

Experimental measurements of the effective thermal conductivity of a lithium titanate (Li_2TiO_3) pebbles-packed bed

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Abstract

Tritium breeding materials, which have the ability to react with neutrons and produce tritium, are required to fuel fusion reactions. Tritium is produced inside the fusion blanket by neutron irradiation of lithium ceramics (tritium breeders). Using the lithium ceramics in form of pebbles-packed bed is a promising concept for fusion blankets, and worldwide efforts have been dedicated to its R&D. Thermal properties of lithium ceramic pebble beds have a significant impact upon the thermal performance of the fusion blanket. Specifically, the effective thermal conductivity of lithium ceramic pebble beds is important for the design and analysis of fusion blankets. The experimental apparatus of this study was designed and built based on the principles of the steady state and axial heat flow methods in order to conduct the required measurements. The objective of this study is to measure the effective thermal conductivity of a Li_2TiO_3 pebble bed as a function of the average bed temperature. The pebble bed has pebbles of 1.7–2.0 mm diameter and a packing fraction of 61%. Helium at atmospheric pressure was used as a filling gas. The experimental results showed that the effective thermal conductivity decreased from 1.40 to 0.94 W/m K with the increase of the average bed temperature from 50 to 500 °C. The results presented in this work will help to create a database of the effective thermal conductivity of Li_2TiO_3 pebble beds which can be used for the design and analysis of fusion blankets.

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1. Introduction

Lithium ceramics have been selected as tritium breeders in many designs of fusion blanket. Lithium ceramic breeding blanket is considered one of the most promising fusion blankets and worldwide efforts have been devoted to its R&D. In the breeding blanket, lithium ceramics are used as tritium breeders and beryllium is used for neutron multiplication. The candidate ceramics for this breeding blanket are lithium orthosilicate (Li_4SiO_4), lithium titanate (Li_2TiO_3), lithium zirconate (Li_2ZrO_3) and lithium oxide (Li_2O). The material-form options for the tritium breeders in a fusion blanket are sintered block and pebble bed. The sintered blocks have some problems such as uncertainty of thermal conductance at breeder/structure interface and cracking due to high thermal stresses. Accurate prediction of the thermal conductance at breeder/structure interface is very difficult due to some factors including breeder cracking, cracked fragment relocation, sintering, creep, and swelling. These factors work to change the gap between solid breeder and structural materi-

als during blanket operation. To reduce the uncertainty in the interface thermal conductance, the pebble bed was proposed as a material form for the tritium breeders and neutron multipliers in many fusion blankets.

Pebble beds are used in many applications, such as nuclear reactor fuel rods, thermal insulation, heat exchangers, chemical catalysts, automotive catalytic converters, and recently, fusion blankets. The pebble bed consists of two phases: solid phase which is the pebbles and the filling gas. Using pebble beds in fusion blanket has several advantages. First, bed characteristics (such as size and shape of pebbles, pressure of cover gas, and bed packing) can be tailored to obtain the required thermal characteristics. Second, the effective thermal conductivity can be controlled by adjusting bed characteristics. Third, tritium produced in the pebble beds can be easily removed by the purge gas through the pores' network. Tritium extraction from the lithium ceramics, by means of a purge gas, has been successfully used in several previous experiments. Fourth, loading of pebbles into complex geometries can be achieved. Also, mixing of ceramic breeder and neutron multiplier can be implemented to improve tritium breeding. In addition, with small pebbles, the temperature gradient and, therefore, the thermal stresses in the pebbles should be insignificant to cause pebble cracking.

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Tritium breeders should have good thermal properties, especially thermal conductivity, as well as satisfactory tritium breeding characteristics. The thermal properties of tritium breeders are important for the blanket design and development because the nuclear heat produced by the fusion has to be transferred to the coolant through the blanket materials (e.g. tritium breeders, neutron multiplier, structure, etc.). Therefore the effective thermal conductivity and interface thermal conductance of the lithium ceramic pebble beds are key thermal properties for the fusion blankets. In order to study the heat transfer in the blanket, the effective thermal conductivity and the interface thermal conductance of the lithium ceramics pebble beds have to be well measured and characterized. A database of these two properties is required for the blanket R&D. The effective thermal conductivity, k_{eff} , of pebble beds depends on several parameters. Some of these parameters have significant impact on k_{eff} such as thermal conductivity of pebbles and filling gas, gas pressure, bed porosity, and pebbles' deformation. Other parameters have less impact on k_{eff} such as pebbles' size, shape, and surface roughness [1]. The design of the breeding blankets requires reliable database of the thermal properties of the ceramic breeder pebble beds.

The objective of this study is to measure the effective thermal conductivity of a Li_2TiO_3 pebble bed as a function of the average bed temperature in the range of 50–500 °C. The experimental apparatus, used to conduct the required measurements, is presented in the following section. After that, three models of effective conductivity of pebble beds are summarized. Then the predictions of these models are compared with the measured values of k_{eff} in Section 5. Then the paper is concluded with the conclusions and future work.

2. Experimental apparatus

The experimental apparatus of this work was designed and built to measure the effective thermal conductivity, k_{eff} , of different lithium ceramic pebble-packed beds. Its design is based on the principles of the steady state and the axial heat flow methods. The main concept of the experiment is to generate a temperature gradient across the pebble bed and create 1D heat transfer region around the bed's centerline. Then heat flux and temperatures across the pebble bed are measured to calculate the effective thermal conductivity using Fourier's law of heat conduction after steady state conditions are reached. The experimental apparatus, shown in Fig. 1, consists of the following main parts: pebbles container, heater, heating block, heat flux smoother, heat sink, thermocouples, heat flux sensors, heat flux meter, helium chamber, radiation shield, and vacuum chamber. The heater is located above the pebble bed and a heat sink is placed under the pebble bed in order to generate an axial heat flow through the pebble bed. An 800-W band heater, with maximum operating temperature of 760 °C and watt density of 47 W/in.², was used. The heat flows from the heater to the heating block (copper disc), and flows downward through a heat flux smoother to the pebble bed. Then the heat flows from the pebble bed to the heat-flux-sensors disc, then to the heat flux meter, and eventually to the heat sink. The heat flux smoother, made of copper disc, is directly placed above the pebble bed in order to provide a uniform surface temperature.

The heat flux, through the pebble bed, is measured by two heat flux sensors. These two sensors are welded to a stainless steel disc, forming the heat-flux-sensors disc. The heat-flux-sensors disc is placed at the bottom end of the pebbles bed. The pebbles container is a hollow cylinder with inner diameter of 89 mm, outer diameter of 107 mm and height of 50 mm (see Fig. 2). The container was manufactured from glass–mica ceramic, which has a low thermal conductivity (0.042 W/m K). Five profile thermocouples (Ø1.60 mm probe diameter) are

used to measure the temperature profile of the pebble bed. These profile thermocouples can measure the temperature at two sensing points along their probe. The thermocouples are secured in their locations by having array of holes at the pebbles' container. Fig. 2 shows the distribution of the thermocouples inside the pebble bed and the small solid circles (10 mm from the centerline) indicate the locations of the two sensing points in each thermocouple. The thermocouples are type-K with special limits of error: ± 1.1 °C or $\pm 0.4\%$ T (which is greater). The measured temperatures and heat flux are captured by a data acquisition unit, which is connected to a personal computer to store all the experimental measurements. The heat flux meter (HFM) was used to obtain a second independent measurement of the heat flux. The main concept of this HFM is to create a temperature gradient across a reference material (has well-known thermal conductivity) and then Fourier's law of 1D-heat conduction (Eq. (1)) is used to calculate the heat flux. The heat flux meter consists of solid cylinder made of stainless steel 316 (as a reference material) and four thermocouples. The heat sink consists of a copper solid cylinder and a copper tube, where cold water is circulated. The circulating cold water enables the heat sink to provide a uniform low temperature (16–20 °C) at the upper surface of the heat sink. The test article is placed inside a stainless steel chamber (helium chamber in Fig. 1) to control the pressure and type of the used gas. When the vacuum pressure, inside the helium chamber, reaches a value of less than 1 mT, the helium chamber is fed with helium and kept at atmospheric pressure. A Pyrex bell jar was used to form a vacuum chamber around the helium chamber. The space between the helium chamber and the bell jar is evacuated by a mechanical pump in order to eliminate any convective heat loss. In addition a radiation shield is placed between the helium chamber and the vacuum chamber in order to minimize the heat loss by radiation. The radiation shield has a can-shape and is made of polished stainless steel sheet. The reflective surfaces of the radiation shield have a significant role in reducing the heat loss by radiation. At the beginning of the experimental runs, the heater and mechanical pump are turned on simultaneously to remove the moisture and any outgases from the helium chamber.

3. Data analysis

For each experimental run, the temperatures and heat flux across the pebble bed are monitored until the steady state condition is reached. Then the measured data are stored in a personal computer and used to calculate the effective thermal conductivity, k_{eff} . Calculation of k_{eff} by the steady state method requires the knowledge of the heat flux, q , through the bed, and temperature difference, ΔT , across a thickness, Δx . Then Fourier's law (in 1D-heat conduction) is used to calculate k_{eff} using the following equation:

$$q = -k_{\text{eff}} \frac{\Delta T}{\Delta x} \quad (1)$$

The steps of calculating k_{eff} can be summarized as follows:

1. Every thermocouple measures temperature at two different points (at the same horizontal level, see Fig. 2). When the difference in the two measured temperatures of the same thermocouple is smaller than the error limit (± 1.1 °C), the average of the two values are calculated and used to draw the bed thermal profile, otherwise the data is rejected. This check is made for the five thermocouples, located inside the pebble bed. Then these five average temperatures are used to draw the bed thermal profile (see Fig. 3).
2. Temperature difference, ΔT , across the bed is the difference between temperature of TC1 and temperature of TC5 and the thickness, Δx , is 40 mm (see Fig. 2).

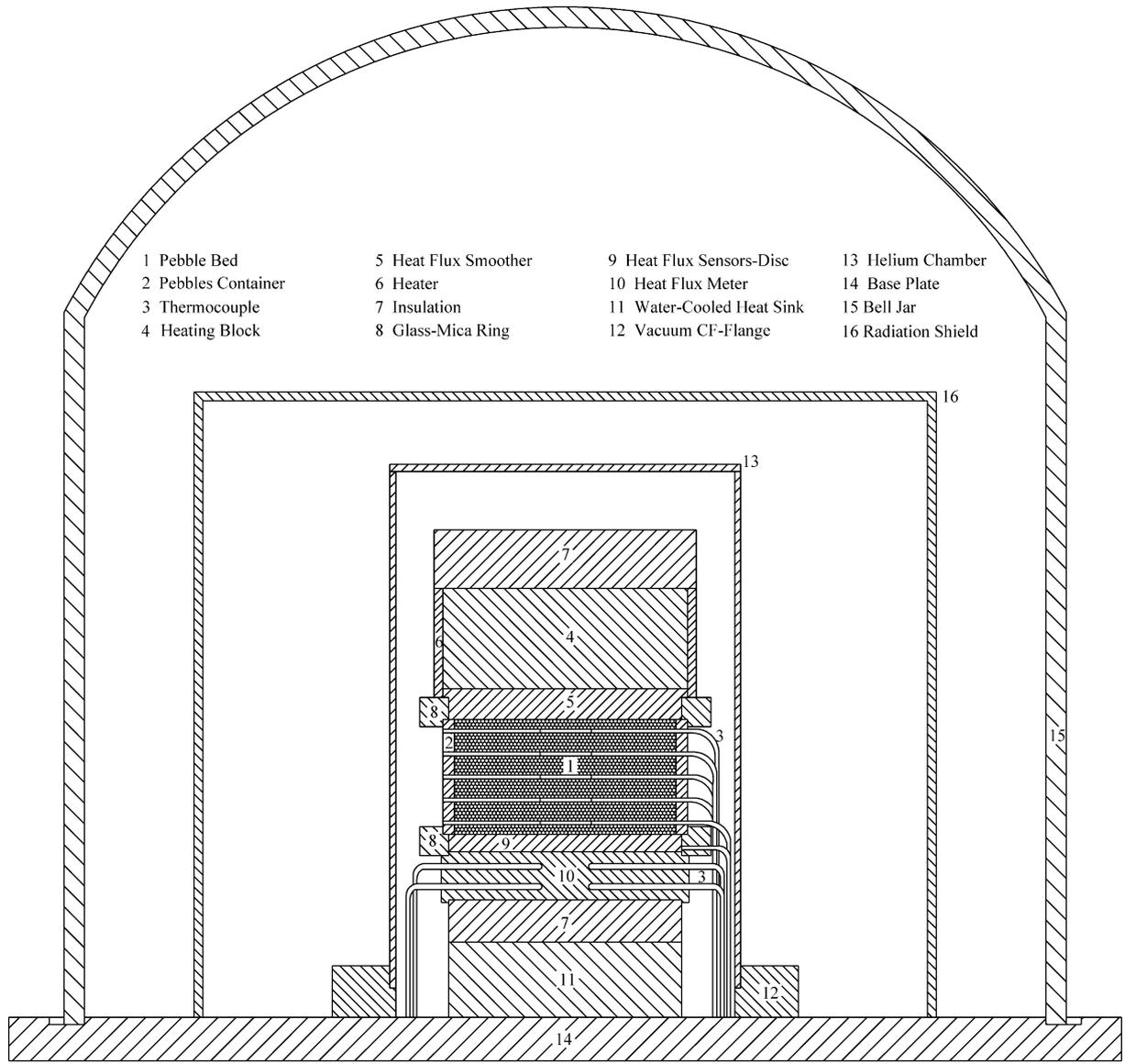


Fig. 1. Schematic drawing of the experimental apparatus.

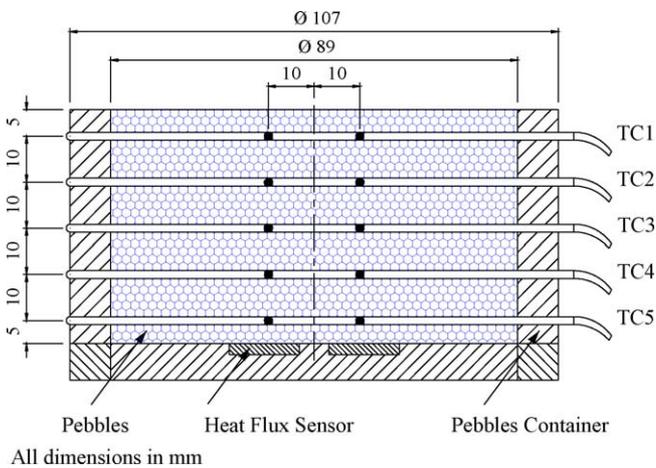


Fig. 2. Distribution of the thermocouples inside the pebble bed.

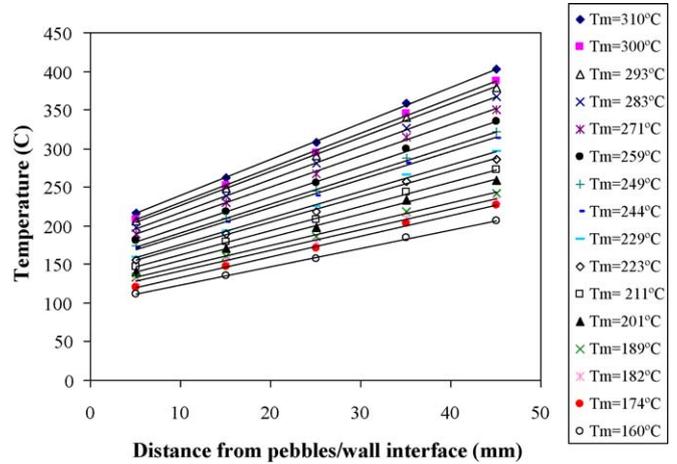


Fig. 3. Thermal profile of the Li₂TiO₃ pebble bed at different temperatures.

3. The heat flux, q , is taken as the average of two readings of the heat flux sensors.
4. Eq. (1) is used to calculate k_{eff} .

4. Models of effective conductivity of pebble beds

Several models have been presented, in the past, to predict the effective thermal conductivity of pebble beds. Three of these models are selected for the purpose of comparison with the present experimental results.

4.1. Schlunder, Zehner, and Bauer model

The Schlunder, Zehner, and Bauer (SZB) model [2–5] is widely used to predict the effective thermal conductivity of pebble beds. This model can be used for beds of pebbles of any shape and size distribution, but having the same thermal conductivity. In this model, the bed is simulated by a standard cell, which contains two contacting pebbles. The SZB model takes into account the conduction and radiation effects throughout the bed. The model does not assume point contact between pebbles; it considers the effect of contact area size. The SZB model considers three paths for the heat flow. The first path represents molecular conduction and radiation through the gas. The second path represents conduction through both the solid and the gas phases with radiation between pebbles surfaces. The third path represents the solid–solid conduction. Based on parallel resistances, the bed effective thermal conductivity, k_{eff} , is calculated using the following correlation [2]:

$$\frac{k_{\text{eff}}}{k_g} = (1 - \sqrt{1 - \varepsilon}) \left[\frac{\varepsilon}{(\varepsilon - 1) + \frac{k_g}{k_D}} + \varepsilon \frac{k_R}{k_g} \right] + \sqrt{1 - \varepsilon} \left[\phi \frac{k_s}{k_g} + (1 - \phi) \frac{k_{\text{SO}}^*}{k_g} \right] \quad (2)$$

$$\frac{k_{\text{SO}}^*}{k_g} = \frac{2}{a} \left[\frac{B \left(\frac{k_s}{k_g} + \frac{k_R}{k_g} - 1 \right) \frac{k_g}{k_D} \ln \frac{\left(\frac{k_s}{k_g} + \frac{k_R}{k_g} \right) \frac{k_g}{k_D}}{B \left[1 + \left(\frac{k_g}{k_D} - 1 \right) \left(\frac{k_s}{k_g} + \frac{k_R}{k_g} \right) \right]} - \frac{B - 1}{a} \frac{k_g}{k_D} + \frac{B + 1}{2B} \left(\frac{k_R}{k_D} - B \left[1 + \left(\frac{k_g}{k_D} - 1 \right) \frac{k_R}{k_g} \right] \right) \right] \quad (3)$$

where

$$a = \left[1 + \left(\frac{k_R}{k_g} - B \frac{k_D}{k_g} \right) \frac{k_g}{k_s} \right] \frac{k_g}{k_D} - B \left(\frac{k_g}{k_D} - 1 \right) \left(1 + \frac{k_R}{k_g} \frac{k_g}{k_s} \right) \quad (4)$$

where k_g is thermal conductivity of gas, ε is the bed porosity, k_R is the equivalent thermal conductivity due to radiation, k_s is the thermal conductivity of pebble, ϕ is the pebble-to-pebble contact area, and k_D is the equivalent thermal conductivity due to molecular heat flow. The deformation factor, B , can be calculated as follows:

$$B = C \left(\frac{1 - \varepsilon}{\varepsilon} \right)^m \quad (5)$$

Both C and m should be determined from the experimental data. The amount of heat transfer by radiation depends on the

absolute temperature, T , of the packing, the radiation characteristics of the pebbles, and the packing geometry. For small temperature gradients, the radiative heat transfer was described by Damkohler’s equivalent thermal conductivity, k_R , as follows [3]:

$$\frac{k_R}{k_g} = \frac{0.04\sigma}{(2/\gamma - 1)} \left(\frac{T}{100} \right)^3 \frac{X_R}{k_g} \quad (6)$$

σ is the Stefan–Boltzmann constant and γ is the radiative emissivity. X_R is the effective radiation length that characterizes the distance between the pebbles’s surfaces:

$$\frac{k_g}{k_D} = 1 + \frac{2\lambda}{X_D} \left(\frac{2}{\alpha} - 1 \right) \quad (7)$$

$$X_R = R_F d, \quad X_D = D_F d \quad (8)$$

where d is the pebble diameter, R_F is the shape factor for the interstitial energy transport by radiation, D_F is the shape factor for the interstitial energy transport by molecular flow, α is the accommodation coefficient, and λ is the mean free path of the gas molecules.

4.2. Shapiro et al. model

In 2004 Shapiro et al. [6] published their model for predicting effective thermal conductivity of ceramic powder packed beds. In this model, the pebble bed is assumed to be composed of identical pebbles that contact each other over area of πa^2 . The heat is transferred through the pebble bed via the solid pebbles, through the gas and via the contact areas. The model was simplified by assuming the pebbles to be cylinders of radius d and height L , aligned in the same x -direction and contacting each other through a contact point of thickness δ and circular area of radius a . The effective thermal conductivity, k_{eff} , of a pebble packed bed was defined by

$$k_{\text{eff}} = \frac{4}{\pi} k_s \frac{1 + \delta/L}{1 + \frac{\delta}{L} \left(\frac{k_s}{k_a} \right) \left(\frac{1}{1 - (a/d)^2} \right) \left(\frac{k_\delta}{k_a} + \frac{1}{(d/a)^2 - 1} \right)^{-1}} \quad (9)$$

where δ is the gap between pebbles, k_s is the thermal conductivity of solid pebble, and k_a is the thermal conductivity of solid pebble within contact region.

$$k_\delta = k_g \left[\frac{B}{p\delta} + 1 \right]^{-1} \quad (10)$$

$$B = \frac{4\nu}{\nu + 1} \frac{2 - \alpha}{\alpha} \lambda p_0 Pr^{-1} \quad (11)$$

where ν is the specific heat ratio of the gas, λ is the mean free path at atmospheric pressure, Pr is the Prandtl number, α is the accommodation coefficient, k_g is the gas thermal conductivity

at atmospheric pressure, p is the gas pressure, and p_0 is the atmospheric pressure. The values of gap thickness, δ , and contact area size, a , are selected to obtain the best possible fit with the experimental data.

4.3. Okazaki et al. model

Okazaki et al. [7] published their model, in 1981, to predict the effective thermal conductivity of pebble beds. In this model, the variations in pebble size and pebble thermal conductivity were considered. The authors proposed a unit cell model, where the heat flux through the pebble bed is divided into: (i) heat flux through the gaseous phase with a relative surface A_v and (ii) heat flux through the solid phase with a relative surface A_s . The heat flux is determined by the contributions of the heat transfer through the gas near the contact point and the heat transfer through the pebble. For pebble beds formed by spheres that have similar size, the relation between the coordination number, N , and the porosity, ε , was given as

$$N = 13.84 - \sqrt{232\varepsilon - 57.18} \quad (12)$$

Then the effective number of contact points ($n = N/6$) on a hemisphere can be calculated. This unit cell model consists of a solid part and a macro-void part. The fractional areas of the solid part and the macro-void part were calculated as follows:

$$A_s = \frac{3(1 - \varepsilon)}{2}, \quad A_v = \frac{3\varepsilon - 1}{2} \quad \text{for } \varepsilon \geq 0.33 \quad (13)$$

The effective thermal conductivity of the solid part, k_{es} , can be calculated by

$$\frac{k_{es}}{k_g} = 2n \left(\frac{\kappa}{\kappa - 1} \right)^2 \times \left\{ \ln[\kappa - (\kappa - 1)\cos\theta_0] - \left(\frac{\kappa - 1}{\kappa} \right) (1 - \cos\theta_0) \right\} \quad (14)$$

where

$$\kappa = \frac{k_s}{k_g}, \quad \theta_0 = \sin^{-1} \sqrt{\frac{1}{n}} \quad (15)$$

The effective thermal conductivity, k_{eff} , is then formulated as follows:

$$\frac{k_{eff}}{k_g} = A_v k_s + A_s k_s \left(\frac{k_{es}}{k_g} \right) \quad (16)$$

5. Results and discussion

The effective thermal conductivity, k_{eff} , of a Li_2TiO_3 pebble bed was experimentally measured in this study. More than 120 experimental tests (runs) were performed to report the values of k_{eff} as a function of the bed's temperature. The pebble bed is single size (pebbles' diameter is 1.7–2.0 mm) with a packing fraction of 61%. The measured density of the Li_2TiO_3 pebbles is 2.93 g/cm^3 with $83 \pm 4\%$ T.D. [8]. Stagnant helium at atmospheric pressure was used as a filling gas. After reaching the

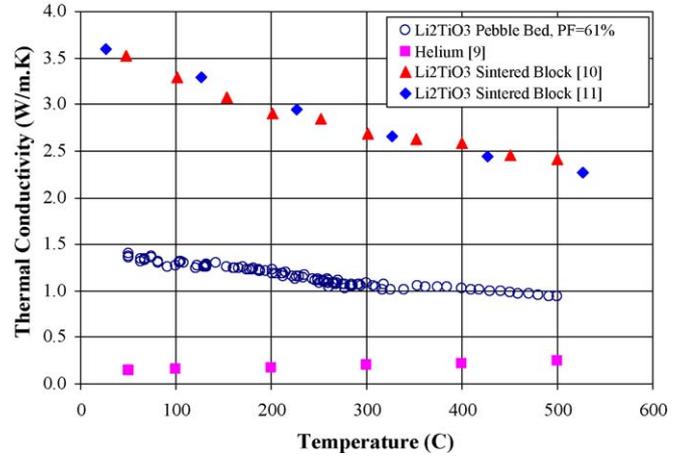


Fig. 4. Thermal conductivity of Li_2TiO_3 (pebble bed and sintered block) and helium.

steady state for each experimental run, the measured temperatures of the pebble bed were used to plot its thermal profile (temperature distribution). Fig. 3 shows several thermal profiles of the Li_2TiO_3 pebble bed obtained at different average bed temperatures, T_m (from 160 to 310 °C). The temperatures of each run can be fitted well by a straight line, indicating that the effective thermal conductivity across the pebble bed is practically constant.

Fig. 4 shows the thermal conductivity of both Li_2TiO_3 (sintered block) and helium in addition to the effective thermal conductivity of Li_2TiO_3 pebble bed obtained by the present study. It is observed that with the increase of temperature, thermal conductivity of Li_2TiO_3 (sintered block) decreases and thermal conductivity of helium increases. The thermal conductivity of Li_2TiO_3 (sintered block) decreases from 3.53 W/m K at 50 °C to 2.41 W/m K at 500 °C. With the same range of temperature, the thermal conductivity of helium increases from 0.15 to 0.24 W/m K. The experimental results, given in Fig. 4, of this study show that the effective thermal conductivity, k_{eff} , decreases from 1.40 to 0.94 W/m K with the increase of the average bed temperature from 50 to 500 °C. In other words, value of k_{eff} at 500 °C is 67% of its value at 50 °C. The decrease of thermal conductivity of Li_2TiO_3 (sintered block) with increase of temperature has a bigger impact on k_{eff} than that of increase of thermal conductivity of helium. Thermal conduction through the solid pebbles and thermal conduction through the contact areas are expected to dominate when the filling gas is stagnant and its thermal conductivity is small compared to that of the pebbles [12]. For pebble beds with stagnant gas, conduction and radiation are the dominant modes of heat transfer inside the pebble bed. The conduction contribution, which is usually the most significant, depends not only on the thermal conductivity of the pebbles and the filling gas, but also on the contact areas among the pebbles. Radiation becomes more dominant at high temperatures with large temperature differences across the pebble bed, particularly when the pebbles have large sizes [13]. Schotte [14] showed that radiation becomes significant at temperatures above 400 °C for 1 mm pebbles and above 1500 °C for 0.1 mm

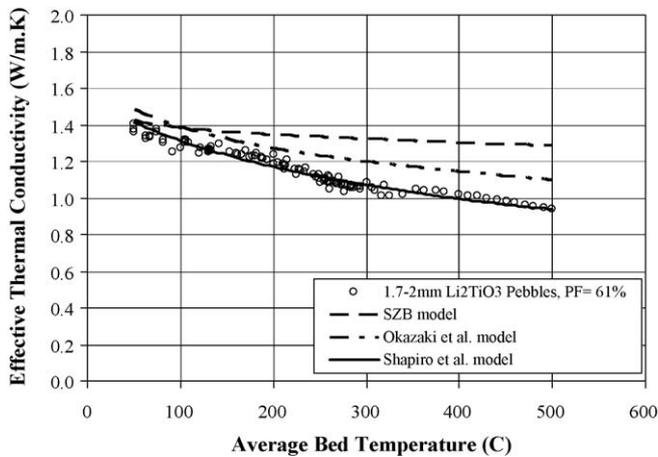


Fig. 5. Effective thermal conductivity of Li_2TiO_3 pebble bed (experimental results and models' predictions).

pebbles. Also, Hill and Wilhelm [15] studied the radiative and conductive heat transfer in a pebble bed of 3.81 mm-diameter alumina spheres. In their study, the ratio of heat transfer by radiation to that transferred by conduction was estimated to increase from 0.1 at 100 °C to 1.2 at 1000 °C (as average bed temperature).

Fig. 5 shows the measured effective conductivity of the Li_2TiO_3 pebble bed compared with the predictions of three models: Okazaki et al. model, Shapiro et al. model and SZB model. The present experimental values of k_{eff} agree reasonably well with those values predicted by Shapiro et al. model. However the predicted values of k_{eff} , by SZB and Okazaki et al. models, are higher than the measured values especially at temperatures from 300 to 500 °C. The SZB model over predicts the values of k_{eff} by 22, 27, and 37% at temperatures 300, 400, and 500 °C, respectively. Also, Okazaki et al. model over predicts the values of k_{eff} by 11, 12, and 17% at temperatures 300, 400, and 500 °C, respectively.

The effective thermal conductivity of Li_2TiO_3 pebble beds was not measured before (to our knowledge) in the temperature range covered in this work. However, it was experimentally measured in previous studies [16,17] for a temperature range of 425–775 °C. The measured values of k_{eff} in these studies are not included in the comparison, shown in Fig. 5, because their temperature range is beyond the temperature range of this study. Values of k_{eff} , in the temperature range of 50–500 °C, are required for the thermal analysis and modeling of the ITER test blanket modules, particularly for transient and off-normal events.

6. Conclusions and future work

The effective thermal conductivity of Li_2TiO_3 pebble beds is a key property for the design and analysis of fusion breeding blankets. The published data on k_{eff} of Li_2TiO_3 pebble beds is insufficient and more data is still needed. An experimental apparatus was designed and built, based on the principles of steady state and axial heat flow methods, in order to conduct

the required measurements in this study. The effective thermal conductivity, k_{eff} , of a Li_2TiO_3 pebble bed was measured as a function of the average bed temperature in the range of 50–500 °C. The pebble bed is single size (1.7–2 mm diameter pebbles) with a packing fraction of 61%. Helium at atmospheric pressure was used as a cover gas. The experimental results showed that k_{eff} decreased from 1.40 to 0.94 W/m K with the increase of temperature from 50 to 500 °C. The measured values of k_{eff} of the Li_2TiO_3 pebble bed were compared with the predictions of the three models of Okazaki et al., Shapiro et al. and SZB. The measured values of k_{eff} agree reasonably well with the predicted values by Shapiro et al. model. The presented values of k_{eff} will help to create a complete and reliable database of the thermal properties of the lithium ceramics pebble beds. This database is required for thermal analysis and modeling of ITER test blanket modules. In the near future, k_{eff} will be measured at a higher temperature range (from 500 to 800 °C) after making the required modifications in the current experimental apparatus.

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