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TRITIUM-RELATED MATERIALS PROBLEMS IN FUSION REACTORS

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TRITIUM-RELATED MATERIALS PROBLEMS IN FUSION REACTORS*

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I. INTRODUCTION

Our goal here is to describe the pressing materials problems that must be solved before tritium can be used to produce energy economically. Unlike most of the other materials problems that have been discussed in the previous lectures of this series, the tritium problem is intimately connected with on-site chemical processing. Therefore, to fully appreciate it, one must have some understanding both of tritium and its peculiarities and

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of the chemical processing requirements of a thermonuclear fusion power plant.

Tritium and deuterium are heavy isotopes of hydrogen that can undergo nuclear fusion with relative ease. This process is expressed by the reaction:

Tritium is a necessary fuel in thermonuclear reactors and is therefore at the heart of all fuel-cycle problems.

Because tritium has a half-life of only 12.3 years (1), it is not found naturally in useful concentrations and must, therefore, be bred, usually in nuclear reactors. This is expensive (about \$10,000/g), but the cost is not firm because new production reactors have not been built for decades.

Tritium is a hazard to man and the environment. Although the biological effectiveness of its radiation is generally taken as unity (the same as gamma rays), tritium is believed to be three times as toxic as gamma rays in chronic, low-level doses (2). Tritium is absorbed easily through the lungs or skin. The maximum permissible concentration (mpc) in air is about one part per trillion. A lethal dose may be received from a single breath of tritiated water vapor or from a single drop of tritiated liquid on the skin. The biochemical effects of tritium have been studied extensively (3) and are fairly well understood.

The chemical behavior of tritium is nearly identical to that of normal hydrogen: it burns, explodes, and can be incorporated into biological molecules; it permeates and embrittles metals as hydrogen does. Embrittlement may also be caused by helium produced in the radioactive decay of tritium (4).

BREEDING TRITIUM

Tritium is usually bred in nuclear reactors using lithium as the breeding material. Lithium can absorb a neutron and produce tritium and helium:

Fusion reaction (1) produces a neutron that can be used to create more tritium via reaction (2). Other important breeder reactions include that of ${}^7\text{Li}$,

and the neutron-multiplying reaction of beryllium,

The product neutrons in reactions (3) and (4) can also be used to breed tritium via reaction (2).

Several classes of breeding materials have been considered:

- pure lithium metal with isotopic enrichments
- lithium alloys, including intermetallic compounds (LiAl , Li_4Sn , and Li_7Pb_2)
- molten salts (Li_2BeF_4 and $\text{LiNO}_3/\text{LiNO}_2$ eutectic)
- solid ceramics (LiAlO_2 , Li_2O , Li_2C_2 , Li_3N , LiD , and $\text{Li}_2\text{Be}_2\text{O}_3$)

These materials may be used singly or in combination, with or without neutron multipliers and reflectors. Their effectiveness depends on the design of the reactor. In general, as atoms other than neutron multipliers are added to lithium to form compounds or

alloys, the breeding ability of the combination decreases. Thus, LiAl is a poorer breeder than pure lithium, which in turn is poorer than Li_2Pb_7 , where lead is a neutron multiplier.

The breeding material is contained in a blanket layer just outside the innermost wall of the reactor. Most of the neutrons produced are absorbed in the blanket; it is here that tritium is produced and that neutron energy is deposited. This general scheme is shown in Fig. 1.

FIG. 1. Schematic diagram of net tritium breeding in a fusion power plant.

RECOVERING BRED TRITIUM

The various breeding materials show vastly different behaviors with respect to the release of tritium. The rate of tritium recovery from the breeding material usually depends on the concentration of tritium in the blanket. The concentration must climb to its steady-state value before recovery can proceed at a steady rate. This behavior is shown graphically in Fig. 2, where the shaded area gives the required start-up inventory. This is the

FIG. 2. Graphic representation of tritium start-up-inventory requirements as determined by recovery processing.

inventory required to keep the plant going until the recovery rate equals the burn rate, i.e., the break-even rate.

Assuming that the rate of recovery of tritium from the blanket is directly proportional to the tritium concentration in the breeding material, it can be shown that the ratio of break-even inventory I_{be} to steady-state inventory $C_{ss}V$ is given by

$$\frac{I_{be}}{C_{ss}V} = 1 - \left\{ \left(1 - \frac{1}{\alpha} \right) \left[1 - \ln \left(1 - \frac{1}{\alpha} \right) \right] \right\}, \quad (5)$$

where α is the breeding ratio (5). Evaluated numerically for different values of α , this expression gives the data shown in Table I.

TABLE I.

TRITIUM INVENTORIES AS A FUNCTION OF BREEDING RATIO

α	$I_{be}/C_{ss}V$
1.01	0.944
1.05	0.807
1.10	0.691
1.20	0.535
1.30	0.431
1.40	0.356

We can see that if it is important for a plant to become very quickly self-sufficient in tritium, the blanket materials chosen should have neutronic properties that allow high breeding ratios. In general, this means avoidance of non-breeder materials that capture neutrons, and inclusion of neutron multipliers such as Be or Pb. The required break-even inventory is further reduced by choosing materials that have a low steady-state tritium concentration C_{ss} and by minimizing the volume V of breeding material. This requires the use of solid materials that show low (but not zero) solubility and high diffusivity for tritium. The solubility should not be zero because in a radiation field, all solids develop internal voids that would then act as permanent sinks for tritium. In general, these relationships between solubility, diffusivity, and radiation stability in solid breeding materials are unknown. Breeding materials are expected

to be in powder or liquid form rather than in massive solid form so that processing kinetics will be fast.

There are three major means of recovering tritium:

- permeation,
- trapping by adsorption, and
- trapping by absorption.

Molten salt extraction from liquid lithium followed by gas sparging is being investigated and may also be feasible.

Permeation

In the permeation recovery process, tritium, in liquid-lithium, high-pressure-helium, or potassium-vapor coolants, contacts one side of a selectively permeable membrane, such as niobium. The popular Nb-1% Zr alloy might also be used as a membrane material, but due to the strong chemical affinity of Zr for tritium, it could show premature embrittlement from helium. Other possible permeation membranes are V, Ta, and Pd. On the other side of the membrane, the tritium partial pressure is lower so the tritium permeates the membrane. The rate of this continuous process is usually $R_1 = K_1 (P_1^{1/2} - P_2^{1/2})$, where K_1 is a constant and P_1 and P_2 are the tritium partial pressures on the two sides of the membrane. Ample evidence (6) shows that when the permeation rate is surface-limited, it is better described by $R_1 = K_2 (P_1 - P_2)$. In either case, as shown in Fig. 3, the recovery rate increases as P_1 (or the blanket tritium inventory) increases. As just described, it is zero at start-up.

At one time, it was suggested that adding an excess of normal hydrogen to the very dilute tritium stream might increase the recovery rate by some kind of sweeping action during the permeation process, but this has since been shown to be a hindrance rather than a help (7).

FIG. 3. Schematic diagram of a permeation-type process for recovery of tritium from a He or Li coolant stream.

Absorption

As shown in Fig. 4, the trapping processes are generally cyclical and require regeneration of the trap absorbents and adsorbents. Molecular tritium can be absorbed from a vacuum or from flowing helium or lithium streams if they come into contact with certain metals (typically cerium, scandium, or titanium) to form a stable hydride. The dissociation pressures of a few of the more stable binary hydrides are shown in Fig. 5. Uranium has been shown to be capable of removing 99.9% of the D_2 from a flowing stream of argon containing 1% D_2 , but this is not enough for T_2 recovery in a breeding cycle recovery process. On the basis of chemical reactivity, price, and dissociation pressures at both high and low temperatures, cerium has been chosen as a promising regenerable getter (tritium absorber) (8). Some ternary hydrides might be as good or better, but our knowledge of these more complex systems is very scanty in comparison. Long-term poisoning effects need to be studied.

FIG. 4. Schematic diagram of a trapping-type process for recovery of tritium from a He stream.

FIG. 5. Equilibrium hydrogen pressure over some substoichiometric binary metal hydrides as a function of temperature.

Adsorption

Tritium or tritiated vapors (e.g., tritiated water vapor) can be removed from flowing gases by selective

adsorbents: molecular sieves, silica gel, or activated charcoal. These adsorbents are particularly effective when operated at low temperatures, but recovery processes capable of operating at high temperatures are usually preferred for reasons of thermodynamic efficiency. This is particularly true if the reactor coolant stream functions as part of the recovery cycle and if a significant fraction of that stream must be processed to supply the needed tritium.

In both trapping processes, first-order kinetics require the recovery rate R_2 to be proportional to $(P_1 - P_e)$, where P_1 is the partial pressure of the tritium-bearing species in the gas and P_e is the equilibrium partial pressure of the species being trapped at that particular temperature. Again, the recovery rate depends strongly on P_1 , which is a principal factor in determining the blanket's tritium inventory.

Recovery of tritium from liquid lithium coolant is expected to be difficult when the lithium concentration is kept below 10 ppm by weight. For this concentration, the blanket inventory could be about 5 kg (\$50,000,000), but as shown in Fig. 6, the tritium partial pressure available to drive a separation process would be only 10^{-6} Torr at 1000 K. Thus, large processing equipment might be required.

Figure 7 shows that adding aluminum to the lithium substantially raises the partial pressure. Tritium has been produced in this alloy for defense purposes for many years (9,10). The phase diagram for the system (Fig. 8) clearly shows that these intermetallic compounds, LiAl and Li_2Al , probably could not be used as solid powders because of sintering effects at several hundred degrees Celsius.

An encouraging recent development is the renewed activity with LiAlO_2 powders. The tritium-breeding reaction is

FIG. 8. Graphic representation of the pressure-composition-temperature relationship for the hydrogen-lithium system in the dilute range.

Tritium and tritiated water show a low solubility in the solid powder. Particles given the equivalent of a 1-h exposure to the neutron flux of a fusion reactor have released tritium in a mean time of 4.3 h at 600°C (10,11). It appears on the basis of recent work, that at high temperatures, tritium blanket inventories could be kept to a few hundred grams in a large power plant using the oxide reaction. There are still a few possible problems and unanswered questions, however.

The high temperatures required to release the tritiated water quickly from LiAlO_2 may make the use of refractory metals or carbides in structural members necessary, but these materials are chemically attacked by water vapor at high temperatures. Our current plan in response to this water vapor problem is to construct the blanket from lithium beryllate (12) and excess

FIG. 7. The influence of alloying lithium with 50% aluminum on the equilibrium partial pressure of hydrogen in the two-phase (high hydrogen content) region.

FIG. 8. The lithium-aluminum phase diagram.

beryllium as a partially sintered mix of powders. The breeding reaction would then have the following chemical result:

The excess beryllium would effectively scavenge the oxygen freed in the breeding reaction, would be compatible with the breeding compound, and would act as a neutron and energy multiplier in the blanket.

The availability of beryllium for this blanket design is a big question. Without further exploration and discovery, current supplies could soon be exhausted (13). Also, continuous irradiation of either material ($\text{Li}_2\text{Be}_2\text{O}_3$ or Be) may induce swelling and internal void formation, which could permanently tie up a much larger amount of tritium than presently expected. The presence of Be could aggravate this behavior due to its high (n,α) cross section. Nothing has been done experimentally on this particular problem. In general, these problems on breeding and recovery are common to all fusion reactor concepts and are independent of the plasma confinement concept.

CONTAINING TRITIUM

Environmental control of tritium in fusion reactors presents a challenging set of problems. As we have noted above, tritium behaves just like hydrogen: it permeates metals and plastics, it readily becomes airborne, and once mixed with normal water, it cannot be conveniently separated. These potential escape routes are shown in Fig. 9.

FIG. 9. Schematic drawing of a fusion power plant showing tritium escape routes.

Permeation Losses

The high tritium concentrations or partial pressures that provide the high recovery rates desired from the breeding blanket may also cause high rates of permeation loss to the environment. For example, the reference design of the theta-pinch reactor calls for a liquid-lithium breeding blanket. To keep permeation losses to less than 30 Ci/day, the partial pressure of tritium has to be no more than 10^{-10} Torr above the lithium (14). This is an incredibly low partial pressure to maintain for a product in an industrial situation, and to date, no way of doing this has been found.

An obvious way to reduce some permeation losses is to use hydrogen-impermeable heat exchangers. This can be done by dropping the coolant operating temperature (and efficiency), by adding additional heat exchange loops, or by adding new materials to the heat exchangers. Some of these new materials are barrier metals (copper, tungsten) (15), barrier oxides (oxidation films,

BeO (7,16), and metal getters (titanium, cerium), and, as shown in Fig. 10, all might be placed into laminated (triplex) tubing to reduce or eliminate permeation losses. The virtue of oxide barriers is that, as shown in Fig. 11, they are far less permeable than the least permeable of metals (tungsten). However, they suffer from the disadvantage of being difficult to fabricate. This is especially true if pore-free thin films are required as the intermediate layer.

FIG. 10. Schematic wall cross section of a possible triplex heat-exchanger tube designed to reduce tritium escape via the permeation mechanism.

Liquid Loss

High-level tritium in waste water is usually immobilized by reacting it to form concrete, which can then be encapsulated and buried (17). In the future, it may be possible to remove the HTO from waste water selectively by laser isotope-

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FIG. 11. The relative permeabilities of stainless steel, tungsten, and alumina.

separation techniques and thus greatly reduce the amount of solid waste to be buried. Progress to date has been slight however (18,19).

Gaseous Loss

Tritium in gaseous wastes can be reduced to low levels with trapping similar to those used for recovering tritium from the breeding blanket. Tritiated hydrocarbons may be adsorbed directly or oxidized to trap the tritium as tritiated water on

molecular sieves. This oxidation would normally be done at a few hundred degrees Celsius in an oxygen-rich stream flowing over precious-metal catalysts. The gaseous species HT, DT, and T₂ can be trapped directly by cryogenic absorption or adsorption. Usually, however, they are trace contaminants in air and can therefore be catalytically converted to water or reacted with certain hot transition metal oxides (CuO, MnO₂) to form water. Tritiated water is then trapped on molecular sieves at or below room temperature. The removed fraction of tritiated water can be increased by deliberately swamping the gas flow with water vapor after the first trapping, but this adds greatly to the liquid-waste disposal problem. The general scheme is shown in Fig. 12.

FIG. 12. Schematic diagram of a system using catalytic oxidation for removing tritium from other gases.

Two new state-of-the-art gaseous-effluent control systems have been built in the U.S. recently. They both employ catalytic oxidation, followed by adsorption of tritiated water on a molecular sieve (20,21). To date, they have not seen enough operation to demonstrate satisfactory long-term performance, but they certainly represent our present best hopes for keeping the gaseous release of tritium and its compounds to an acceptable minimum. If these systems do not perform up to expectations, much additional work on process design, catalyst development, and molecular sieve development will be required. Again, these containment problems are common to all fusion reactor concepts.

FUEL RECYCLING

Recycling the fuel in a fusion reactor is necessary because most of it does not react before it is exhausted from the plasma region. Fortunately, suitable materials and standards of construction exist for every part of the fuel-recycling system where large amounts of tritium must be handled at relatively high pressures. Other parts of the fuel cycle need substantial additional materials research and development before fusion power can become a reality.

Several compounds must be removed from the fuel before it can be reused:

- helium ash
- hydrogen from the D(T,H)T reaction [which is present in smaller quantities than in the normal D(T,n)He reaction]
- methanes
- traces of air

The fuel-recycling loop is fairly simple and is represented in Fig. 13.

Recycling problems depend more on the reactor concept than do other problems discussed so far. Mirror reactors are steady-state devices with continuous neutral beam injection and exhaust, and neutral injection requires separate deuterium and tritium streams. Tokamaks operate in a pulsed mode (about a 1-min cycle) with neutral beam or pellet injection and with between-cycle exhaust. The theta-pinch reactor is short-pulsed (a few seconds per pulse), and mixed isotopes in the fuel supply are desirable. Finally, laser-driven microexplosions are very short pulses (milliseconds), and several pulses per second are expected. All the reactors operate under high-vacuum conditions. In a mirror reactor, the fuel exhausts directly to an electrical direct convertor that recovers most of the kinetic energy of the escaping particles.

Neutral beam injection allows preheated (high-energy) particles to be fed into the plasma across magnetic field

FIG. 13. Simplified schematic diagram of the fuel-recycling loop on a fusion reactor.

lines. Some necessary charge-exchange reactions are velocity-dependent rather than energy-dependent, so isotopic purity increases the efficiency of neutral beam injection. The separation of deuterium and tritium (along with the removal of hydrogen) will undoubtedly be accomplished by means of cryogenic distillation, an existing technology. Purely physical operations, such as distillation, must be supplemented by chemical methods to break down the DT, HD, and HT molecules as in Fig. 14. This is routinely done now,

FIG. 14. Diagram of a cryogenic distillation column with a catalytic disproportionator.

but efficiency should be improved. In particular, it would be worthwhile to study the effect of the tritium radiation field on the kinetics of re-equilibration in a solution of partially separated diatomic molecules. Development of catalysts to promote the exchange reactions at liquid-hydrogen temperatures would further reduce costs and tritium inventory requirements.

Carbon First Wall

The innermost reactor wall may be lined on the plasma side with a graphite curtain to reduce neutron-radiation damage and to lower the atomic number of materials sputtered from the first wall (22). In mirror reactors, the direct-converter collection electrodes may also be made of graphite because of its high-temperature properties. Energetic particles escaping from the plasma will be implanted in the curtain materials. A certain amount of the implanted tritium will be lost from the fuel stream because of chemical interaction with the curtain materials, and this poses another unexplored inventory problem. Very crude calculations suggest the possibility of permanently trapping tens of kilograms of tritium in graphite components.

Methane Formation

In low-pressure plumbing systems, tritium reacts with hydrocarbons and other carbon compounds to form methane, by an unknown, radiolytic mechanism. This is shown in Fig. 15 on the basis of data recently taken by G. Morris at the Lawrence Livermore Laboratory (LLL). The presence of methane in the fuel supply is undesirable because it can plug the cryogenic still needed for hydrogen isotope separation and can introduce impurities into the plasma.

Some insight into the mechanisms involved may be obtained from the recent research of Balooch and Olander (23), showing that hydrogen is added to C, CH, CH₂, and CH₃ free radicals on the surface of graphite at 506 K. Some of their experimental data and theory are shown in Fig. 16 and Eq. (8). The theory is obtained from an approximate solution to the time-dependent non-

FIG. 15. Data showing the synthesis of tritiated methanes in relatively clean stainless-steel tritium-filled vessels: (1) 25°C, drawn tubing, freon washed; (2) 100°C, drawn tubing, vapor degreased; (3) 25°C, machined surface, vapor degreased, vacuum baked at 100°C for 24 h; (4) 100°C, drawn tubing, vapor degreased.

linear differential equation describing the interactions of a chopped molecular beam of hydrogen atoms with the surface.

$$\epsilon_{CH_4} = \frac{3K_1K_2k_3\eta(C_0/H)^2 \sin\phi_{CH_4}}{\omega + (H^2D/2)^{1/2}\omega^{1/2}}, \quad (8)$$

where:

ϵ = the apparent reaction probability = $[CH_4]/[H_2]$ at the detector,

K_1 = the equilibrium constant for H, C, and CH on the surface,

K_2 = the equilibrium constant for H, CH, and CH_2 on the surface,

FIG. 16. Data and theory of methane formation by reaction of graphite with chopped atomic hydrogen beam, showing the variation of methane reaction probability with beam intensity. The different symbols for the prism plane represent replicating runs.

- k_3 = the rate constant for $CH_2(ads) + H(ads) \rightarrow CH_3(ads)$,
 η = the sticking probability of H atoms in the beam to the surface,
 C_0 = the bulk concentration of H atoms just below the surface,
 H = the solubility coefficient relating C_0 to the surface concentration,
 ϕ = the methane phase lag relative to the hydrogen at the detector,
 ω = the modulation frequency of the beam, and

D = the diffusion coefficient of hydrogen atoms in the bulk graphite.

In the case of tritium in a fuel system, each decay produces a large number of dissociations in the gas phase, and also an uncombined free tritium atom that remains from the original T_2 molecule. These free atoms may react in much the same fashion as those in the molecular beam experiment. Whether additional methane will be formed by energetic implanted tritons or deuterons reacting with graphite is unknown.

LASER-FUSION FUELING

The materials problems associated with safety and containment in laser fusion are identical to those for CTR. Likewise, the requirement to breed an excess of tritium persists, and since the energy-producing reactions are the same, the motivation is still strong to operate the breeding blanket at a high temperature so that it may be a part of a thermal conversion cycle. The fuel-cycle problems, however, are a bit different, and as one might expect, they involve materials problems.

The conceptual designs of laser-fusion power plants are in a very primitive stage compared to those using magnetically-confined plasmas. In general, however, the fueling scheme preferred (for reasons of simplicity) is the injection of frozen DT pellets at a high repetitious rate (10 to 100 Hz). Other parts of the laser-fusion fuel cycle would be much the same as those already described for magnetic-confinement systems.

Cryogenic Targets

Heating and compressing solid pellets of DT to fusion conditions requires lasers of enormous power. The laser requirements are relaxed by a significant factor if the frozen DT is in the form of a spherical hollow shell (24). Liquid hydrogen droplets with internal cavities have recently been produced (25), but the sphericity and concentricity of the internal and external surfaces were inadequate for use as laser-fusion targets. Improvements on the existing techniques and development of other methods would be of very great interest.

Remarkably, most of the cryogenic properties of DT are unknown. Reasonable estimates have been made (26), but experimental verification has not yet been provided. Although we speak generally of DT, in any real DT system, three different molecular species (D_2 , DT, and T_2) actually co-exist. Each species shows different physical properties because the isotope effect is quite large. It is this difference that forms the basis of isotopic separation by cryogenic distillation. Figure 17 contains the simplest possible ternary phase diagram for DT. It assumes regular solution behavior between species and phases and ignores complications that could arise from the varying amounts of ortho and para D_2 and T_2 that might be present. As can be seen, liquid that freezes will change composition, and depending on the kinetics of the phase change, spatial segregation of the molecular species could occur during formation of a pellet.

Current Targets

Present targets are mostly microspheres filled with high-pressure DT gas. Curiously enough, more goal-directed

FIG. 17. Estimated ternary phase diagram for the DT system in the vicinity of the phase boundaries.

effort probably has been expended on these relatively simple targets than on all the other materials-related tritium problems confronting us in CTR with magnetic confinement. A photograph of typical DT-filled microspheres in an interference microscope is shown in Fig. 18. Those that are found to have the proper size, wall thickness, and degree of perfection are mounted for subsequent testing in the laser facility. A mounted target is shown in Fig. 19. Only about 1 microsphere out of 10,000,000 can pass the rigorous selection tests and go on to be shot in the laser (27).

FIG. 18. Photograph of DT-filled glass microspheres under an interference microscope. Only one shows acceptable sphericity.

Development of techniques to give a higher survival rate would be of obvious benefit.

Pressurization is accomplished by heating the microspheres in a suitable pressurized vessel so that DT dissolves in and diffuses through the glass until the internal and external pressures are equal. Cooling under pressure traps the gas inside the microsphere. As soon as the external pressure is released, the trapped gas begins to escape slowly by diffusion. The rate is reduced by storage in a refrigerated location, but the exasperating aspect of this off-gassing is that it varies by as much as a factor of 100 from target and is predictable only on a statistical basis (28).

Figures 20 and 21 show two well-established phenomena. The first is that the outgassing transient for a batch

FIG. 19. A gas-filled microsphere mounted on a 5- μ m stalk for further experimentation.

of spheres is non-exponential. (The reasons for this are not well understood.) The second is the variability between targets, which is believed to result from differences in glass composition. Long-term storage of surplus targets for later experiments is of questionable practicality because nondestructive absolute determination of the amount of tritium in a single target is not presently possible and interpretation of laser-fusion experiments depends strongly on this information. Work in this area should continue.

FIG. 20. Unexplained non-exponential outgassing behavior of pressurized laser targets.

CONCLUSIONS

Materials problems in fusion reactors of all kinds arise in the context of breeding, recovering, containing, and recycling tritium. Technically, all these problems are probably solvable, but the economic feasibility of the solutions is still in question. The treatment of materials problems here is necessarily very general, but the references should provide a useful guide to the current literature on these topics.

FIG. 21. Non-repeatability of outgassing behavior for tritium-filled microencapsules.

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