# MATERIALS COMPATIBILITY CONSIDERATIONS FOR A FUSION-FISSION HYBRID REACTOR DESIGN*

**MASTER**

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## ABSTRACT

The Tandem Mirror Hybrid Reactor is a fusion reactor concept that incorporates a fission-suppressed breeding blanket for the production of $^{233}U$ to be used in conventional fission power reactors. The present paper reports on compatibility considerations related to the blanket design. These considerations include solid-solid interactions and liquid metal corrosion. Potential problems are discussed relative to the reference blanket operating temperature (490°C) and the recycling time of breeding materials (<1 year).

## INTRODUCTION

The use of fusion energy to produce fuel for conventional light water fission reactors has the potential to supply large quantities of fissile fuel while also reducing the required nuclear performance and engineering constraints of a conventional commercial fusion reactor.[1] One promising concept for such a fusion-fission hybrid reactor incorporates a direct cooled fission-suppressed blanket consisting of a cylindrical module with a beryllium multiplier in mobile pebble form, thorium metal snap-ring overlays (possibly coated) on the beryllium pebbles, and a liquid lithium coolant which is pumped directly through the pebble bed.[2] The analysis and discussion in the present paper is based on this specific design concept. Potential compatibility problems in this concept include both solid-solid and liquid metal-solid interactions. Solid-solid interactions temperature (490°C) and the recycling time of breeding materials (<1 year).

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1. Be — structural alloy (currently Fe-12 Cr-1 Mo [HT-9])
2. Th — structural alloy
3. Be — Th (plus possible barrier coating)

In addition, self-welding may occur at contact points between the beryllium pebbles (Be-Be) or thorium rings (Th-Th). Liquid metal interactions are those of molten lithium with the thorium, steel, and beryllium. Corrosion of these materials by lithium under temperature and concentration gradients and deposition of dissolved species in the cooler regions of the lithium circuit are of major importance to reactor design. Effects of impurities in the solid and liquid blanket materials (principally O, N, C, and T) on compatibility must also be examined. Although the number of compatibility concerns is large, the maximum operating temperature in the present reactor concept is low (490°C) in terms of conventional liquid metal heat transfer systems (for example, fast breeder reactors). Also, to minimize radiation effects as well as fissioning, the beryllium and thorium are replaced after several months of neutron exposure.

In the present paper, many of the possible compatibility reactions are reviewed in terms of available thermochemical and kinetic data. Results from compatibility reaction couples exposed in static lithium at 350, 450, and 550°C are also presented. The purpose of these experiments was to examine the reactions between beryllium and stainless steel in direct contact and across a molten lithium gap.

## SOLID-SOLID INTERACTIONS

The most significant solid-solid interactions are anticipated to be those involving metallic beryllium. This is because beryllium forms very stable intermetallic compounds (beryllides) with both transition and actinide metals. Furthermore, because the beryllium atom is relatively small, the solid-state diffusion rates that control interfacial reactions with beryllium tend to be correspondingly high.

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In the current reference design concept,[2] beryllium pebbles 3 cm in diameter are overlaid with a thorium snap ring. The maximum temperature at the contact interface is estimated to be 475°C. There are no experimental data on the interdiffusion of thorium and beryllium; however, thermodynamic data indicate the metals should react to form the compound $Be_{13}Th$. The compound can be expected to form as a uniform layer at the Be-Th interface and to grow at a rate controlled by the diffusion of beryllium through the developing layer (i.e., with a half-power time dependence). The mutual solubilities of beryllium and thorium are negligible at 500°C (ref. 3) so that interdiffusion should involve primarily the growth of the intermetallic compound with little solid solution alloying. The thorium density in the compound is sufficiently low that the build-up of $^{233}U$ should not constitute a hot spot problem. However, the compound would complicate the removal of the thorium rings from the beryllium, in which case it may be necessary to employ a diffusion barrier between the two metals. Possible barriers include CVD-deposited or ion-plated molybdenum, applied to the inside of the thorium ring, or a stable carbide coating (for example, TiC) applied to the exterior of the beryllium pebble. Either approach would require an experimental evaluation.

The reaction of beryllium with the containment alloy, HT-9, constitutes another potential compatibility problem. The absence of nickel as an alloying element in HT-9 should reduce the severity of interalloying with beryllium compared to that for an austenitic stainless steel. Nevertheless, the interdiffusion between iron and beryllium at the proposed operating temperature could limit the containment lifetime.

The only solid-state thorium reaction of engineering significance appears to be the beryllium-thorium reaction referred to above. Various fuel irradiation capsules and at least one reactor experiment (SRE) have operated with thorium in contact with austenitic stainless steel without significant interaction up to 650°C (ref. 4). Of most concern will be the tendency for carbon to be transferred from HT-9 to the thorium, which can example, TiC) applied to the exterior of the beryllium pebble. Either approach would require an experimental evaluation.

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The extent of self-welding between beryllium-beryllium and thorium-thorium contact points is critical to the removal of beryllium pebbles after the prescribed radiation period. Unfortunately, there is little experimental information on self-welding tendencies of these elements. Thermodynamic considerations indicate that a thin oxide coating could develop on the thorium surfaces exposed to lithium containing small amounts of dissolved oxygen, but the beryllium surfaces will be oxide free. The growth rate of interfacial attachment areas (necks) between pebbles has been calculated[2] assuming that volume diffusion is the rate-controlling mechanism. On this basis, the attachment of thorium to thorium is predicted to be negligible, and the attachment rate for beryllium is estimated to be <0.12 mm/y (0.06 mm/irradiation period). Although this is an extremely slow rate in terms of the fraction of the pebble circumference affected, it could nevertheless limit the mobility of the pebbles. Furthermore, other transport processes could operate to permit interfacial attachment, including surface diffusion and a dissolution/deposition process through the lithium melt. Thus, it is important to establish the nature of the external beryllium surface that will exist in the blanket (for example, whether it will develop an intermetallic or carbide layer in contact with lithium, as discussed below).

If beryllium self-welding is shown experimentally to be a potential problem at 475°C, there are various coatings with acceptable neutronic properties that can be applied to the pebbles to limit the volume and surface diffusion rates of beryllium. One logical approach would be to take advantage of the very stable and refractory intermetallic compounds formed between beryllium and such metals as chromium, iron, and molybdenum. Coatings of the latter metals could be applied in pure form to the beryllium surface and then reacted at about 800°C to produce the beryllide compound. Producing a nitride or carbide coating on the beryllium could also be an effective approach, but achieving chemical stability in the presence of HT-9 and thorium may be more difficult than for a beryllide.

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## LIQUID METAL CORROSION/MASS TRANSFER

The direct-cooled blanket concept is predicated on the chemical compatibility of beryllium, thorium, and HT-9 with liquid lithium. The compatibility issue here transcends more than just the resistance of these individual materials to lithium at design temperatures. A larger concern is how the presence of any one material in the circuit will affect the corrosion performance of the others. For example, all of the solid-solid interactions discussed above may be fundamentally affected by the presence of lithium.

Lithium corrosion can take several forms: dissolution, alloying, impurity (including carbon) transfer, intergranular penetration, and thermal-gradient and dissimilar metal mass transfer. Each type of corrosion reaction can lead to specific problems. For example, the mass transfer and subsequent deposition of dissolved species can cause flow restrictions with attendant hot spots in the pebble bed (although this effect could be mitigated by the one-half year residence time), while in cases where intergranular penetration may occur, the mechanical integrity of the structural alloy could be compromised. Impurity transfer can result in the formation of surface compounds on the solid blanket components or in the carburization/decarburization of a steel. Impurities can also promote intergranular attack in lithium systems.

Corrosion data for thorium in liquid lithium are not available in the open literature and will be needed for a complete compatibility assessment of the blanket system. However, the excellent compatibility of thorium with sodium[5] indicates reasonable compatibility with lithium. Whether a diffusion barrier is used against the thorium to prevent interalloying with beryllium will obviously affect the lithium compatibility question. Thermodynamic calculations indicate that at 500°C, $ThO_2$ will not be reduced by lithium containing more than 5 ppm (by weight) of oxygen. Thus, even in the absence of a deliberate barrier, wetted surfaces of thorium metal would probably be covered by a thin oxide that might serve to reduce beryllium interalloying. Carbide coatings have also been suggested as possible diffusion barriers. While such coatings may be compatible with lithium,[6] it will be difficult to find carbide coatings that are simultaneously compatible with two strong carbide formers, thorium and beryllium, in the same system.

Little is known about the compatibility of beryllium with lithium, particularly at the proposed exposure temperature ($\leq$475°C). However, no Be-Li compounds have been reported and there appears to be only limited solubility of beryllium in lithium.[7] As discussed below, the dissolution of beryllium in lithium may be exacerbated by the presence of nickel-

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Corrosion studies of ferrous alloys in liquid lithium have been ongoing in support of fusion reactors for several years.[8,9] The fusion breeder effort can therefore draw from this relevant work. Studies to date have indicated the nature and, to a lesser extent, the rate of temperature gradient mass transfer that will occur from the hotter to the cooler regions of iron-based alloy heat transfer circuits containing lithium. While mass transfer rates depend on a number of factors, the dissolution and deposition rates appear generally acceptable for HT-9 at the projected reactor operating time and temperature.

Another important compatibility reaction is that of carbon transfer between the structural alloy and the lithium.[10,11] In many cases, a ferritic steel can decarburize in lithium with an attendant change in its mechanical properties. The direction and extent of carbon transfer depend on the relative carbon activities in the lithium and steel as they are affected by temperature (including the temperature differential of a system), alloy composition, and heat treatment.[10,11] Microstructural manipulation can aid in minimizing carbon redistribution in lithium-steel systems, and such carbon transfer should be relatively sluggish at the operative temperatures of the present blanket system (<490°C). However, both the thorium and beryllium surfaces in the lithium circuit are effective sinks for carbon given the initial carbon activity of HT-9.

The rate-controlling step for the release of atoms from the steel to the molten lithium can be diffusion across the solid/liquid interface, diffusion across the stationary liquid boundary layer, or solid-state diffusion. If diffusion in the liquid-lithium boundary layer is rate-controlling for the dissolution process (as is often the case), the fusion reactor's magnetic field can affect the mass transfer rate.[12] A transverse magnetic field would act to restrict turbulence such that a thinner effective boundary layer is produced, and a higher corrosion rate would be expected. Magnetic fields may similarly influence deposition rates. There have also been several observations of preferential accumulations of iron and iron-manganese deposits in the areas of electromagnetic pumps in lithium circuits.[13,14]

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## IMPURITY EFFECTS

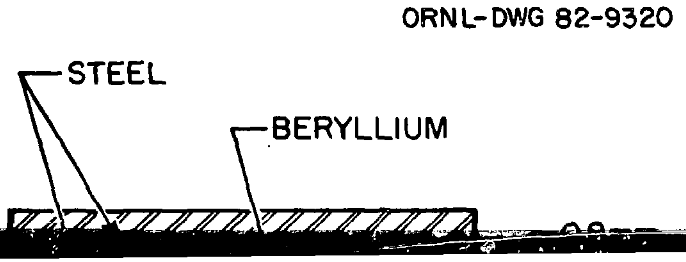
The corrosion of beryllium can be exacerbated by its reaction with impurities in the lithium. Thermochemical calculations involving the Be-Li-C-O-N system[7] have shown that BeO and 2 $BeO \cdot Li_2O$ are not stable under conditions of normal impurity limits for lithium. However, the surface oxide on the beryllium pebbles exposed to the lithium will dissolve fairly rapidly upon exposure, thereby resulting in a higher than normal oxygen level in the lithium. Although oxygen impurities do not normally affect the dissolution behavior in lithium, concentrations above about 5 ppm would result in oxidation of any uncoated thorium in the system. If these films were as protective as those formed on thorium in air at 500°C, their presence would actually be beneficial.

Nitrogen will be a more detrimental impurity in lithium than oxygen since it can react with beryllium to form $Be_3N_2$, which has been determined to be a stable compound in the Li-Be-O-C-N system at typical impurity levels.[7] In addition, results from the experimental work reviewed in ref. 9 have indicated that higher levels of nitrogen in lithium accelerate, and possibly change, the corrosion reactions of steel with lithium. Because of the lack of thermodynamic data on the Th-N system, we cannot assess the influence of nitrogen on the compatibility of thorium with the other blanket materials.

As discussed above, the activities of carbon in lithium and in the structural steel determine whether carburization or decarburization of the steel will occur. Typical steel carbides ($M_{23}C_6$, $M_6C$) are thermodynamically more stable than stoichiometric thorium carbide and are about as (or possibly less) stable as $Be_2C$. A carbide coating on the thorium metal (for example, TiC) would be stable in lithium so long as it is also more stable than $Be_2C$. Thorium carbide is marginal in this respect.

## RESULTS OF Li-Be COMPATIBILITY EXPERIMENTS

Diffusion couples of type 316 stainless steel and beryllium (shown schematically in Fig. 1) were held in static lithium at 350, 450, and 550°C. Examination of couples



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Diffusion couples of type 316 stainless steel and beryllium (shown schematically in Fig. 1) were held in static lithium at 350, 450, and 550°C. Examination of couples exposed for 1000 and 3000 h showed significant interalloying between the beryllium and steel at 450 and 550°C, the depth increasing with temperature (Fig. 2). Additionally, as shown in Fig. 3, a layer of BeNi, identified by

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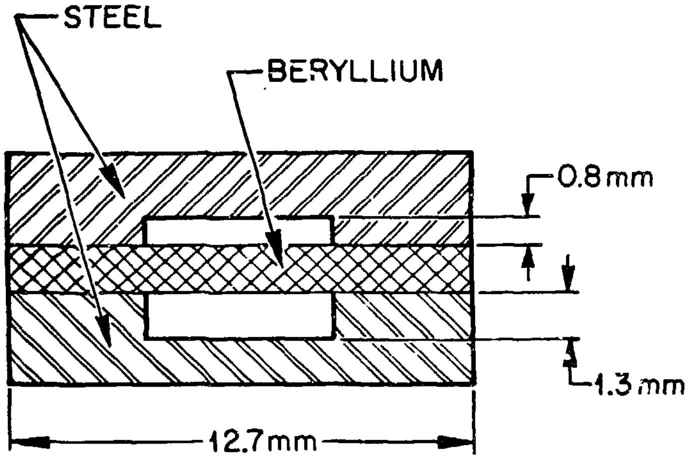


Fig. 1. Schematic drawing of diffusion couples exposed to static lithium.

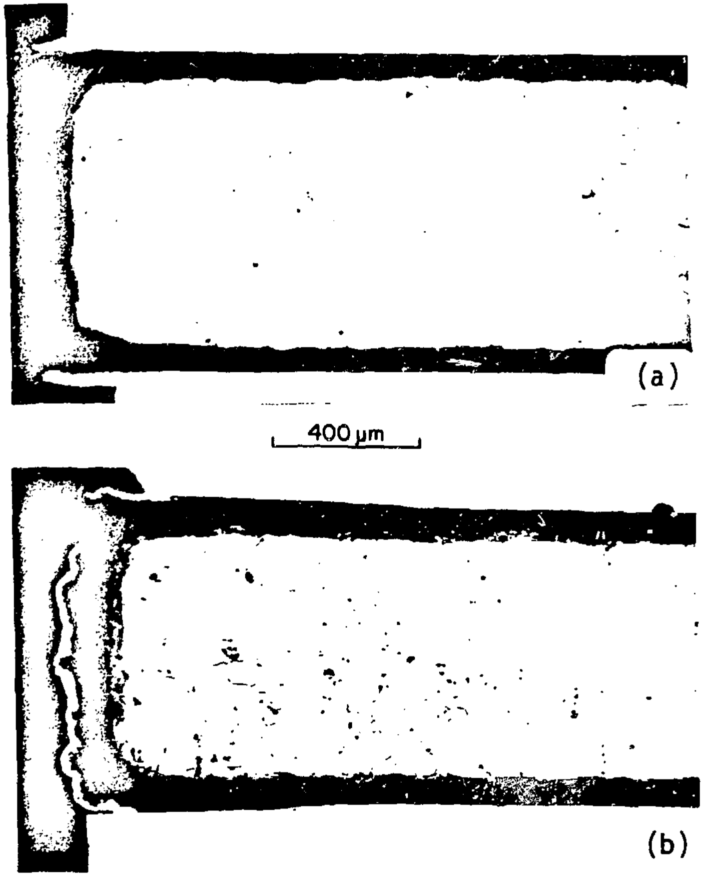
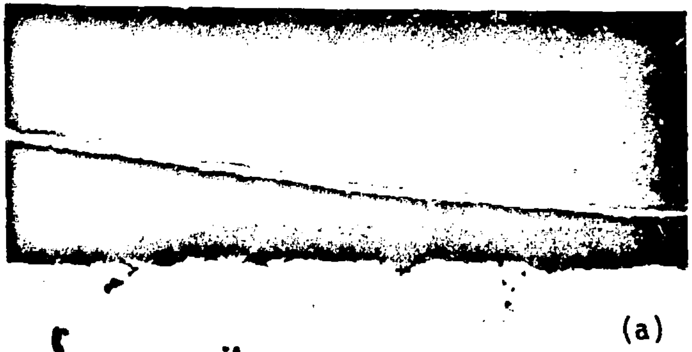
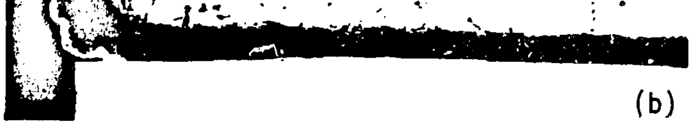


Fig. 2. Type 316 stainless steel/Be/type 316 stainless steel reaction couples exposed in static lithium for 3000 h. (a) 450°C. (b) 550°C.





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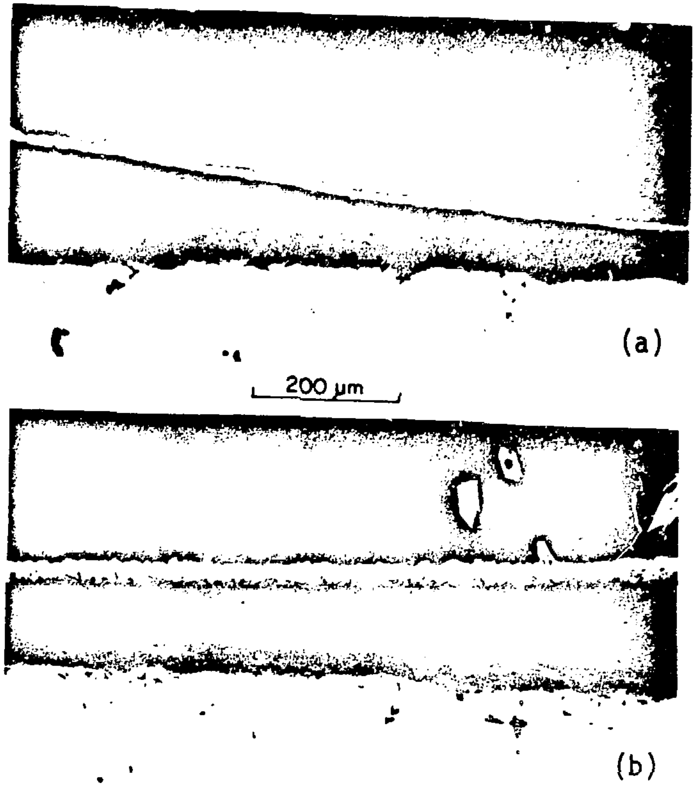

Fig. 3. Cross-sections of beryllium that have been exposed for 3000 h in static lithium with type 316 stainless steel in close proximity. Detached material is BeNi. (a) 450°C. (b) 550°C.

x-ray diffraction, was formed within lithium-filled gaps which separated the beryllium and steel at distances of 0.8 and 1.3 mm, respectively. The 350°C diffusion couple showed only a superficial beryllium-nickel reaction layer at points of direct contact. No evidence of beryllium-iron interalloying was found in these tests. Iron-chromium steels would therefore be preferable to those containing nickel.

## SUMMARY OF ISSUES

Any compatibility assessment of the present fusion breeder blanket design concept is complicated by the number of blanket components. Solid-solid interactions include self-welding of the thorium and beryllium (in the presence of lithium), the interalloying of thorium and beryllium, and the interalloying of beryllium and the structural steel. Of highest priority are the self-welding reactions, because these will determine whether barrier coatings must be applied to the pebbles to guarantee their removability. Possible coatings for the thorium metal are $ThO_2$, ThC, or TiC. Possible coatings for the beryllium metal are $Be_{12}Fe$, $Be_{12}Cr$, or $Be_{12}Mo$. Next in priority are the possible reactions of beryllium with thorium. The same coating candidates listed above could also be used to reduce the extent of the latter reaction. Last in priority are the reactions between beryllium and HT-9, which should be relatively slow at 450°C.

In the area of liquid-solid interactions, major concerns are (1) temperature gradient mass transfer of the containment alloy (HT-9) and/or breeding materials by lithium with attendant reduction of the blanket fluid passages by chromium, iron, or beryllium deposition and (2) mass transfer of metallic and nonmetallic (impurity) elements among the chemically dissimilar blanket and structural materials. The use of a structural steel of low nickel activity and the relatively low operating temperature ($<490°C$) will reduce the pebbles to guarantee their removability. Possible coatings for the thorium metal are $ThO_2$, ThC, or TiC. Possible coatings for the beryllium metal are $Be_{12}Fe$, $Be_{12}Cr$, or $Be_{12}Mo$. Next in priority are the possible reactions of beryllium with thorium. The same coating candidates listed above could also be used to reduce the extent of the latter reaction. Last in priority are the reactions between beryllium and HT-9, which should be relatively slow at 450°C.

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In both the solid-solid and liquid-solid cases, the relatively low operating temperature and short recycling time of this fusion breeder concept reduce the compatibility concerns discussed above.

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