, the program for nuclear electrical power sources for space applications has nicely validated the use of lithium in niobium at temperatures up to 1200°C.⁵ Further, work for the Air Force at Battelle on structures suitable for hypersonic aircraft has led to the rolling of niobium sheets in 3 ft widths and the fabrication of these sheets into shell structures suitable for aircraft wings and fuselages.⁶ As a consequence, the feasibility of fabricating niobium into the type of structure shown in Fig. 2 has been demonstrated, although it is clear that structures of this type will be expensive and will require stringent quality control measures.

Another major problem associated with the blanket region is radiation damage to the vacuum wall.³ This problem is even more severe than the radiation damage problem in a fast breeder reactor because in this instance we would like to have the vacuum wall last for 20 years to avoid replacing large radioactive parts, whereas in the fast breeder reactor the relatively small fuel elements would be replaced at intervals of a year or less. The extensive radiation damage work currently underway in the Fast Breeder Reactor program will prove enormously helpful to the fusion reactor effort, but additional work will be required because the initial neutron energy in fission reactors is only 1 or 2 MeV whereas in fusion reactors it is about 14 MeV. This may induce radiation damage mechanisms that we have not yet been able to envision.

Power Conversion System

There appear to be a number of reasonably straightforward ways in which the heat energy developed in the blanket region can be employed to produce

electricity. If the blanket region is made of stainless steel and cooled with lithium at about 500°C , it would be possible to raise steam at perhaps 450°C to drive a conventional steam power plant as indicated in Fig. 3, and this should give an overall thermal efficiency about equal to the 40% of that for the best fossil fuel plants. The steam generator could be similar to that for an LMFR, and the rest of the steam plant would be conventional. A second approach would be to employ the heat in a gas turbine cycle, in which case there would be a strong incentive to make use of a high temperature blanket region to obtain a good thermal efficiency. Commercial gas turbines are now operating with turbine inlet temperatures of around 1700°F . With niobium as the structural material in the blanket and a 1700°F turbine inlet temperature, it should be possible to obtain an overall thermal efficiency for the gas turbine plant within a few percentage points of that obtainable with current fossil fuel-fired steam plants. Figure 4 shows a schematic diagram for a gas turbine plant of this type.⁷

A third and still more attractive approach, although novel, would be to employ a potassium-steam binary vapor cycle.⁸ With this system it would be possible to get an overall thermodynamic cycle efficiency of around 58% as opposed to the 40% thermal efficiency of current fossil fuel-fired plants. This would reduce the waste heat rejection per kilowatt of electricity by more than a factor of 2, and would reduce the fuel consumption by about 30% as compared to a conventional 1000°F steam cycle. Figure 5 shows a schematic diagram of such a power plant. Note that this type of plant also shows promise for obtaining a high thermal efficiency, reduced fuel consumption, and reduced heat rejection for both fission reactor and fossil fuel-fired power plants.^{9,10}

Limitations on Tritium Concentration

The most obvious limitation on tritium concentration is that imposed by the cost of the tritium for the initial inventory in starting up a reactor. A firm price is not available for tritium, but an estimate can be made assuming that it could be generated in central station reactors interchangeably

Fig. 3. Flow diagram for a steam power conversion system with an intermediate NaK loop for tritium removal.

Fig. 4. Flow Sheet Showing Pressures and Temperatures in a Helium Gas-Turbine Cycle Coupled to a Theronuclear Reactor

Fig. 5. Flow diagram for the potassium-steam binary-vapor-cycle power conversion system.

with plutonium. Assuming that the main element in the cost would be neutrons, the costs for the two materials would be inversely proportional to the atomic weight. Thus, the current market price for central station reactor plutonium of about \$10/g implies a tritium price of about \$800/g, or about \$1,000,000/kg. To be conservative one can assume that special charges will increase this price to roughly \$2,000,000/kg. If the amount of tritium to be employed in a mission were to be 1 kg, the cost on this basis would run about \$2,000,000. However, if a thermonuclear reactor economy is once built up, it is likely to be capable of a breeding ratio of around 1.3 to 1.5, and this could yield a surfeit of tritium. It is difficult to predict what the cost of tritium would be under those conditions, but it seems likely that it will drop to perhaps \$20,000/kg. As will be shown later, other considerations make it vital to keep the tritium inventory to a low level, and hence the doubling time becomes only a matter of a few months.

A second important consideration in estimating the permissible inventory of tritium is imposed by concern for hydrogen embrittlement of the structural material. For niobium it appears that the tritium concentration in lithium should be kept below about 100 ppm in order to avoid difficulty with embrittlement of the niobium. Inasmuch as a typical full-scale thermonuclear reactor¹¹ will require a lithium inventory of the order of 400,000 kg, this would imply a tritium inventory in the lithium of around 20 kg, or a capital investment of the order of \$44,000,000, an unpleasantly high level.

A third and particularly difficult set of requirements is imposed by environmental considerations. The ease with which tritium can diffuse through metal walls make it difficult to contain. Studies of reactor safety problems likely to be posed by full-scale thermonuclear reactors indicate that by far and away the most difficult is that posed by tritium.^{12,13} These studies indicate that there is an extremely strong incentive to keep the tritium concentration in the lithium blanket to something of the order of 1 ppm. If this can be done, it reduces the capital investment in tritium for the initial startup to a minor level and provides a large margin with respect to any possible difficulties from hydrogen embrittlement of the structural metal.

Some notion of the difficulty associated with efforts to design a system that will not permit tritium to leak into the surroundings at a serious rate is given by Fig. 6 which shows the tritium permeation rate through 1000 m² of 1 mm thick of typical metals and alloys as a function of temperature. These curves have been plotted for a partial pressure on the liquid metal side equivalent to that given by a tritium concentration of only 1 ppm in lithium. The value of 1000 m² of metal wall was chosen because it is of the same order as the surface area of the torus containing the plasma. The heat transfer surface required to transmit the heat from the lithium to a working fluid is of the order of 10 to 100 times higher, thus making the heat exchanger the principal "window" through which the tritium will tend to diffuse.

Tritium Recovery System

The tritium generated in the lithium blanket as a consequence of neutron absorptions in the lithium might be removed by any of quite a number of different techniques. Obvious approaches include fractional crystallization or fractional distillation, but studies have shown that neither of these approaches will make possible the 1 ppm concentration desired. However, one can make a virtue of necessity, take advantage of the high permeation rate of the hydrogen into the heat exchanger, and set about removing the tritium from the secondary fluid circuit. This, of course, is not practicable if water is employed because then an isotope separation process would be required and this would be enormously expensive because it would be necessary to reduce the tritium concentration to a much lower level than the deuterium concentration in sea water. Obviously, a much better approach is to make use of a secondary fluid that does not contain any hydrogen, i.e., helium or potassium. It is for this reason that the choice of the power conversion system has a very important effect on the choice of the tritium removal system.

If one examines the helium power conversion system, it immediately becomes evident that one might include activated alumina dryers in a bypass circuit. One might depend on the presence of a small amount of oxygen in the helium to react with the hydrogen as rapidly as it diffused through the heat

Fig. 6. Effects of temperature on the estimated permeation rate of tritium at a partial pressure of 10^{-7} torr through 1 mm thick sheets of $10,000 \text{ m}^2$ of metals and alloys. The estimates were made by extrapolating from the limited data given in Ref. 21. Experimental data at low hydrogen partial pressures are badly needed to determine the validity of these estimates.

exchanger walls into the helium. The resulting moisture would then be removed in the aluminum oxide dryer. It would also be possible to include a copper oxide bed in the bypass stream ahead of the activated aluminum dryer. This is a better approach because niobium in the temperature range above about 800°C is very susceptible to attack by oxygen, a point that will be considered in detail in a companion paper. This problem appears so formidable, in fact, that in the writer's judgment it would be better to make use of the potassium vapor cycle of Fig. 5. This has the advantage that it will yield a substantially higher thermal efficiency than can be obtained with the helium gas turbine system, and the materials problems appear to be much more manageable.

The system envisioned is indicated in Fig. 7. About 99% of the tritium would diffuse from the lithium into the potassium boiler. A review of the literature, particularly Ref. 14, indicated that cold trapping tritium from the potassium would be a good basis for the design of a tritium removal system. This stems from the fact that the solubility of tritium in potassium falls off rapidly as the temperature is reduced, so that potassium tritide can be removed with a cold trap and filter system. Even better performance can be obtained by an AEC patented process that involves adding a small amount of lithium to the potassium.¹⁵ This is very effective because of both the much stronger affinity of hydrogen for lithium and the lower solubility of the lithium tritide in potassium.^{14,15}

In view of the above, a tritium removal system was designed to remove tritium from the potassium Rankine cycle system of Fig. 5 by cold trapping LiT. Inasmuch as there is a substantial amount of data available on normal ~~hydrogen in potassium and lithium~~ but very little on tritium in potassium, the calculations were carried out first for normal hydrogen and, subsequently, corrections were applied to convert the results to the tritium case.

When making the analysis no comprehensive set of data could be found in the literature for the concentration of KH in potassium as a function of both temperature and hydrogen overpressure, hence the data presented in Ref. 14 for 22% Na-78% K were employed. The limited data available^{17,18} indicate

Fig. 7. Cold trap system for removing tritium from a controlled fusion reactor.

that the solubility of KH in potassium is about the same as NaH in Na, hence use of the data for NaK should give an acceptable approximation for present purposes. Extrapolation of Fig. 6 on p. 167 of Ref. 14 indicates that the hydrogen concentration in the potassium would be 0.001 ppm at 80°C (the melting point of potassium is 62.5°C).

The controlling consideration in the design of the tritium recovery system is maintaining a tritium permeation rate into the steam system of the order of only a few curies per day, that is, the equivalent of only a few cubic centimeters per day at standard pressure and temperature. To achieve such a low permeation rate it is necessary to introduce a more effective diffusion barrier than that represented by the stainless steel walls in the potassium condenser-steam generator. One way of doing this would be to apply a coating of tungsten to the potassium side of these tubes. Another approach would be to employ a coating of either gold or a ceramic on the steam side of the tubes. It would be advantageous to use gold because the technology for plating it is on much firmer ground, but the ceramic coating appears to be not only much less expensive, but, at least potentially, it may also be substantially more effective at the very low partial pressure of tritium envisioned here. This stems from the fact that the principal obstacle to permeation through the ceramic layer will be the dissociation of the tritium at the surface of the ceramic, and when this becomes the rate controlling step, the permeation rate should vary linearly with the partial pressure instead of as the square root. If transition to this as the rate controlling step occurs at a partial pressure substantially above that maintainable with the cold trap in the potassium system, the permeation rate into the steam might be kept to a sufficiently low level from the standpoint of environmental considerations to permit a substantially higher tritium concentration in the potassium and lithium than envisioned in the flow sheet of Fig. 7.

The bulk of the tritium flowing through the plant will be handled by the above system. However, it is also necessary to employ some additional auxiliary pieces of equipment to recover the tritium which diffuses through hot walls into the high vacuum system and appears in the discharge from the

high vacuum system pumps. One must also expect some tritium to leak into the atmosphere of the room containing the tritium handling equipment, and this in turn means that the moisture should be removed from the ventilating air supplied to the room before it reaches the room, and any tritium that leaks into the room air should be oxidized to form water vapor and the water vapor should be removed with a dryer prior to discharging the air from the room. It may be that the room can be ventilated in a routine fashion under normal conditions if the tritium leakage rate into the room air can be kept sufficiently low, but provisions must be made in any case to cope with a tritium leak. If such a leak occurs, the room air then must be processed to remove moisture that would include the tritium.

There will also be tritium leakage from the potassium system into the argon buffer gas system for the potassium turbine shaft seals. In addition, one must expect some tritium leakage from the stainless steel casing of the potassium turbine into the enclosure surrounding it, and this tritium must also be recovered.

The tritium and deuterium being circulated through the reactor will become contaminated by normal hydrogen because about one atom of normal hydrogen is formed per 60 fusion reactions. Thus, an isotope separation system is required to keep the concentration of normal hydrogen in the deuterium-tritium mixture to about 1%. Of course, operating experience may disclose that somewhat higher concentrations of normal hydrogen can be permitted, but, for purposes of this design study, the 1% level was assumed. It was further assumed that a still for separating normal hydrogen from deuterium and tritium would be employed, and that the tritium-rich stream coming out of the column will contain only 1% normal hydrogen, while the tritium-lean stream coming out the other end of the column would have a tritium concentration of about 1%. The latter stream would be fed to a bed of calcium oxide to tie the tritium up as CaOH . This material would have to be sequestered, but its volume would be sufficiently small so that a 20 year accumulation from the reactor would still represent a rather modest inventory, i.e., about 20 tons of CaOH . If desired, the entire quantity could be kept in the tritium system equipment room, and this has been assumed for the plant layout of Fig. 8.

A schematic diagram showing the relation of the various components in the tritium system is shown in Fig. 9. No attempt has been made to indicate such refinements as the point at which the various streams containing mixtures of normal hydrogen and tritium would be fed into the isotope separation column. In fact, it will probably be necessary to gain some operating experience before decisions of this character can be made.

It should be noted that all of the walls containing the high vacuum system will be relatively cold, i.e., below 200°C, and hence tritium diffusion through these walls should be extremely low. Thus, tritium contamination of the atmosphere in the reactor cell or in the equipment rooms should be very low indeed. To achieve this it will, of course, be necessary to make use of all-welded joints throughout the various elements of the tritium system except for a few places where maintenance requirements make it virtually essential to use a bolted flange. Such cases must be kept to an absolute minimum.

Back Diffusion of Hydrogen from the Steam System

The difficulties posed by the diffusion of tritium into the steam system raise questions as to the extent to which back diffusion of hydrogen from the steam system might occur and lead to dilution of the tritium by normal hydrogen. This could occur as a consequence of oxidation of the metal in the steam generator tube walls by steam, release of hydrogen, and diffusion of the hydrogen through the tube wall into the potassium system. It has been shown that the corrosion rate by steam inside of a steel boiler tube can be measured quantitatively by determining the rate of hydrogen effusion from the outside of the boiler tube.¹⁹ Data presented in Ref. 12 indicate that, for a system having a boiler tube matrix of the proportions considered here, the back diffusion of normal hydrogen through plain stainless steel tubes would amount to 1.5 kg/yr at a steam temperature of 600°F. In searching for data at the higher temperature of interest in this study, it was found in Ref. 20 that the corrosion rate for type 316 stainless steel amounts to ~0.0003 in./yr. This corresponds to a hydrogen generation rate of ~40 kg/yr so that the

hydrogen in-leakage to the potassium system could amount on an annual basis to about 25% of the tritium production.

Use of a tungsten coating on the potassium side of the tube walls would inhibit this back diffusion to a substantially lower level, but it is difficult to estimate just how great the reduction might be. However, even without taking credit for this possibility, it appears that coping with the dilution would not be too expensive because an isotope separation process is required in any event, as cited above.

If a ceramic coating were applied to the steam side of the tubes in the potassium condenser-steam boiler, both the corrosion rate and the back diffusion rate of hydrogen should be reduced drastically. Thus, it would be desirable to apply such a coating not only to reduce the tritium leakage rate into the steam system but also to reduce the dilution of the main tritium inventory by normal hydrogen.

Uncertainties in the Analysis

In reviewing the above analysis in an effort to appraise the uncertainties, a number of points become evident. The calculations are conservative in that they were based on permeation coefficients obtained with metal tube walls completely free of oxide films; the oxide film on the steam side of the boiler tubes would certainly act to reduce the tritium diffusion rate. This would be even more likely to be the case if the steam side of the tubes were coated with a thin but adherent and nonporous film of a ceramic such as aluminum or titanium oxide. Under any circumstances, new experimental data are badly needed for permeation of hydrogen through niobium and through stainless steel having a thin, dense, pore-free layer of oxide at hydrogen partial pressures of as low as 10^{-7} mm. Experimental data are also badly needed on the volume of cold trap required as a function of temperature, and LiT concentration in potassium. It may be necessary to use some other material such as zirconium or yttrium to getter the tritium in order to avoid an excessive volume of potassium in the cold trap.²²

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