ANL/FPP/TM-184

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DE85 000775

ARGONNE NATIONAL LABORATORY
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THERMAL CONDUCTIVITIES FOR SINTERED AND
SPHERE-PAC Li_2O AND $\gamma\text{-LiAlO}_2$ SOLID BREEDERS
WITH AND WITHOUT IRRADIATION EFFECTS

by

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Fusion Power Program

July 1984

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Work supported by the Office of Fusion Energy
U. S. Department of Energy

* Materials Science and Technology Division
**Chemical Technology Division

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ABSTRACT

Thermal conductivities (k , k_{eff}) have been estimated for sintered and sphere-pac Li_2O and $\gamma\text{-LiAlO}_2$ with and without neutron irradiation effects. The estimation is based on (1) data from unirradiated UO_2 , Li_2O , and $\gamma\text{-LiAlO}_2$; (2) data from irradiated dielectric insulator materials; and (3) relatively simple physical models. Comparison of model predictions with limited ex- and in-reactor data found reasonable agreement, thus lending credence for their use in design applications. The impact of thermal conductivities on tritium breeding and power generation in fusion solid-breeder blankets is briefly highlighted.

1. INTRODUCTION

The technological importance of thermal conductivity to fusion solid-breeder blankets is uniquely related to the two basic functions of the blanket, i.e., tritium generation and recovery, and heat generation and removal. The uniqueness originates from (1) the dependence of blanket tritium breeding and heat generation capabilities on solid/non-solid breeder volume fractions, (2) the temperature limits (upper and lower) within which solid breeder blankets are designed and operated, and (3) the dependence of tritium recovery and heat removal on solid breeder temperatures. While thermal conductivity is generally an important thermophysical property of materials, it is a critical property for fusion solid-breeder blankets.

This paper is concerned with the thermal conductivities, k and k_{eff} , respectively, for sintered and sphere-pac solid breeders (Li_2O and $\gamma\text{-LiAlO}_2$). Sintered Li_2O and $\gamma\text{-LiAlO}_2$ are the conventional solid breeder ceramics featured in many blanket designs and materials studies. One of the major problems in blanket temperature control with sintered solid breeders is the thermal conductance at breeder/structure interface. Accurate prediction of this interface conductance (and therefore breeder temperature control) is very difficult, largely due to factors such as solid breeder cracking, cracked

fragment relocation, sintering, creep, and swelling, which will alter the separation distance between solid breeder and structural materials (coolant tubes, for example) during blanket operation [1]. To reduce the uncertainty in the interface thermal conductance associated with breeder cracking and relocation, sphere-pac solid breeders have been proposed as an alternative. The basic idea of sphere-pac configuration is that with small spherical particles, the temperature gradient and therefore the thermal stresses in the particles should be insignificant to cause particle cracking. Furthermore, since the particles are packed among themselves and in discrete contacts with structural materials (i.e., coolant tube and module wall), variation in the interface conductance during blanket operation should be small, as long as there are no particle sintering, particle breakup, and resettlement. For adequate tritium breeding, a high-volume fraction of solid breeder is required. Based on fission experience with a simple cylindrical fuel element geometry, a solid packing fraction (SPF) of ~87% appears attainable for sphere-pac solid breeders with three particle sizes (1200, 300, and 30 μm diameters) and with particle density ~100% TD. However, both the fabrication of theoretically dense Li_2O and $\gamma\text{-LiAlO}_2$ spheres and the practicality for complex blanket module configurations remain to be demonstrated. Subgrains with submicron sizes are also a mandate for the $\gamma\text{-LiAlO}_2$ spheres for tritium release purposes.

This paper starts with a review of the available thermal conductivity data on unirradiated, sintered Li_2O and $\gamma\text{-LiAlO}_2$. No data exist for unirradiated, sphere-pac solid breeders. A simple model is used to develop a procedure for estimating k_{eff} , with the aid of UO_2 data. Neutron irradiation effects on thermal conductivities are included in a model to obtain quantitative estimates of k and k_{eff} for sintered and sphere-pac Li_2O and $\gamma\text{-LiAlO}_2$.

The model predictions are then compared with the limited ex- and in-reactor data. Finally, the impact of thermal conductivities on tritium breeding and power generation in fusion solid-breeder blankets is briefly highlighted.

2. ANALYSIS AND RESULTS

2.1 Unirradiated, Sintered Solid Breeders

Takahashi and Kikuchi [2] measured thermal diffusivities of unirradiated, sintered Li_2O with variations in temperatures and porosities that cover the range of interest of the Li_2O blanket. They found that the Li_2O thermal conductivity can be represented well ($\pm 3\%$ deviation from data) by the following expression,

$$k(P) = k_0 f(P) \quad W/m-K, \quad (1)$$

where k_0 is the thermal conductivity of a theoretically dense Li_2O ; $f(P)$ is a porosity function (with P the fractional porosity) given by

$$k_0 = \frac{1}{A + BT}, \quad (2)$$

$$f(P) = \frac{1 - P}{1 + \beta P}, \quad (3)$$

and the constants are $A = 2.2 \times 10^{-2}$, $B = 1.784 \times 10^{-4}$, and $\beta = 1.45 \times 10^{-4} \text{ T}$, where T is the temperature in K.

Two sets of thermal conductivity data on unirradiated, sintered $\gamma\text{-LiAlO}_2$ were found in the literature [3,4]. Both sets were included by Suiter [5] in

an assessment of lithium-based oxide ceramics for tritium breeding applications. The data of Gurwell [3] were obtained for a mixed LiAlO_2 (50-75% γ - LiAlO_2 and 25-50% α - LiAlO_2) body at temperatures up to 1200 K. The density was listed as 88.5% of 2.62 g/cm^3 . There were no descriptions, however, on test procedure, environment, or grain size of the sample. The data of Hollenberg [4] were obtained for high-purity, fine-grained ($<1 \text{ }\mu\text{m}$) γ - LiAlO_2 (83.8% TD) by a laser pulse technique. All measurements were made in helium atmosphere to more closely simulate the conditions in a tritium breeding blanket, which also resulted in higher k values than those of Gurwell at comparable temperatures (even though the latter have higher density). Unfortunately, however, Hollenberg measured thermal conductivity only up to 723 K, which is significantly below the upper temperature limit (1273 K) considered for γ - LiAlO_2 in the blanket comparison and selection study [6].

Suiter [5] gave two correlations in which the thermal conductivity data of Gurwell and Hollenberg are represented by separate quadratic functions of temperature. Initial use of these correlations for extrapolation into high temperatures was found unsatisfactory. Both correlations have an inflection point in temperature above which the calculated thermal conductivity increases rapidly with temperature. Such trends have not been observed in the data over the experimental temperature regimes. In fact, Suiter observed that the difference between the two sets of data, although quite large at low temperature, decreases at high temperatures.

In the following analysis, the data of Hollenberg are recorrealted with a different temperature function, i.e.,

$$k = \frac{1}{A + BT} \quad \text{W/m-K} \quad , \quad (4)$$

where A and B are constants found from regression fitting, $A = 7.274 \times 10^{-2}$ and $B = 4.67 \times 10^{-4}$ with T in K. This type of temperature function has a good theoretical basis for phonon lattice conductivity above the Debye temperature, and yielded a correlation coefficient of 98.5%. Equation (4) is hereafter used to calculate thermal conductivities of unirradiated, sintered γ -LiAlO₂ (84% TD) at higher temperatures (>723 K). As an example, Eq. (4) gives a calculated thermal conductivity of 1.58 W/m-K at 1200 K, whereas Gurwell gave 1.78 W/m-K; the difference may be rationalized against material densities (or porosities) and compositions. It should be noted that at still higher temperatures (>1200 K), Eq. (4) may eventually break down as contributions to conduction from both photons and electronic charge carriers become important. An upswing in thermal conductivity with temperature would then be expected. As mentioned before, however, such an upswing has not been observed in the present data sets up to 1200 K.

To get the thermal conductivity for unirradiated, sintered γ -LiAlO₂ at other densities, one needs a relationship for the porosity dependence of thermal conductivity. This porosity dependence should not be significantly different from that of sintered Li₂O. Because of the lack of data, the same porosity dependence of Li₂O is assumed for γ -LiAlO₂, i.e.,

$$k_o = \frac{1}{A + BT} f(P)^{-1} \quad \text{W/m-K} \quad , \quad (5)$$

where f(P) is given by Eq. (3).

2.2 Unirradiated, Sphere-Pac Solid Breeders

As mentioned before, no data exist on the thermal conductivity of sphere-pac solid breeders. Before discussing the UO₂ data and the model, it is

useful to examine the heat transport processes in a sphere-pac solid breeder configuration.

Heat transport in a system composed of close-packed spherical particles involves four distinctive processes: (1) lattice conduction, by which heat is transferred by phonons, photons and electrons within the lattice of solid particles; (2) gas phase conduction, by which heat is transferred through the gas phase in the open spaces between particles and at gas-solid interfaces; (3) radiation heat transport, which is important at high temperatures and depends on surface emissivity; and (4) contact heat transport at solid particle interfaces, which depends on contact pressure and surface roughness. Although each of the above heat transport processes is reasonably understood, the development of a model for the overall thermal conductivity (k_{eff}) of the system is complicated by the complex geometrical particle arrangement and the relative importance of each heat transport process. Mechanistic modelings [7,8] were attempted in the past, which combined several of the above individual processes in series/parallel forms, for single-size particle and for vipac (two particle sizes) beds; no successful approach has been developed for a sphere-pac (three particle sizes) bed.

Moore et al. [7] investigated the effective thermal conductivities of powders with UO_2 or ThO_2 microspheres in several gases from 300 to 1300 K. Filled gas effects on k_{eff} were clearly indicated in the three-particle (1200, 300, and 25-45 μm nominal diameters) blended bed of UO_2 (SPF ~87%) in He, N_2 , and Kr versus temperature at a gas pressure of 0.1 MPa (~1 atm). The relative magnitudes of k_{eff} 's reflect the different thermal conductivities of the three gases, with helium giving the highest k_{eff} . The UO_2 particles used by Moore et al. were prepared by the "sol-gel" process, which resulted in particle density greater than 98% theoretical. By comparing the k_{eff} thus obtained for

the three-particle bed to that of the high-density ($\sim 98\%$ TD), sintered UO_2 , one can assess the reduction of thermal conductivity due to the presence of interparticle spacings and the contact resistance between particles (radiation transport may be ignored because of the relatively low temperatures). Figure 1 shows the comparison of thermal conductivities for unirradiated UO_2 . The solid lines for 98% and 95% TD sintered UO_2 represent data reported in Refs. [9] and [10]. By calculating the ratio between the thermal conductivity of 98% TD sintered UO_2 and the k_{eff} of Moore et al. (obtained with helium gas at 0.1 MPa), one finds that k_{eff} is reduced by a factor of 3.5 at 500 K and by a factor of ~ 2.75 at 1200 K.

When the helium gas pressure was increased, Moore et al. [7] noted an increase in k_{eff} . The gas pressure effect on k_{eff} starts to level off, however, at a helium pressure of 0.6 MPa (~ 6 atm) such that further increase in pressure resulted in only marginal improvement of k_{eff} . The k_{eff} data at 0.2 and 0.6 MPa helium pressures are also plotted in Fig. 1. Based on the 0.6 MPa data, the k_{eff} 's are factors of ~ 2.3 and ~ 1.78 less than those of the 98% TD sintered UO_2 at 500 and 1200 K, respectively.

A simple way to estimate k_{eff} for sphere-pac Li_2O and $\gamma\text{-LiAlO}_2$ is to assume that the same reduction factors in k_{eff} observed in the UO_2 data apply to solid breeders. Using the thermal conductivity data for high-density, sintered solid breeders, one can readily obtain k_{eff} for the sphere-pac solid breeders. In the present case where high-density, sintered data are not available for solid breeders, Eqs. (2) and (5) can be used. The validity of the estimates depends on two assumptions: (1) all three systems (UO_2 , Li_2O , and $\gamma\text{-LiAlO}_2$) are made of high-density ($\sim 100\%$ TD) particles with three different sizes (30, 300, and 1200 μm diameters), and (2) the reduction factors (temperature and helium-pressure dependent) in thermal conductivity observed

Figure 1. Thermal conductivities for unirradiated, sintered, and sphere-pac UO_2 versus temperature and helium fill gas pressure.

in the sphere-pac UO_2 are applicable to sphere-pac Li_2O and $\gamma\text{-LiAlO}_2$. The second assumption also implies the same details in the gas-solid heat transport and in the contact resistance between particles for all three systems. For the UO_2 experiment [7] and for a sphere-pac blanket module where external constraints from the tubing and module wall are relatively small, the contact heat transport is probably of secondary importance, compared to heat transport at gas-solid interfaces.

Near the solid particle contact region, the distance between the spheres becomes smaller than the mean free path of gas molecules. Rarefied gas properties are therefore expected in this region. Heat transfer under such conditions is often represented in terms of a "temperature jump distance" at the gas/solid boundaries [11]. This distance depends on the accommodation coefficient for the gas and solid surfaces, and on several gas properties (e.g., thermal conductivity, viscosity, specific heat, etc.). While the latter helium gas properties are identical in the sphere-pac UO_2 and solid breeder systems, the accommodation coefficients can be quite different, as they involve both gas and solid. Kennard [11] gives a simple model in which the molecular masses of the gas and solid completely determine the accommodation coefficient. The model predicts a much lower accommodation coefficient (~ 0.03) for UO_2/He than those for $\text{Li}_2\text{O}/\text{He}$ (~ 0.21) and $\gamma\text{-LiAlO}_2/\text{He}$ (~ 0.11). This would result in a larger temperature jump distance for UO_2/He than for solid breeders/He. Since the temperature jump distance represents incomplete heat transfer near the gas/solid interface, the larger the temperature jump distance, the less effective the heat transfer. This means that the gas-phase heat transport contribution to k_{eff} will be more for UO_2 than for solid breeders. On this basis the estimated k_{eff} for sphere-pac solid breeders may be conservative (i.e., lower than the actual values). One needs to be careful with the

Kennard model, however, since other factors (such as surface roughness) can have an overriding effect. In their model for the k_{eff} of sphere-pac UO_2 , Moore et al. [7] treated the accommodation coefficient as a variable in their calculations to match the data. The values used vary from 0.1 to 0.4, which are all significantly higher than 0.03, as calculated from the Kennard model.

A flaw inherent in the above approach becomes apparent when one considers the irradiation effects on k_{eff} . The flaw arises from not separating the contribution to k_{eff} from the individual heat transport processes. Displacement and capture damages in solid breeders induced by fast neutron bombardment cause most change in the lattice thermal conductivity and very little change to the other heat transport processes, except perhaps for the contact conduction (due to radiation-enhanced sintering).

Since one needs to consider only the lattice conduction part to include irradiation effects on k_{eff} for the sphere-pac systems, the following model is used first to separate the contributions to k_{eff} from individual heat transport processes in the UO_2 data. A general model for k_{eff} of a sphere-pac system may be written as

$$\lambda_{eff} = \sum_i \sum_j f_{ij} \lambda_j, \quad (6)$$

where $\lambda_{eff} = 1/k_{eff}$ is the overall thermal resistance of the system, λ_j is the thermal resistance of the j th heat transport process, and f_{ij} is a geometrical factor for the j th process reflecting a series/parallel combination of λ_j with other λ_i 's.

In the analysis below, Eq. (6) is simplified to contain only two terms, i.e.,

$$\lambda_{\text{eff}} = \lambda_l + \bar{\lambda}_{g-s} \quad , \quad (7a)$$

or

$$\frac{1}{k_{\text{eff}}} = \frac{1}{k_l} + \frac{1}{\bar{k}_{g-s}} \quad , \quad (7b)$$

where subscripts l and $g-s$ denote lattice and gas/solid interface, respectively; f_{ij} is assumed to be one for all i and j . The new term, $\bar{\lambda}_{g-s}$ (or $1/\bar{k}_{g-s}$), in a sense represents the sum of the thermal resistances from all processes except that due to lattice conduction. With thermal conductivity data on the sphere-pac and high-density, sintered UO_2 , one can calculate $\bar{\lambda}_{g-s}$ directly from Eq. (7a) as a function of temperature and helium fill-gas pressure. These $\bar{\lambda}_{g-s}$ values are then assumed applicable to the sphere-pac Li_2O and $\gamma-LiAlO_2$, with or without irradiation. Using Eq. (7a) again with k_l from high-density, sintered solid breeders, Eqs. (2) and (5), one can calculate the corresponding k_{eff} 's for sphere-pac solid breeders. The k_l 's will, of course, be different for the unirradiated and irradiated Li_2O and $\gamma-LiAlO_2$.

Before discussing the irradiation effects on k_l , it would be of interest to compare the k_{eff} obtained from the two procedures for unirradiated, sphere-pac solid breeders. The results are shown in Figs. 2 and 3 for Li_2O and $\gamma-LiAlO_2$, respectively. Notice that Procedure 1 (i.e., assuming identical k reduction factors for UO_2 , Li_2O , and $\gamma-LiAlO_2$) resulted in higher estimated k_{eff} for Li_2O but lower estimated k_{eff} for $\gamma-LiAlO_2$ than those obtained from Procedure 2, Eq. (7); the reason will become clear later.

Figure 2. Comparison of effective thermal conductivity (k_{eff}) for unirradiated, sphere-pac Li_2O made of high-density ($\sim 100\%$ TD) particles with three different sizes (30, 300, and 1200 μm diameters). Procedures 1 and 2 are, respectively, based on a simplistic approach and Eq. (7) referred to in the text.

Figure 3. Comparison of effective thermal conductivity (k_{eff}) for unirradiated, sphere-pac $\gamma-LiAlO_2$ made of high-density ($\sim 100\%$ TD) particles with three different sizes (30, 300, and 1200 μm diameters). Procedures 1 and 2 are, respectively, based on a simplistic approach and Eq. (7) referred to in the text.

2.3 Irradiation Effects on Lattice Thermal Conductivity (k_L)

The conduction of heat in dielectric solid at low to medium temperatures can generally be regarded either as propagation of anharmonic elastic waves through a continuum, or as interaction between quanta of thermal energy called phonons. At higher temperatures, higher-frequency electromagnetic internal radiation (photons) and thermal conduction by electronic charge carriers may become important. The UO_2 data (Fig. 1) used in this work show that k_L initially decreases with increasing temperature, but the curve turns around at ~ 2000 K, indicating significant conduction contribution from photons and electronic charge carriers all the way up to the UO_2 melting point (3123 K). Thermal conductivity data of Li_2O [2] and γ - $LiAlO_2$ [3,4] over the temperature ranges investigated (473-1173 K for Li_2O , 373-1200 K for $LiAlO_2$), on the other hand, exhibit a continuous reduction of k_L with temperature, even though their rates of change with temperature become small toward the high-temperature ends. Since these experimental temperature ranges match closely with the temperature ranges in which the two solid breeders will be operating in a blanket, phonon conduction appears to be the dominant component; therefore subsequent discussion will focus only on this term.

2.4 Sintered Material

In an ideal lattice with no defects (i.e., interstitials, vacancies, dislocations, etc.), phonons can interact with each other by collision where one has to distinguish between a normal (N-process) and an Umklapp process (U-process) [12]. In both processes a new phonon (three-phonon process) is created by the collision of two phonons. The energy of the new phonon equals the sum of the energies of the colliding phonons (conservation of energy). In the N-process the direction of energy flow is maintained after the collision, but

in the U-process a change of the energy flow direction occurs, thus imposing a resistance to heat flow. At temperatures above the Debye temperature, the majority of the phonon collisions are the U-processes. According to the kinetic theory of gases, an approximate but useful relationship for the phonon conductivity in an ideal lattice is

$$k_{\ell} = \frac{1}{3} C v \Lambda_0, \quad \Lambda_0 > a_0; \quad (8a)$$

$$k_{\min} = \frac{1}{3} C v a_0, \quad \Lambda_0 < a_0, \quad (8b)$$

where C is the heat capacity per unit volume, v is the phonon wave velocity in the solid, Λ_0 is the attenuation length (or mean free path) of the lattice phonon waves, and $k_{\min}$ is the minimum lattice conductivity when the phonon wave length approaches atomic dimensions (a_0). In many solids $k_{\min}$ is not easily revealed because of melting and the other high-temperature conduction components (i.e., photons and electronic charge carriers).

For a non-ideal lattice with defects, phonon scattering will occur at these defects, in addition to the U-processes. These defects in general change the mean free path rather than the other factors in Eq. (8). Similar to the thermal resistance concept described earlier for the overall thermal resistance of a sphere-pac system on a macroscopic level, one can write on an atomic level

$$\frac{1}{\Lambda} = \frac{1}{\Lambda_0} + \sum_i \frac{1}{\Lambda_i}, \quad (9)$$

where Λ_i is the mean free path for phonon scattering by the i th type of defect initially present in an unirradiated material. It has been shown [13] by theory and confirmed by experiment that $\Lambda_0 = \alpha/T$, where α is a constant characteristic of the material, and T is the temperature. The phonon-defect mean free path (Λ_i), on the other hand, is generally independent of temperature.

Ignoring $k_{\min}$ and simple manipulation of Eqs. (8) and (9) show that the lattice thermal conductivity k_L of unirradiated material can be written as

$$k_L = \frac{1}{A + BT} \quad , \quad (10)$$

a functional form observed experimentally for both Li_2O and $\gamma\text{-LiAlO}_2$ (see preceding section).

The effect of neutron irradiation is to introduce additional defects (via radiation damage) into the material. These defects may include displaced atoms, bubbles, voids, and cracks. They give rise to extra sources of phonon scattering, and the effect may be represented by additional mean free paths Λ_j for each of the j th type of defect introduced by irradiation. The total effective mean free path that determines the thermal conductivity of irradiated material is then

$$\frac{1}{\Lambda_{\text{eff}}} = \frac{1}{\Lambda} + \sum_j \frac{1}{\Lambda_j} \quad , \quad (11)$$

where Λ is given by Eq. (9).

The lattice thermal conductivity of irradiated material (k_{irr}) can be expressed in terms of the original unirradiated k_L as

$$k_{\text{irr}} = \alpha_{\text{irr}} k_L \quad , \quad (12)$$

where

$$\alpha_{irr} = \frac{k_{irr}}{k_2} = \frac{1}{1 + \frac{\beta'}{A + BT}} \quad (13)$$

Each of the constants A, B, and β' in Eq. (13) has a well-defined physical meaning. Constant A embodies the mean free path for phonon-defect scattering; the defects are those initially present in an unirradiated material. Constant B embodies the mean free path for phonon-phonon scattering and is characteristic of the crystal structure of the material. Finally, constant β' embodies the mean free path for phonon-defect scattering; the defects are those produced by irradiation. Thus if k_2 and constants A, B, and β' are known, k_{irr} can be readily obtained as a function of temperature.

Experimental information on the Λ_j 's for various defects are not available for any of the three solids (UO_2 , Li_2O , and $\gamma-LiAlO_2$) to permit direct use of Eq. (11). Post-irradiation thermal conductivity measurements [14] of UO_2 irradiated at low temperatures (273-373 K) to low fluences ($1.1-10 \times 10^{21}$ n/m²) showed ~50% reduction from the unirradiated value at low temperatures (<373 K). When the measurements were extended to higher temperatures, annealing stages were observed at 473 and 673 K. Even after annealing at 873 K, some damage still remained in a specimen exposed to 8.7×10^{21} n/m². Another set of annealing experiments of Ross [15] on UO_2 irradiated at 773 K demonstrated the complicated nature of the damage. The results showed that while an 873 K anneal removes most of the damage from the lightly irradiated specimens, the damage in the more heavily irradiated specimens (4.7×10^{21} n/m²) is practically unaffected. The latter responded to higher annealing temperatures,

but the amount of recovery appears to saturate at about 50% over 1173 K. The annealing stages seen at 673 K in lightly irradiated material moves into higher temperatures and divides into two components at higher exposures. Each stage is presumably associated with the annihilation of irradiation-induced lattice defects, but their identifications have not been established.

In a recent study by Hurley and Clinard [16], thermal diffusivities were measured for Al_2O_3 , Y_2O_3 -1% ZrO_2 , MgAl_2O_4 and MgO irradiated at relatively high temperatures (925 and 1100 K) and fast neutron exposures (up to $2 \times 10^{26} \text{ n/m}^2$). Room-temperature measurements showed a 10 to 75% reduction in thermal diffusivity (hence thermal conductivity) for Al_2O_3 , Y_2O_3 -1% ZrO_2 , and MgO . The reduction, however, saturates after 0.5 to $2 \times 10^{26} \text{ n/m}^2$, depending on the irradiation temperature. The single-crystal spinel MgAl_2O_4 , on the other hand, showed little sensitivity to fast neutron irradiation. The single-crystal spinel MgAl_2O_4 has a complex cubic crystal structure. It is also one of the most dimensionally stable insulator materials under neutron irradiation. Compared to the non-cubic, hexagonal Al_2O_3 which swelled 3% $\Delta V/V_0$ after $1 \times 10^{26} \text{ n/m}^2$, MgAl_2O_4 exhibits virtually zero swelling. Transmission electron microscopy [17] showed that in Al_2O_3 , unfaulted interstitial dislocation loops can easily form, leading to a bias for precipitation of interstitials, thus leaving vacancies to form cavities (or voids). Achieving the stoichiometric ratio of vacancies for cavities in MgAl_2O_4 would be more difficult than in Al_2O_3 ; fewer interstitial loops form because of difficult loop nucleation, and these loops remain faulted, thus acting as less effective sinks for interstitials. Recent Ph.D. thesis research by Parker [18] with weak-beam transmission electron microscopy showed that the defects in MgAl_2O_4 are primarily dislocation loops. These loops were found to grow in size with increasing irradiation temperatures from 673 to 823 K, while their densities decrease at

higher temperatures. No precipitation reactions were found in control specimens after ~6 months at 823 K.

The absence of voids, the fewer dislocation loops, and the larger loop sizes all indicate that the irradiation-induced defects in MgAl_2O_4 are less effective scattering centers for phonons, hence the small effect on thermal conductivity. The main point of the above discussion of MgAl_2O_4 is that in treating irradiation effects on the lattice thermal conductivity of solid breeders, one needs to consider the difference in solid-breeder crystal structures. The crystal structure of Li_2O resembles that of UO_2 ; the metal and oxygen atoms occupy reverse positions (corners, faces, interstices) in a fluorite-type (UO_2) unit cell, however. Both swelling (up to ~6% $\Delta V/V_0$ after 1 at.% ^6Li burnup) [19,20] and evidence of helium bubbles [21] have been observed in the Li_2O pellets irradiated in Argonne's Experimental Breeder Reactor-II (EBR-II). Based on these results, it seems reasonable to assume a 50% reduction in the Li_2O lattice thermal conductivity after $0.5\text{--}2 \times 10^{26} \text{ n/m}^2$ fast neutron exposure. The $\gamma\text{-LiAlO}_2$, on the other hand, has a tetragonal crystal structure [5]. This type of structure is not the same as that of a spinel, but $\gamma\text{-LiAlO}_2$ is well known for its thermal stability. Under neutron irradiation, the FUBR-1A [19] experiment in EBR-II showed essentially zero swelling at three irradiation temperatures (773, 973, and 1173 K) to a burnup comparable to that of Li_2O . The TRIO-1 experiment [22] in the Oak Ridge Research Reactor (ORR) irradiated at 673-973 K to ~0.2 at.% ^6Li burnup showed excellent dimensional stability, even for the relatively low-density (~65% TD) $\gamma\text{-LiAlO}_2$. A 10% reduction in thermal conductivity has been inferred from the thermocouple readings and the preliminary post-test heat transfer analysis [23]. Based on these results, a 25% reduction in the $\gamma\text{-LiAlO}_2$ lattice thermal conductivity is assumed after $0.5 - 2 \times 10^{26} \text{ n/m}^2$ fast neutron exposure. The

choice of these reduction factors may be conservative, but they at least reflect the difference of radiation damage in Li_2O and $\gamma\text{-LiAlO}_2$.

Rearranging Eq. (13) gives

$$\beta' = \left(\frac{1}{\alpha_{\text{irr}}} - 1 \right) (A + BT) \quad . \quad (14)$$

Recalling that $\alpha_{\text{irr}} = k_{\text{irr}}/k_\lambda$ and with the reduction factors assumed above (α_{irr} equal to 0.5 and 0.75, respectively, for Li_2O and $\gamma\text{-LiAlO}_2$), the values of β' at $T = 400$ K are 9.34×10^{-2} for Li_2O and 8.65×10^{-2} for $\gamma\text{-LiAlO}_2$. The final expression for k_{irr} after $0.5 - 2 \times 10^{26} \text{ n/m}^2$ is

$$k_{\text{irr}}(P) = \frac{k_\lambda}{1 + \beta'/(A + BT)} f(P) \quad \text{W/m-K} \quad (15)$$

for sintered Li_2O and $\gamma\text{-LiAlO}_2$, where $f(P)$ is the porosity function given before for the unirradiated materials, and different values of A , B , and β' from Eqs. (2), (5), and (14) must be used for Li_2O and $\gamma\text{-LiAlO}_2$.

Figures 4 and 5 show the calculated thermal conductivities for irradiated, sintered Li_2O and $\gamma\text{-LiAlO}_2$, respectively, at three densities (70, 85, and 98% dense). Compared to k_0 of unirradiated solid breeders given by Eqs. (2) and (5), the reduction of k_{irr} is larger at low temperatures, becoming less toward high temperatures.

2.5 Sphere-Pac Material

As discussed earlier for unirradiated, sphere-pac solid breeders, neutron irradiation should cause most change in k_λ , while affecting very little the other heat transport processes in a sphere-pac system. All one needs to do with the sphere-pac solid breeders is to replace λ_λ (or k_λ) in Eq. (7) by

Figure 4. Irradiation effect on thermal conductivity of sintered Li_2O .

Figure 5. Irradiation effect on thermal conductivity of sintered $\gamma\text{-LiAlO}_2$.

λ_{irr} (or k_{irr}), assuming $\bar{\lambda}_{g-s}$ (or $\bar{k}_{g-s}$) remains unchanged from the unirradiated values. The series combination of thermal resistances, or the parallel combination of thermal conductivity and conductance in a sphere-pac system, can be mathematically written as

$$k_{eff}^{irr} = \alpha_{irr}^{sp} k_{eff}^{irr} \quad , \quad (16)$$

where

k_{eff}^{irr} , k_{eff}^{irr} = effective thermal conductivities for unirradiated and irradiated sphere-pac material, respectively, and

$$\alpha_{irr}^{sp} = \frac{\alpha_{irr}}{\alpha_{irr} + \gamma(1 - P_{sp})(1 - \alpha_{irr})} \quad , \quad (17)$$

where $P_{sp} = 1 - SPF = 13\%$, for a solid packing fraction (SPF) of 87%, and $\gamma = k_{eff}/k_s$ (i.e., the ratio of thermal conductivities of unirradiated, sphere-pac, and sintered material).

Notice that the gas-solid heat transport effect is implicitly included in γ . For a given fill-gas pressure, γ varies with temperature; for a given temperature, γ varies with fill-gas pressure. Final results of k_{eff}^{irr} are displayed in Figs. 6 and 7, respectively, for Li_2O and $\gamma-LiAlO_2$ made of high-density (~100% TD) spherical particles with three sizes (30, 300, and 1200 μm diameters). Compared to the unirradiated k_{eff} , the irradiated k_{eff} is 12 to

Figure 6. Irradiation effect on thermal conductivity of sphere-pac Li_2O .

Figure 7. Irradiation effect on thermal conductivity of sphere-pac $\gamma\text{-LiAlO}_2$.

19% lower for Li_2O and 6 to 12% lower for $\gamma\text{-LiAlO}_2$ at 1000 K. Part of the differences in percent reduction resulted from the different reduction factors (50 versus 25%) assumed for Li_2O and $\gamma\text{-LiAlO}_2$; part also depends upon the relative magnitudes of λ_ℓ and $\bar{\lambda}_{g-s}$, i.e.,

$$\frac{\Delta k_{\text{eff}}}{k_{\text{eff}}} = 1 - \frac{1 + (\lambda_\ell / \bar{\lambda}_{g-s})}{(1 + F \lambda_\ell / \bar{\lambda}_{g-s})}, \quad (18)$$

where k_{eff} is that of the unirradiated system, Δk_{eff} is the difference of k_{eff} between unirradiated and irradiated systems, and F is the reduction factor of k_ℓ due to irradiation.

The relative magnitudes of λ_ℓ and $\bar{\lambda}_{g-s}$ (or k_ℓ and $\bar{k}_{g-s}$) also explain the difference in k_{eff} estimated for unirradiated sphere-pac Li_2O and $\gamma\text{-LiAlO}_2$ noted earlier in Figs. 2 and 3. Equation (7) shows that when $\bar{\lambda}_{g-s} \gg \lambda_\ell$, k_{eff} will be dominated by $\bar{\lambda}_{g-s}$. Irradiation effects, which change only λ_ℓ , will be minor on k_{eff} . The k_ℓ of Li_2O is a factor of ~ 2 higher than that of $\gamma\text{-LiAlO}_2$, so that λ_ℓ of Li_2O is $\sim 1/2$ of that of $\gamma\text{-LiAlO}_2$. Since the same $\bar{\lambda}_{g-s}$ is used for both systems, this means λ_ℓ is relatively more important to λ_{eff} (and k_{eff}) for $\gamma\text{-LiAlO}_2$ than Li_2O . The same kind of argument may be used to explain the TRIO-1 thermal conductivity results [23]. Since the $\gamma\text{-LiAlO}_2$ pellets in TRIO-1 contain a rather high fraction (~ 0.35) of porosity and their microstructure remain unchanged during irradiation, the thermal resistance, λ_p , due to the as-fabricated pores should remain large (if not dominant) during irradiation. Irradiation-induced defects will increase λ_ℓ somewhat, but if λ_ℓ is substantially lower than λ_p , irradiation effects on thermal conductivity of low-density (but microstructurally stable) material should be minor.

3. DISCUSSION

Thermal conductivities of packed beds have been investigated in other non-fusion technology areas, fluidized and packed beds, for example. Experimental results from one such investigation [24] of a packed bed of spherical particles with stagnant interstitial fluid are shown in Figs. 8 to 10. Notations in these figures, k_B , k_g , and k_s , are the thermal conductivities of the bulk (equivalent to k_{eff} in this paper), gas, and solid, respectively. For sphere-pac solid breeder blankets, the fill gas is generally helium, with a gas thermal conductivity given by [25]:

$$k_g = 3.366 \times 10^{-3} T^{0.668} \quad \text{W/m-K} \quad (19)$$

From Eq. (19) one can calculate k_g of helium as a function of temperature. From Eqs. (2) and (5) the k_s (or k_o) of Li_2O and $\gamma\text{-LiAlO}_2$ can be calculated. Values of k_B (or k_{eff}) for unirradiated, sphere-pac solid breeders can be read directly from Figs. 2 and 3. Finally, the values of k_B and k_g for unirradiated, sphere-pac UO_2 can be obtained readily from Fig. 1. Calculated ratios of k_s/k_g and k_B/k_g for UO_2 , Li_2O and $\gamma\text{-LiAlO}_2$ at helium fill-gas pressures of 1, 2, and 6 atm are shown in Figs. 8, 9, and 10, respectively. Excellent agreements were found between the calculated values and the curve corresponding to a porosity fraction $\epsilon = 0.163$ for the 6-atm case (Fig. 10), whereas lower k_B/k_g values were calculated for the 1- and 2-atm cases (Figs. 8 and 9).

In deriving Eqs. (13) and (17) for irradiation effects on thermal conductivity, a parameter β' was introduced and determined from the low-temperature reduction factor observed in many dielectric solids. The form of the equa-

Figure 8. Comparison of calculated thermal conductivities (at 1 atm He fill gas pressure) with data on a packed bed of spherical particles with stagnant interstitial fluid.

Figure 9. Comparison of calculated thermal conductivities (at 2 atm He fill gas pressure) with data on a packed bed of spherical particles with stagnant interstitial fluid.

Figure 10. Comparison of calculated thermal conductivities (at 6 atm He fill gas pressure) with data on a packed bed of spherical particles with stagnant interstitial fluid.

tions suggests that α_{irr} and α_{irr}^{sp} increase towards unity as temperature increases from 400 K. Thus the effect of irradiation on thermal conductivity is less drastic at high temperatures. Notice also that α_{in}^{sp} in general is larger than α_{irr} because irradiation affects mainly the solid (i.e., lattice) component of the conductivity in the sphere-pac case. However, the absolute values of k_{eff}^{irr} are still lower than those of k_{irr} because k_{eff} is lower than k_L for unirradiated materials.

There are reasons to believe that the thermal conductivity estimates will be conservative. In fact, the effect of irradiation on k_L and k_{eff} may be even less at higher temperatures. The β' in Eq. (13) assumed constant in this work will in fact decrease with increasing temperature due to more effective annealing of the damage induced by irradiation. The α_{irr} and α_{irr}^{sp} will increase towards unity even faster than indicated by Eqs. (13) and (17). Thermal and radiation-enhanced sinterings could further improve the thermal conductivities at high temperatures.

Thermal stability tests [26], conducted for 8 h at 1173 K on 85%-dense Li_2O and $\gamma-LiAlO_2$, showed that the latter is stable, but the grains in Li_2O grew from a 6- μm average to 60-100 μm . Since grain growth generally occurs concurrent with sintering, pore closure seems highly likely in Li_2O over a relatively short period at 1173 K. The Li_2O grain growth results in the FUBR-1A irradiation experiment [27] indicated a factor of 2 growth in grain size at 973 K, and a factor of 6 growth at 1173 K, both after a fast neutron exposure of $\sim 6.3 \times 10^{25} \text{ n/m}^2$. In the same experiment, the $\gamma-LiAlO_2$ exhibited essentially no grain growth, even though the starting grain size ($< 1 \mu m$) is considerably smaller than that of Li_2O . In the OG-5 irradiation experiment [28] in ORR, the major fraction of pores in the low-density (68%-dense) Li_2O became closed pores for irradiation temperatures $> 1123 \text{ K}$ after a fast fluence of

$2 \times 10^{25} \text{ n/m}^2$; the pores remained open for irradiation temperatures $< 1023 \text{ K}$. The pores in the low-density (70% TD) LiAlO_2 , on the other hand, remained open up to 1273 K , after a fast neutron exposure of $2.7 \times 10^{25} \text{ n/m}^2$. Effects of capture versus fast-neutron displacement damages on sintering are not easily discernible in the OG-5 experiment, primarily because highly depleted ^6Li was used which burned up at much lower fluences. The $\gamma\text{-LiAlO}_2$ data appear to corroborate the data of Wiswall and Wirsing [29] between 1248 and 1273 K , although the latter revealed some powder agglomeration.

The various ex-reactor and in-reactor results indicate that grain growth and sintering are more likely to occur in Li_2O ($> 1023 \text{ K}$) than in $\gamma\text{-LiAlO}_2$ ($> 1273 \text{ K}$). Besides the temperature dependence, solid-state sintering generally increases with time t at temperature ($\propto t^m$, $m < 1$), and with decreasing particle size r ($\propto r^{-n}$, $n \lesssim 1$). For the sphere-pac solid breeders, stage-I sintering (i.e., neck growth) appears likely to occur at sufficiently high temperatures ($> 0.6 T_m$ where T_m is the melting point 1706 and 1883 K , respectively, for Li_2O and $\gamma\text{-LiAlO}_2$) between the fine particles (i.e., with $30\text{-}\mu\text{m}$ diameters). The net result of this neck growth is to reduce the contact resistance between particles, resulting in an increase of k_{eff} at high temperatures. Quantitative assessment of the sintering effect on k_{eff} cannot be made at this time because of the lack of solid-breeder sintering data.

The technological importance of thermal conductivity (k and k_{eff}) to fusion solid-breeder blankets has been emphasized at the beginning of this paper. For solid breeder blankets cooled with pressurized water at a neutron wall loading of 5 MW/m^2 , the calculations of Misra [30] showed the effects of thermal conductivities on volume fractions of solid breeder, structure, and coolant in an outboard blanket module of 50-cm depth (Table 1). Even though these results are based on a one-dimensional unit-cell calculation, a substan-

TABLE 1. THERMAL CONDUCTIVITY EFFECTS ON BLANKET VOLUME FRACTIONS
OF SOLID BREEDER, STRUCTURE, AND COOLANT^a

	Li ₂ O				γ -LiAlO ₂		
	Front ^b	Middle	Rear		Front	Middle	Rear
SOLID BREEDER							
Unirradiated k	0.58	0.68	0.93		0.58	0.68	0.93
Irradiated k	0.45	0.57	0.91		0.55	0.65	0.92
STRUCTURE (HT-9)							
Unirradiated k	0.21	0.18	0.06		0.22	0.18	0.05
Irradiated k	0.30	0.25	0.07		0.23	0.20	0.06
COOLANT (H ₂ O)							
Unirradiated k	0.21	0.14	0.01		0.20	0.14	0.02
Irradiated k	0.25	0.18	0.02		0.22	0.15	0.02

^aResults from Ref. [30].

^bFront, middle, and rear refer to regions near first wall, middle, and rear of the blanket.

tial decrease in solid-breeder volume fraction and a concomitant increase in structure and coolant volume fractions are noted, especially for Li_2O near the first wall region, where both nuclear heating and tritium production are high. Reduction of breeder volume fraction in a $\gamma\text{-LiAlO}_2$ blanket is not as drastic as Li_2O , partly because the irradiation effect on k is smaller and partly because the assumed design temperature window (623 to 1273 K) is wider than that of Li_2O (683 to 1073 K).

Decreased volume fraction of solid breeders and increased volume fractions of structure and coolant due to change in solid-breeder thermal conductivity have serious implications on the blanket tritium breeding potential. Among the various solid-breeder candidate materials, Li_2O is the only one that can achieve a tritium breeding ratio greater than one (based on unirradiated properties) without the use of neutron multipliers. In a sphere-pac blanket design with pressurized water coolant, the tritium breeding ratio based on an unirradiated k_{eff} of 3.4 W/m-K and a breeder volume fraction of 67% is 1.14; the tritium breeding ratio with a k_{eff} of 1.56 W/m-K and a breeder volume fraction of 53% is calculated to be 1.05.

4. SUMMARY

Thermal conductivities (k and k_{eff}) for sintered and sphere-pac Li_2O and $\gamma\text{-LiAlO}_2$ have been estimated with and without irradiation effects. The estimations are believed to be somewhat conservative because (1) the thermal resistance $\bar{\lambda}_{g-s}$ in the UO_2/He system could be higher than those for $\text{Li}_2\text{O}/\text{He}$ and $\gamma\text{-LiAlO}_2/\text{He}$ (i.e., difference in the accommodation coefficients); (2) the β' , which incorporates irradiation effects on thermal conductivities, is likely to decrease with increasing temperature due to more effective annealing of radia-

tion damage; and (3) thermal and radiation-enhanced sinterings have not been included, which should result in increases of k and k_{eff} at high temperatures.

Impacts of thermal conductivities on solid-breeder blanket performance are also briefly discussed. Their technological importance is illustrated by examples on blanket solid-breeder volume fractions, which translate into blanket tritium breeding potential. Experimental determinations of solid-breeder thermal conductivities are highly warranted because of their significance to a viable fusion solid-breeder blanket.

ACKNOWLEDGMENT

The authors express their thanks to Drs. D. L. Smith and F. A. Nichols for their review and comments, to Dr. R. G. Clemmer for discussion of the TRIO-1 thermal conductivity results, and to Lee Ann Ciarlette for preparing this manuscript.

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