EFFECTIVE THERMAL CONDUCTIVITY OF LITHIUM CERAMIC PEBBLE BEDS FOR FUSION BLANKETS: A REVIEW

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The use of lithium ceramic pebble beds has been considered in many blanket designs for the fusion reactors. Lithium ceramics have received a significant interest as tritium breeders for the fusion blankets during the last three decades. The thermal performance of the lithium ceramic pebble beds plays a key role for the fusion blankets. In order to study the heat transfer in the blanket, the effective thermal conductivity of the lithium ceramics pebble beds has to be well measured and characterized. The data of effective thermal conductivity of lithium ceramic pebble beds is important for the blanket design. Several studies have been dedicated to investigate the effective conductivity of the lithium ceramics pebble beds. The objective of this work is to review and compare the available data, presented by various studies, of effective conductivity of lithium ceramic pebble beds in order to address the current status of these data.

I. INTRODUCTION

The lithium ceramic blanket is a promising blanket concept for the fusion reactors, and worldwide efforts have been dedicated to its R&D. Lithium ceramics have received a significant interest as solid breeders for the fusion blankets during the last three decades. The solid breeder candidates are lithium oxide, lithium orthosilicate, lithium titanate and lithium metazirconate. The potential tritium breeding materials are required to have good thermal properties as well as satisfactory tritium breeding characteristics. The thermal conductivity is considered a critical property for the fusion solid breeder blankets. In order to study the heat transfer in the blanket, effective conductivity of the lithium ceramics pebble beds has to be well measured and known.

II. PARAMETERS AFFECTING EFFECTIVE CONDUCTIVITY OF PEBBLE BEDS

The effective thermal conductivity of pebble beds is influenced by many parameters with different degrees. Some of these parameters have significant impact on thermal conductivity of the pebble beds, such as thermal conductivity of solid pebbles and filling gas, gas pressure, bed deformation and bed packing fraction. Other parameters have less impact such as pebble size and surface roughness. In the following paragraphs, the effects of these parameters are presented in more details.

II.A. Conductivity of Pebbles

Thermal conductivity of solid pebbles has a direct impact on effective conductivity of the pebble beds. Effective conductivity of pebble bed is directly proportional to conductivity of the pebbles. A previous study[1] showed that effective conductivity of Li_2O pebble bed is larger than those of Li_2TiO_3, Li_2ZrO_3 and Li_4SiO_4 pebble beds because thermal conductivity of Li_2O is larger than those of the aforementioned lithium ceramics.

II.B. Conductivity of Gas

A previous study[2] revealed effect of gas conductivity on effective conductivity of pebble beds where vacuum, air, nitrogen and helium were used as a filling gas. The results showed that using helium as a filling gas with beryllium pebble bed gives the highest thermal conductivity while the smallest conductivity was obtained with vacuum at the same temperatures. The values of effective conductivity obtained with air and nitrogen were close and smaller than those obtained with helium by a factor of 2.7. Although the gas conductivity is much lower than that of the pebbles, the gas contribution had a significant effect on effective conductivity of pebble beds.

II.C. Ratio of Solid to Gas Conductivity

Thermal conductivities of pebbles and filling gas have direct impact on effective conductivity of the pebble beds. For this kind of pebbles/gas medium, the ratio of solid to gas conductivity (k_s/k_g) should be considered when studying effective conductivity of pebble bed. This ratio plays an important role in the heat transfer throughout the pebble bed. With low values (2 to 15) of k_s/k_g, the heat flux tends to be more uniform throughout the solid and gas regions of the bed. When this ratio is high (> 15), the heat flux prefers to follow the path of higher conductivity regions (pebbles and contact areas among pebbles). Therefore, the pebble beds with low value of k_s/k_g are less affected by changes at the contact areas and bed deformation.

II.D. Gas Pressure

According to the molecular theory of gases, thermal conductivity of a gas is generally independent of its pressure. This remains valid as long as the mean free path of the gas molecules is small compared with the dimensions of the local space. The mean free path is inversely proportional to the gas pressure, i.e., it increases with the decrease of pressure. When the pressure is low enough such that the mean free path (λ) is at the same order as the distance (L) between adjacent solid surfaces, the gas molecules can cross the gap and hit the solid surface without interacting with other gas molecules. When the gap between the pebbles becomes small enough, heat conduction through the filling gas is strongly affected by the Knudsen domain. For large Knudsen number ($Kn = \lambda/L$), thermal conductivity of the gas depends on both dimensions of the gaps and mean free path of the gas molecules. The pressure dependence of the mean free path will cause, in such cases, a pressure dependence of effective conductivity of the pebble beds. This effect is more noticeable with small pebble sizes and higher ratios of k_s/k_g. Some previous experiments confirmed the effect of low gas pressure on k_{eff}, i.e., when the gas thermal conductivity is in the Knudsen domain. Figure 1 shows effect of helium pressure on effective conductivity of Li_4SiO_4 (0.25-0.6mm) and Li_2ZrO_3 (1mm) pebble beds.[1] Also, previous experimental results[3] on gas pressure dependence of effective conductivity of Li_2ZrO_3 (1.2mm) pebble bed are shown in Fig. 1. The results show that effective conductivity of the pebble beds increases by a factor of ~1.9 with the increase of helium pressure from 4 to 100KPa. With a binary (0.1 & 4mm) Al/He pebble bed, effective conductivity increased by up to a factor of 2 with the increase of pressure from 0.1 to 4atm.[4]

II.E. Bed Packing Fraction

Bed packing is a main characteristic of any pebble bed and it has a significant impact on the bed thermal behavior. Bed packing is affected by many factors such as packing technique, pebble arrangement (single or binary size), pebble size relative to bed size, and range of pebble size. The packing fraction is independent of the pebble size, provided that the pebbles are small compared to the bed dimensions. It has been proved that the minimum container dimension should exceed 10 times the pebble diameter to ensure a reliable packing fraction of around 62%. In general, the packing fraction ranges from 60 to 64% for single size beds while it ranges from 80 to 84% for binary size beds. In a previous study[5] the results showed that effective conductivity of beryllium/helium pebble bed is directly proportional to the packing fraction.

II.F. Pebble Size

The pebble size has direct impact on two bed characteristics, namely; near-wall region and distance between adjacent pebbles. The latter is an important parameter for determining the Knudsen number and influencing the local conductivity of the gas if operating in the Knudsen regime. The ratio of pebble size to bed dimension determines the characteristics of the near-wall region where the packing and thus bed thermal behavior acts differently compared to bulk region of the bed.

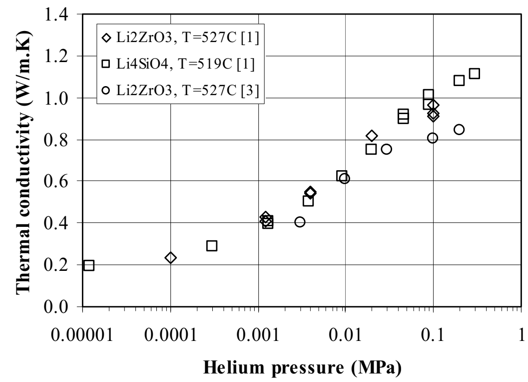

Fig. 1. Effective conductivity of lithium ceramics pebble beds versus helium pressure

II.G. Pebble Surface Roughness

The surface roughness of a pebble affects its contact behavior with the neighboring pebbles and consequently the contact areas among pebbles. This would affect both the contact thermal resistance, which affects k_{eff}, and the pebble-to-pebble interaction, which affects the bed stress/strain mechanics. In addition, the volume of gas trapped among the pebbles is influenced by the pebbles' surface roughness.

II.H. Contact Area and Bed Deformation

The contact area between pebbles directly affects the amount of heat flux across it especially for pebble beds with high k_s/k_g ratio. When the pebbles are subjected to stress, large enough to cause deformation, the contact area increases and consequently more heat is expected to flow through it. The effect of bed deformation on effective conductivity of Li_2ZrO_3 pebble bed was studied.[6] No effect was found within 10% error for loads up to 1.3MPa. Also, Li_2TiO_3 and Li_4SiO_4 pebble beds were investigated[7] at temperatures up to 800°C and bed strain up to 4%. The results showed that the increase of thermal conductivity with increasing bed strain is small and became negligible at high temperatures. This small change in thermal conductivity was also reported[8] with Li_2ZrO_3 pebble bed and loads up to 1.7MPa. It was concluded that the effect of deformation on thermal conductivity can be neglected for lithium ceramic pebble beds due to the small k_s/k_g ratio.

III. THERMAL CONDUCTIVITY OF SINTERED PRODUCT LITHIUM CERAMICS

III.A. Thermal Conductivity of Li_2TiO_3

Thermal conductivity of a 95% Li_2TiO_3 sample was calculated from the values of thermal diffusivity and heat capacity, measured by laser-flash method.[9] The resulting values of thermal conductivity are shown in Fig. 2. A reference fit for thermal conductivity, k, of Li_2TiO_3, as a function of temperature, T, and porosity, p, is given by:[10]

$$k = (1-p)^{2.9}\,(5.35-4.78\times10^{-3}\,T+2.87\times10^{-6}\,T^2) \qquad (1)$$

Where $0.14 \le p \le 0.25$, $400 \le T \le 1400K$ and k in $W/m.K$
Also, thermal conductivity of Li_2TiO_3 pellets was reported by the following empirical equation:[11]

$$k = [(1-p)/(1+\beta p)]\times(4.77-5.11\times10^{-3}T+3.12\times10^{-6}T^2) \qquad (2)$$

$$\beta = 1.06 - 2.88\times10^{-4}\,T, \qquad (3)$$

p = 0.07-0.27, ρ = 73-93% TD, T = 300-1050K and k in $W/m.K$. Where p is porosity, ρ is the density and β is an empirical parameter. In addition, the thermal conductivity of Li_2TiO_3 was measured, in another study[12], as a function of temperature. Figure 2 shows the thermal conductivity of Li_2TiO_3, reported by different studies[9-12], plotted versus temperature.

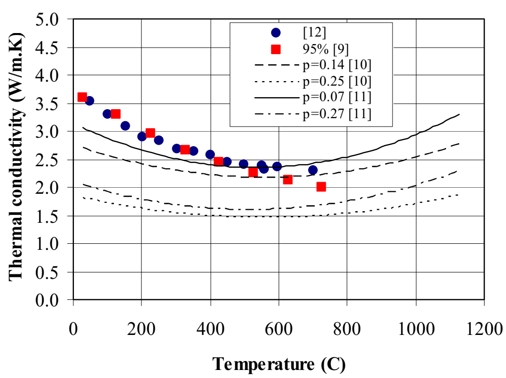

Fig. 2. Thermal conductivity of Li_2TiO_3

III.B. Thermal Conductivity of Li_4SiO_4

Thermal conductivity of Li_4SiO_4 was given as:[13]

$$k = (1.98 + 850/T)\,(1-p_o)/[1+p_o\,(1.95-8\times10^{-4}\times T)] \qquad (4)$$

Where T is temperature in K, p_o = 6.7% is porosity of the pebbles and k in $W/m.K$. In another study,[14] thermal conductivity of Li_4SiO_4 was reported as:

$$k = (1-p)^{5/3}(2.49)[(1+2.06\times10^{-3}\,T)^{-1}+1.85\times10^{-10}\,T^3] \qquad (5)$$

Where p is porosity ($0.16 \le p \le 0.3$), T is temperature ($373 \le T \le 873K$) and k in $W/m.K$. Also, thermal conductivity of Li_4SiO_4 was presented by another study.[12] Figure 3 shows thermal conductivity of Li_4SiO_4, given by different references[12-14], plotted versus the temperature.

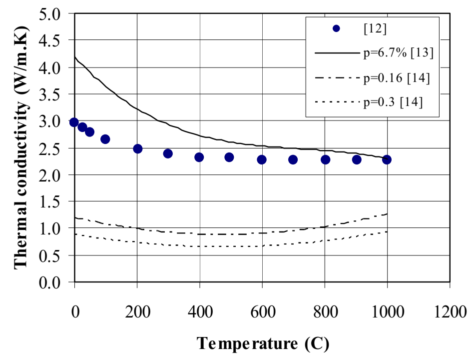

Fig. 3. Thermal conductivity of Li_4SiO_4

III.C. Thermal Conductivity of Li_2O

Figure 4 shows thermal conductivity of Li_2O, measured by previous studies.[15,16] Thermal conductivity, k, of Li_2O was reported by the following equation:[15]

$$k\,(W/m.K) = (1-p)^{1.94}\times[0.022+1.784\times10^{-4}\,T\,]^{-1} \qquad (6)$$

Where p (70.8-93.4%) is the volume fraction porosity and T is temperature in K. Also, thermal conductivity of Li_2O was measured[16] in a temperature range of $473 \le T \le 1173K$ and a porosity range of $0.066 \le p \le 0.292$. A good correlation of k is given by:[17]

$$k\,(\mathrm{W/m.K}) = (1-p)^{1.96}\,[39.79/(1+7.067\times10^{-3}T)] \qquad (7)$$

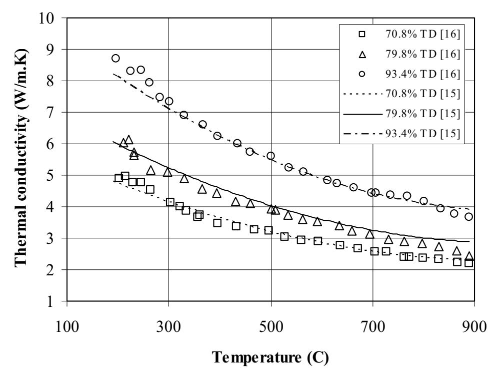

Fig. 4. Thermal conductivity of Li_2O

III.D. Thermal Conductivity of Li_2ZrO_3

Gierszewski[18] summarized thermal conductivity data for Li_2ZrO_3 in a temperature range of $373 \le T \le 1063K$ and a porosity range of $0.187 \le p \le 0.211$. The data correlate well with the following equation:

$$k = (1-p)^{5/3}[(3.643/(1+0.00155T))+7.579\times10^{-10}T^3] \qquad (8)$$

Also, thermal conductivity of Li_2ZrO_3 was measured in other works.[19,20] Figure 5 shows thermal conductivity of Li_2ZrO_3, as given by different studies.[18-20]

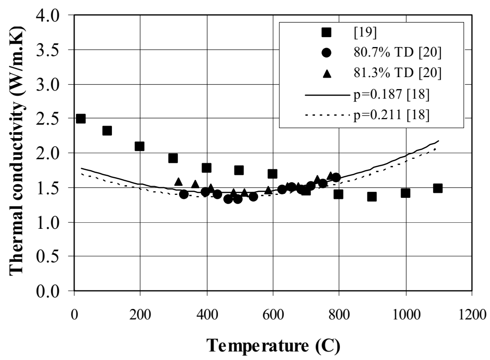

Fig. 5. Thermal conductivity of Li_2ZrO_3

IV. THERMAL CONDUCTIVITY OF HELIUM

Helium (He) is used as a purge gas with the solid breeder pebble beds. Thermal conductivity of He has a significant impact on effective conductivity of these pebble beds. Thermal conductivity of He, as a function of temperature (K), is given by:[21]

$$k\ (W/m.K) = 3.366 \times 10^{-3}\ T^{0.668} \quad (9)$$

Also, k was measured and correlated as follows:[22]

$$k = 0.55+3.353\times10^{-3}T-2.117\times10^{-7}T^2-6.626\times10^{-11}T^3 \quad (10)$$

Where k is in $mW/cm.K$ and T is in Kelvin. Figure 6 shows thermal conductivity of He as given by equations (9 & 10) and other works.[23,24] The reported results[24] show that thermal conductivity of He does not change with pressure in the range of 0.01 to 0.24MPa over a temperature range of 77-1227°C. Also the results presented in another study[25] show that thermal conductivity of He has neglected change with pressure in the range of 2.9 to 10.22MPa at 35, 65, 106.5 and 155°C.

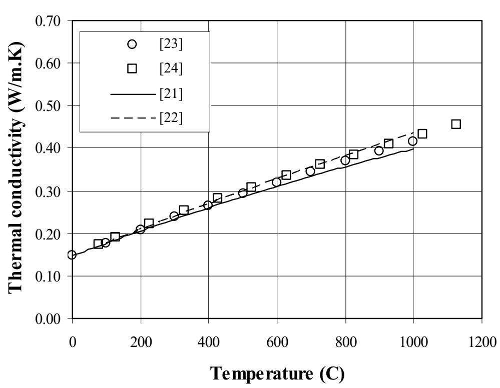

Fig. 6. Thermal conductivity of helium

V. EFFECTIVE CONDUCTIVITY OF LITHIUM CERAMIC PEBBLE BEDS

Effective thermal conductivity of solid breeder pebble beds is very important For the R&D of solid breeder blankets. Having a complete and reliable database for this property is required to achieve the needed progress in the R&D of fusion blanket. In the following sections, the previous studies on effective conductivity of lithium ceramic pebble beds are summarized.

V.A. Hatano et al.[26]

In 2003, Hatano et al.[26] presented their study on effective thermal conductivity of Li_2TiO_3 pebble bed. The hot wire method was used to measure effective thermal conductivity of single and binary size beds of Li_2TiO_3 pebbles. The effective conductivity was measured as a function of temperature (from 420 to 775°C). The packing fraction was ~ 60% using 1.91mm diameter pebbles for the single size pebble bed, while it was ~ 80% when using two sizes (0.28 and 1.91mm diameter pebbles) for the binary size pebble bed. Helium was used as a cover gas and its pressure was adjusted from vacuum to 0.2MPa. The results of this work are presented in Fig. 7.

V.B. Enoeda et al.[1]

In 2001, Enoeda et al.[1] measured effective conductivity of Li_2TiO_3, Li_4SiO_4, Li_2ZrO_3, and Li_2O pebble beds using the hot wire method. The measurement conditions for each breeder are given in TABLE I. The temperature ranged from 425 to 775°C for all runs. The results of this work are shown in Fig. 7, 8, 9, and 10 for Li_2TiO_3, Li_4SiO_4, Li_2ZrO_3, and Li_2O respectively.

TABLE I. Measurements conditions[1]

Breeder / Diameter (mm)	He Pressure (MPa)	Density (g/cm^3)	Packing Density (g/cm^3)
Li_2O / 0.85-1.18	0.1	1.61	0.999 (62.1% PF)
Li_2TiO_3 / 0.8-1.2	0.1	3.1	1.83 (59% PF)
Li_2ZrO_3 / 0.8-1.2	0.0001-0.1	3.6	1.921 (53.4% PF)
Li_4SiO_4 / 0.25-0.63	0.0001-0.2	2.32	1.407 (62.5% PF)

V.C. Dalle Donne et al.[27]

In 2000, Dalle Donne et al.[27] measured effective conductivity of a Li_4SiO_4 pebble bed as a function of the bed average temperature, see Fig. 8. The experiments were performed using pebbles of 0.25-0.63mm diameter and packing factor of ~65%. Helium was used as a cover gas at different pressures (1 to 3bar). The results showed that for pressures higher than 1bar, the pressure variation has no effect on the bed effective conductivity. Effective conductivity of the bed was correlated by:

$$k\ (W/m.K) = 0.768 + 0.496\times10^{-3}\times T_m\,(°C) \quad (11)$$

V.D. Earnshaw et al.[3]

In 1998, Earnshaw et al.[3] measured effective conductivity of Li_2ZrO_3 pebbles bed in helium over a temperature range of 75-1170°C. The pebbles were quite spherical (1.2mm diameter) and were typically 80 to 82%

dense, as compared with solid lithium zirconate. The packing fraction ranged from 63 to 65%. The resulting average density of the bed, considering both the porosity and the packing, was 2.23±0.07g/cm^3. The data, shown in Fig. 9, can be statistically fitted to the following relation:

$$k = 0.69 + 2.2\times10^{-10}\times T^3(K) \quad (12)$$

V.E. Lorenzetto et al.[28]

In 1995, Lorenzetto et al.[28] presented their results of effective conductivity of Li_2ZrO_3 pebble bed, over a temperature range of 100-1175°C, see Fig. 9. For 1.2mm Li_2ZrO_3 pebbles (82% dense, PF=63%) in 0.1MPa helium, effective conductivity data is fitted by:

$$k\ (W/m.K) = 0.66 + 1.17 \times 10^{-7}\ T^{2.2}\ (°C) \quad (13)$$

V.F. Dalle Donne et al.[29]

In 1994, Dalle Donne et al.[29] measured effective conductivity of Li_4SiO_4 pebbles (0.35-0.6mm) bed versus the bed temperature, see Fig. 8. Helium was used as a cover gas and the packing fraction was 64.4%. The density of the pebbles is 97% of the theoretical value, i.e. 2.32g/cm^3. The data was correlated in the temperature range of 40-720°C by the following equation:

$$k\ \ (W/m.K) = 0.708 + 4.51\times10^{-4}T + 5.66\times10^{-7}\times T^2 \quad (14)$$

V.G. Enoeda et al.[5]

In 1994, Enoeda et al.[5] presented their measurements of effective conductivity of Li_2O pebbles bed. The measurements were performed with ø1mm Li_2O spheres within a temperature range of 150-650°C. Helium, at 1atm, was used in the packed beds. The initial packing fraction of the pebble bed was 48%. Effective thermal conductivity in this study has the tendency to decrease with the temperature increase, see Fig. 10.

V.H. Sullivan et al.[30]

In 1991, Sullivan et al.[30] measured effective thermal conductivity of Li_2ZrO_3 pebble bed in 0.1MPa helium gas over a temperature range of 70-500°C. The Li_2ZrO_3 pebbles are ø1.2mm, 80% dense and the packing fraction is 60%. Figure 9 shows the thermal conductivity for the Li_2ZrO_3 spheres bed presented by this study.[30]

V.I. Dalle Donne and Sordon[13]

In 1990, Dalle Donne and Sordon[13] measured effective conductivity of Li_4SiO_4 pebbles (ø0.5mm) bed. The bed mean temperature ranged from 50 to 350°C and stagnant helium, at 1bar, was used. The experimental values of effective conductivity of Li_4SiO_4/He pebble bed are shown in Fig. 8.

VI. ANALYSIS AND DISCUSSION

VI.A. Results of Li_2TiO_3 Pebble Beds

Figure 7 shows the experimental results of effective conductivity, k_{eff}, of Li_2TiO_3 pebble beds.[26,1] The results show that k_{eff} of the Li_2TiO_3 single size bed[26] increases from 1 to 1.22W/m.K over a temperature range of 420-775°C. While it increases from 1.26 to 1.34W/m.K with the binary size bed[26] for the same temperature range. Increasing packing fraction (from 60 to 80%) of the Li_2TiO_3 pebble beds[26] leads to an increase in k_{eff} by 25% at 420°C and 14% at 775°C. This is because in the binary size bed, the contact areas among pebbles are larger than those in the single size bed. These results indicate the effect of packing fraction on the values of k_{eff}. Also, Fig. 7 shows that k_{eff} of Li_2TiO_3 pebble bed[1] increases from 1.02 to 1.15W/m.K over a temperature range of 425-775°C. All the data, in Fig. 7, have the same trend of increasing k_{eff} with the temperature increase. It is observed that k_{eff} of Li_2TiO_3 pebble bed oscillates (decrease and then increase) with increase of temperature from 620 to 775°C.

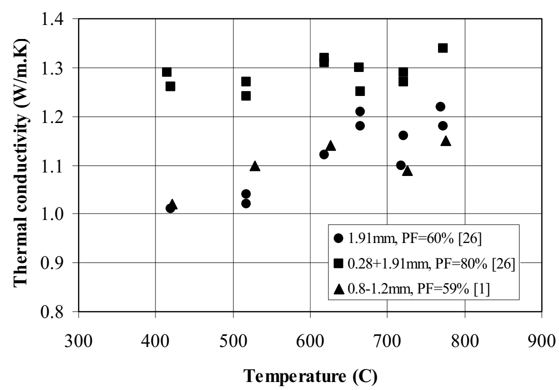

Fig. 7. Effective conductivity of Li_2TiO_3 pebble beds

VI.B. Results of Li_4SiO_4 Pebble Beds

Figure 8 shows effective conductivity, k_{eff}, of Li_4SiO_4 pebble beds presented by previous studies.[1,13,27,29] The results show a negligible temperature dependency of k_{eff} of Li_4SiO_4 pebble bed[1], where k_{eff} ranges from 0.97 to 1.03W/m.K over a temperature range of 425-775°C. However, the results show that k_{eff} of Li_4SiO_4 pebble bed[29] increases from 0.75W/m.K at 75°C to 1.33W/m.K at 725°C. Also, k_{eff} of Li_4SiO_4 pebble bed[27] increases from 0.81 to 1.13W/m.K over a temperature range of 75-725°C. While k_{eff} of Li_4SiO_4 pebble bed[13] increases from 0.74 to 0.9W/m.K with the increase of temperature from 51.6 to 333°C. Results of k_{eff}[27] have the highest values because they have the highest packing fraction (65%), this is true for temperature up to 370°C, however above this temperature values of k_{eff}[29] are the highest. Values of k_{eff}, given by equations (11 & 14) show good agreement with values of k_{eff}[13,1] for temperature up to 570°C however, above 570°C there is a discrepancy in k_{eff} values.

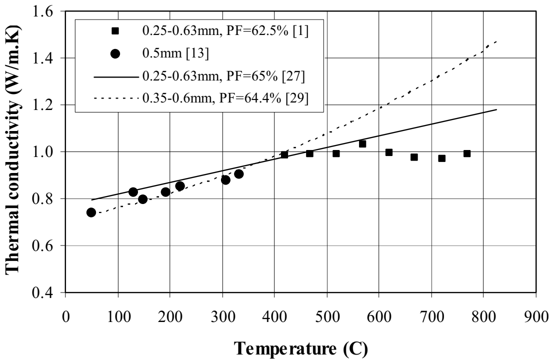

Fig. 8. Effective conductivity of Li_4SiO_4 pebble beds

VI.C. Results of Li_2ZrO_3 Pebble Beds

Figure 9 shows the experimental results reported by four previous studies[1,28,30,3] on effective conductivity of Li_2ZrO_3 pebble beds as a function of bed temperature. All the data shown in Fig. 9 have similar behavior as k_{eff} values increase with the increase of temperature. The results show a small temperature dependence of effective conductivity of Li_2ZrO_3 pebble bed[1] where k_{eff} ranges from 0.9 to 1W/m.K over a temperature range of 425-775°C. However, effective conductivity of Li_2ZrO_3 pebble bed[28] increases from 0.74 to 1.35W/m.K with the increase of bed temperature from 320 to 1170°C. While effective conductivity of Li_2ZrO_3 pebble bed[30] increases from 0.63 to 0.85W/m.K over a temperature range of 70-500°C. Also, the results show that effective conductivity of Li_2ZrO_3 pebble bed[3] increases from 0.64W/m.K at 75°C to 1.33W/m.K at 1167°C. The reported data[3,28] show an increase of k_{eff} by 49% with the increase of temperature from 450 to 950°C. The values of k_{eff}[3,28] show good agreement in the temperature range of 450-1170°C. This is because they have similar pebble size (1.2mm) and packing fraction (63%). For the temperature range of 425-775°C, k_{eff}[1] has the highest values despite the packing fraction of this pebble bed is the lowest one (53.4%).

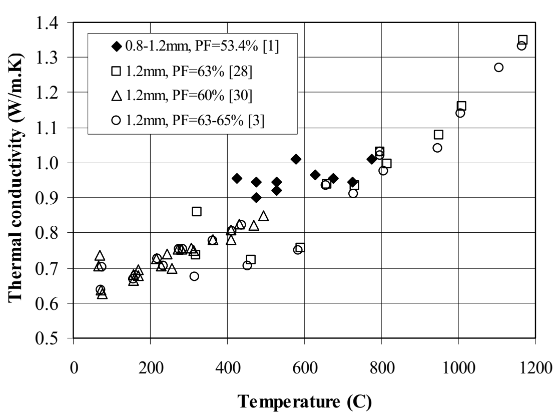

Fig. 9. Effective conductivity of Li_2ZrO_3 pebble beds

VI.D. Results of Li_2O Pebble Beds

Figure 10 shows the experimental results of previous studies[1,5] on effective conductivity of Li_2O pebble beds. The results show a small temperature dependence of effective conductivity of Li_2O pebble bed[1] where k_{eff} oscillates between 1.59 and 1.77W/m.K over a temperature range of 425-770°C. Effective conductivity of Li_2O pebble bed[5] has an average value of 0.9W/m.K and shows a slight tendency of decrease with the increase of temperature. The values of k_{eff}[1] are higher (by ~95%) than those k_{eff}[5] at the same temperature values. This is a result of the difference in the packing fraction (62.1%[1] & 48%[5]) of these pebble beds. To conclude, all the data show poor temperature dependence of k_{eff}, and some values of k_{eff} decrease (dissimilar to other lithium ceramics behavior) with the increase of temperature.

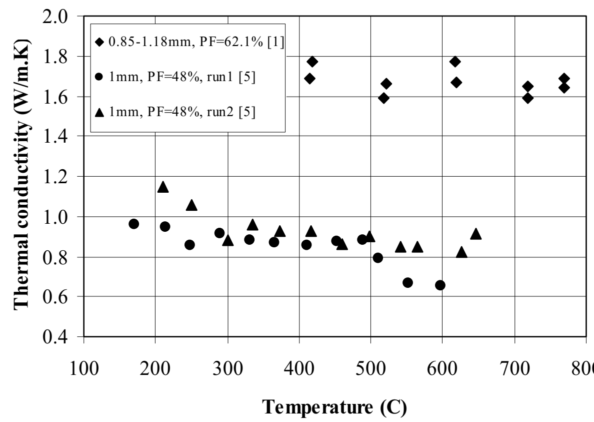

Fig. 10. Effective conductivity of Li_2O pebble beds

VII. CONCLUDING REMARKS

A reasonable number of experimental studies on effective conductivity, k_{eff}, of lithium ceramic pebble beds have been done in the last two decades. In the course of this work, the available experimental studies have been reviewed and compared. The published studies have been successful in identifying the key parameters which affect effective conductivity of the pebble beds. It was observed that all the available studies did not report the uncertainty analysis (experimental errors) of their experimental results on k_{eff} of lithium ceramics pebble beds. The experimental results of Li_2O pebble beds showed a poor temperature dependence of k_{eff}, and some values of k_{eff} decrease (dissimilar to other lithium ceramics behavior) with the increase of temperature. Only two studies,[1,26] presented data on k_{eff} of Li_2TiO_3 pebble beds, and two studies[1,5] presented data on k_{eff} of Li_2O pebble beds. All the data on k_{eff} of Li_2TiO_3 oscillate (decrease and then increase) with the increase of temperature from 620 to 775°C. For Li_4SiO_4 pebble beds, there is a discrepancy among k_{eff} values with the temperature range of 570-825°C. More experimental data, with acceptable level of confidence and accuracy, are still required especially for Li_2TiO_3 and Li_2O pebble beds.

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