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The Thermal Conductivity of Mixed Beryllia/Li Ceramic
in Sphere-Pac Forms

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ABSTRACT

Lithium containing ceramic tritium breeder materials have been envisaged to be deployed within the blanket region of a fusion reactor in several possible configurations. One of these is the sphere-pac configuration. For this configuration an important material parameter is its thermal conductivity (Ksp). It is well known that Ksp demonstrates rather complex behavior as a function of temperature, gas pressure, gas composition, particle size, and packing fraction. The inter-relationship of these parameters has been satisfactorily accounted for with a hierarchical effective media theory (HEMT). For tritium self sufficiency most lithium ceramic breeder materials would require the presence of a neutron-multiplier (e.g., Be or BeO). Here, the influence of configuration on Ksp (i.e., how one put the different solid components together in the sphere-pac bed) becomes important. Using a generalized HEMT (i.e., a model with capability to describe systems with more than one solid material component) we have analyzed in detail the configurational dependence of Ksp for sphere-pac beds composed of lithium ceramic/BeO microspheres. Substantial improvements in Ksp can be achieved if a configuration of lithium ceramic spheres coated with BeO is chosen.

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1. INTRODUCTION

Lithium containing solid ceramic breeder materials are among the leading candidates as tritium breeder materials for fusion power reactors. One common form of these ceramic breeders is as sintered pellets or block format. A potential concern for ceramic materials in this form is the possible fracturing of these materials under relatively low stress levels. This fracturing would lead to a loss in mechanical integrity and possible degradation in thermal response. The sphere-pac (SP) configuration concept provides a possible solution to these problems.[1]

The sphere-pac concept involves the use of a bed of the ceramic breeder material in the form of microspheres of different sizes. The bed is usually permeated through with an inert gas such as helium. The need for a high packing fraction (of the order of 85 to 90%) for a more compact design requires microspheres of a fairly wide distribution of sizes. Such sphere-pac configurations would certainly minimize the potential for fracturing and loss of mechanical integrity. Much experience has been gained on the sphere-pac bed concept from fission technology research.[2] The nature of the sphere-pac configuration is such that its effective thermal conductivity can be significantly lower than the corresponding solid component because of the limited contact between the spheres leading to the bulk thermal conductivity being dominated by the thermal conductivity (K_{sp}) of the gas phase. Concepts that can lead to an enhancement of K_{sp} would be of definite benefits. This work describes a concept that can lead to an enhancement of thermal conductivity for materials of a sphere-pac configuration.

Consideration of the tritium breeding ratio (i.e., >1 for tritium self sufficiency) implies that for most solid breeder materials the addition of a neutron multiplier (e.g., Be or BeO) is a necessity.^[1] Although the formalism presented in this work is in no way dependent on the specific material used for neutron multiplication, the following discussion is couched in terms of BeO. Exactly the same analysis could be employed should Be instead of BeO be chosen as the neutron multiplier only the numerical results would be different. However, since Be has a higher thermal conductivity than that of BeO^[3] one would expect our qualitative conclusions to be further reinforced.

The necessity for neutron multipliers leads to extra considerations for blanket designs that are generally less compact and more involved. However, the drawback of requiring neutron multipliers can, to some extent, be compensated by the enhancement of K_{sp} due to the presence of a highly thermally conductive component such as BeO (conductivity ~ 87 W/MK compared to that of 2.8 W/MK for LiAlO_2 at $T \sim 700$ K).^[3,4] The new notion introduced here is that there exists specific configurations (as compared to more conventional ones) through which the conductive component (BeO) can be introduced into the sphere-pac bed in such a manner so as to further enhance the effective thermal conductivity of the sphere-pac bed. By conventional configurations, one means those in which BeO is introduced in a random manner into sphere-pac beds in the form of spheres similar to that of the solid breeder component. It turns out that by coating the solid breeder spheres (e.g., LiAlO_2) with the right amount of BeO one can achieve an optimized

coated sphere configuration (CSC) thermal conductivity that is greater than that of the conventional mixed sphere configuration (MSC) of the same BeO/solid breeder composition. The basis for this conclusion is described in the following sections.

2. THEORY

We have recently developed the hierarchical effective medium theory (HEMT) in order to estimate the thermal conductivity of finely divided materials.^[5] It is a theory that has no adjustable parameters. For a sphere-pac bed composed of spheres of varying sizes HEMT is a particularly suitable tool. The basis of HEMT relies upon successive embedding of the larger spheres into an effective medium of smaller microspheres in a self-consistent manner.

For the present situation the HEMT needs to be generalized to take into account the presence of two material components, the lithium ceramic breeder and the neutron multiplier material. Furthermore, these two components are arranged in two different configurations, namely, the coated sphere configuration (CSC) and the mixed sphere configuration (MSC). The CSC configuration consists of a sphere-pac bed of touching spheres of LiAlO_2 , each of which is coated with a layer of BeO of appropriate thickness, while the other configuration (MSC) is made up of randomly mixed similar-sized spheres of both LiAlO_2 and BeO. The relative volume fraction of BeO for the two beds is the same (20%). Both beds are assumed to be under a helium

atmosphere. The spheres have diameters of 1200 μm , 300 μm and 35 μm and they have respectively volume fractions of 0.511, 0.173, 0.182. These values are typical of a fission-fuel sphere-pac bed.^[3] These system parameters can, of course, be varied without affecting the basic tenet of the theory.

The thermal conductivity K_s of a coated sphere is calculated as follows^[6]

$$K_s = K_c[2K_c + K_i + 2\eta(K_i - K_c)]/[2K_c + K_i - \eta(K_i - K_c)] \quad (1)$$

where

K_c = thermal conductivity of the coated layer,

K_i = thermal conductivity of the interior component under the coated layer,

η = volume fraction of the interior component.

Using Eq. 1 and treating the coated sphere as if it is made up of one material component only, the standard HEMT can be applied in a straightforward manner.

An important material parameter is the temperature-jump distance, d_j ^[7,8], which characterizes the heat transfer behavior across a gas-solid interface. The temperature jump distance depends on the bulk gas properties and on the geometrical environment of the solid surfaces (e.g., their curvature and roughness). However, on an engineering scale, experimental data has indicated that d_j is insensitive to the material that forms the underlying substrate.

For example, experimental evidence has indicated that little difference exists between the d_j for He/ UO_2 and for He/stainless steel.^[9] Thus, we have taken the values of d_j measured for He/ UO_2 as the input to our calculation.^[8] Because of the insensitivity of d_j to the substrate material, this procedure appears reasonable.

The thermal conductivity of $LiAlO_2$, BeO and He are used.^[3] The temperature variation of thermal conductivity is derived from four sources and arises from the temperature dependence of the solid components, the gas phase, and that of d_j ; d_j is also pressure-sensitive.^[7,8]

A modification of the above approach is necessary for the mixed sphere configuration since there are now two types of unit cells. One is given by a $LiAlO_2$ sphere surrounded by its gaseous environment, the other being a BeO sphere surrounded by its own gas component. The effective thermal conductivities of these two unit cells are calculated via the previous approach. The random mixing of these two types of unit cells give rise to our sphere-pac bed. Note that in this configuration none of the two components is necessarily considered as a continuous medium. In a natural manner, this allows application of Bruggeman's symmetrical effective medium theory (SEMT)^[6] to compute the conductivity of this sphere-pac bed. Using these two generalizations to HEMT we have calculated the thermal conductivity of a sphere-pac bed in two configurations, i.e., the mixed sphere and the coated sphere configurations.

3. RESULTS AND DISCUSSION

Figure 1 shows the calculated thermal conductivity of a sphere-pac bed in the mixed sphere (MSC) as well as the coated sphere (CSC) configuration. The theory used is the generalized versions of HEMT discussed in the previous sections. One notes immediately that $[K(CSC)]$ the thermal conductivity of the coated sphere configuration is significantly above the corresponding conductivity for the mixed sphere configuration $[K(MSC)]$. To indicate in a quantitatively manner the comparison between the two configurations we would introduce an enhancement factor ξ defined as follows

$$\xi = \frac{K(CSC) - K(MSC)}{K(MSC)} \times 100\%$$

where ξ is a measure of the percentage enhancement achievable with CSC over that of MSC; it can also be considered a figure of merit for the CSC concept.

The results as shown in Fig. 1 indicates that ξ varies from about 70% at 500K to approximately 30% at 1500K. The broad structure between 400K and 600K is a reflection of the different temperature dependence of the two configurations. In fact, $K(CSC)$ decreases much more rapidly with temperature than does $K(MSC)$. This can be easily understood since in the CSC concept there is a deliberate attempt to enhance the effective contribution of the more conductive component (in this case, BeO) to the overall thermal conductivity of the system. In turn, this leads to the result that over the

Fig. 1. Thermal Conductivity of $\text{LiAlO}_2 + \text{BeO}$ in Different Configurations (see text) Versus Temperature.

temperature range considered $K(\text{CSC})$ decreases more rapidly with temperature since the thermal conductivity of BeO itself decreases much more rapidly with temperature than does that of LiAlO_2 ^[3,4].

The results shown in Fig. 1 have been obtained under a He pressure of 0.1 MPa (about 1 atmosphere). In powder-like systems such as the sphere-pac beds, the regions close to the points of contact between particles can have linear dimension that is comparable or smaller than that of the mean free path of the gas (in our case, He). In that case, there could be significant pressure dependence of the thermal conductivity.^[7] Again, the generalized HEMT was employed to estimate the pressure effect. The results are shown in Fig. 2.

The pressure range being considered is between $P=0.1$ to 0.9 MPa (approximately between one and nine atmospheres) at a temperature of 1000K . Calculations also were carried out for a broad range of temperature. However, the results are qualitatively similar and it is sufficient to center our discussion on the $T=1000^\circ\text{K}$ results.

Figure 2 indicates that for very moderate changes in gas pressure, significant enhancement in thermal conductivity can be achieved. In fact, for the CSC almost a factor of two was gained in thermal conductivity when the gas pressure was increased from 0.1 to 0.9 MPa. Although both $K(\text{CSC})$ and $K(\text{MSC})$ were affected by gas pressure, the CSC system is the more sensitive. This can be seen clearly in the enhancement factor ξ , which increases rapidly with

Fig. 2. Thermal Conductivity of $\text{LiAlO}_2 + \text{BeO}$ in Different Configuration (see text) Versus Gas Pressure.

pressure initially and then tends towards a near-saturation value. Within the range of pressure considered, it is obvious that one has not yet reached saturation.

The results in Fig. 1 and Fig. 2 demonstrate the basic attractiveness of the CSC (i.e., coating) concept. To proceed beyond this phase would require a detailed materials development program; it is nevertheless instructive to highlight the important materials issue that needs to be addressed in such a materials development program. Note that some of these issues are germane to any consideration of solid breeder/multiplier system and are not relevant only to the CSC concept.

1. Tritium Inventory

The question of tritium inventory for the CSC is an important one and needs to be investigated. However, two facts exist that may render the tritium inventory issue a less critical one for the CSC concept. First, $D_T(\text{BeO})$, the tritium diffusion coefficient in BeO, is of the order 10^{-14} - 10^{-15} cm^2/sec at 600°C .^[10] This is comparable to the $D_T(\text{LiAlO}_2)$ extracted from several sets of tritium release data.^[1] Secondly, any sphere-pac bed by design already possesses a high surface area-to-volume ratio, which is highly beneficial to tritium permeation and release from the bulk interior of the solid.

2. Mechanical and Chemical Compatibility

The possible mismatch of elastic and thermal expansion properties between coating and substrate can lead to fracturing of the coating. If the resulting stress on the coating is compressive in character, then the coating can tolerate a much higher stress level without fracturing. To resolve this issue extensive and reliable mechanical properties data would need to be acquired and detailed stress analysis carried out. Validation needs to be obtained via careful experimental testing procedures on the prototypes.

These observations also apply to the issue of chemical compatibility^[11] between the coating and the substrate. Here the problems may originate from possible chemical interactions between the various components, particularly near the interfacial region. In this area the issues can only be resolved via thermodynamic analysis and experimental investigations on the possible chemical interactions as functions of environment.

3. Irradiation Effects

BeO is known to swell under irradiation.^[4] In a materials development program one should aim to quantify the impact of this behavior on the CSC concept for various substrate/coating systems. It is important to investigate possible techniques to suppress or reduce this irradiation-induced swelling phenomenon via methods using impurity additives.

The above description cuts across a wide spectrum of material performance issues. However, this is a requirement typical of an engineering development program if one wishes to proceed beyond the present conceptual analysis stage (which this work has demonstrated) to the eventual goal of system deployment.

4. CONCLUSION

Given two heterogeneous systems whose constituents and compositions are identical, these two systems can still possess significantly different properties if the constituents in the two systems are arranged in different geometrical configurations. In the present case, both the mixed sphere configuration (MSC) and the coated sphere configuration (CSC) have identical material components (BeO and LiAlO_2), the same compositions (i.e., volume fractions) and identical size distributions of the microspheres that make up the two systems. They operate under the same conditions of temperature and He pressures, yet a simple rearrangement of the relative geometrical configuration between the two material components lead to a significant difference in their thermal conductivities.

It is well understood that in order to characterize the properties of a complex heterogeneous medium one requires much more detailed microstructural information beyond simple averages. Differences in the details of the microstructural information, while retaining the same average as given by the volume fractions, can lead to substantial variations in the system properties.

In fact, this point underlies one of the basic difficulties in constructing a satisfactory theory of the properties of heterogeneous media. However, our present concept of a coated sphere configuration capitalizes on this system-dependence on microstructural details in seeking to maximize the long-range connectivity of the better conductive component (namely, BeO). Detailed considerations described in this work have demonstrated the qualitative performance expected of this concept as well as the quantitative enhancement achievable for the overall thermal conductivity of the composite system. It must be emphasized that although the present work concentrates on the $\text{LiAlO}_2/\text{BeO}$ system, the coating concept is potentially applicable to other coating/substrate combination materials. For different coating/substrate combinations the detailed issues in a development program outlined in the last section are likely to require different degrees of emphasis. However, a preliminary assessment of the potential attractiveness of any specific combination can be executed using the tool of a generalized version of HEMT, as described here and whose applicability has been demonstrated in this work.

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