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Thermal conductivity of sintered lithium orthosilicate compacts

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ABSTRACT

The design of solid breeder blankets is strongly affected by the low values of thermal conductivity and density of ceramic breeder pebble beds. A significant rise of both quantities would enhance the thermal performance and lead to an increased tritium breeding ratio. In order to improve these quantities pretreated lithium orthosilicate pebble material was dry pressed and subsequently sintered. The thermal conductivity of cylindrical pellets was determined by the heat pulse method using a laser flash device. A pebble bed characteristic sample was also investigated in order to check the measurement accuracy in comparison with previous results. Furthermore, two samples of low density cellular ceramics were also prepared by infiltration of polymer foams with a ceramic slurry. The thermal conductivity results show that the values are affected both by the particle size and the sample density. Thermal conductivity values of higher than 2 W/m K were obtained using large particles and sintering at 1000 °C.

1. Introduction

For the helium cooled pebble bed (HCPB) blankets, studied within the European Fusion Technology Program, lithium orthosilicate (OSi) and lithium metatitanate, both in form of pebbles, are the candidate breeder materials [1]. The design of this type of blanket is strongly affected by the low values of the ceramic breeder pebble bed thermal conductivity (≈1 W/m K in helium atmosphere) and theoretical density (≈64%). A significant increase of both quantities would significantly improve the thermal performance and would also lead to an increased tritium breeding ratio. Based on these considerations, Sharafat et al. [2] proposed the application of ceramic foams or cellular ceramics as a solid breeder material. It is obvious that in contrast to ordinary foams, which are characterized by a low density, the advanced material must have a large density in order to meet the requirement for a fusion blanket. Examples of such high density materials or experimental results, however, were not given in [2].

From previous research results [3] it was obvious that it is not sufficient to increase pebble contacts by elastic or plastic deformation. A better thermal and mechanical contact can be achieved by forming sintering necks or partial melting.

In the present study we used lithium orthosilicate pebbles developed in collaboration between Research Center Karlsruhe and Schott AG, Mainz, as the starting material. These pebbles are fabricated by a melt-spraying technique in a semi-industrial scale facility. Li_4SiO_4 pebbles with a surplus of 2.5 wt% SiO_2 are produced by melting a mixture of lithium hydroxide and silica powders and then spraying the liquid material in air [4]. Pebbles with diameters between 400 and 600 μm were used as starting material to study the influence of density and microstructure on the thermal conductivity. Low density foams were also included in our study.

2. Experimental

2.1. Sample preparation

Lithium orthosilicate pebbles with diameters in the range of 400–600 μm were used as starting material. These pebbles were either used as-received or crushed to mean particle sizes of ≈250 μm or <50 μm. The samples were dry pressed at different pressures without any binder into cylindrical pellets of 1–2 mm thickness and 12 mm in diameter and subsequently sintered at temperatures of $T_s = 970$ and 1000 °C for 2 h or 6 h, respectively.

In addition, cylindrical pellets of cellular ceramics were prepared by infiltration of polymer foams with an alcoholic slurry (57 wt% Li_4SiO_4-powder). After the infiltration the ceramic foam pellets were dried at room temperature for 3 h and afterwards thermally treated at 970 °C for 2 h.

2.2. Characterization

The microstructure and the pore geometries of the samples were analyzed using scanning electron microscopy (SEM) (JSM 6400, Jeol, Japan). The thermal diffusivity of the ceramic pellets was determined by the heat pulse method using a laser flash device LFA 427 (Netzsch, Germany) at temperatures between 25

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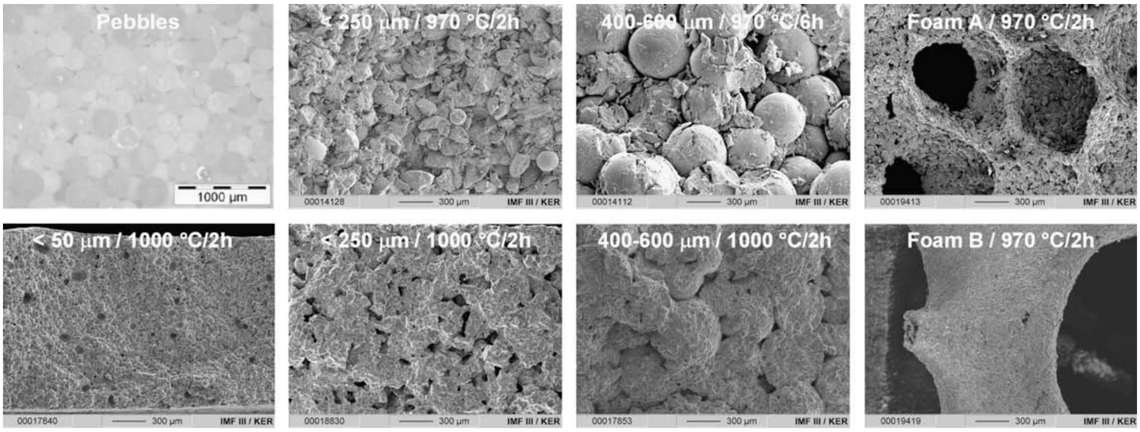

Fig. 1. Optical microscopy of pebbles and microstructure at fracture surfaces of sintered pellets and foams after different heat treatments.

and 750 °C in helium atmosphere. The ceramic foams were measured in argon. The thermal conductivity λ was evaluated from the thermal diffusivity α, the measured OSi specific heat capacity c_p and the OSi density ρ as a function of temperature according to $\lambda(T) = \alpha(T)\ \rho(T)\ c_p(T)$. The densities of the samples were determined geometrically.

In order to check the accuracy of the laser flash technique with previous data [3,5], a pebble bed consisting of OSi pebbles with diameters between 250 and 630 µm was also measured.

Table 1
Characteristic parameters of the investigated samples.

	Sample	T_S (°C)/Dwell time (h)/press capacity (kN)	Sinter density (% TD)	Thermal conductivity at 600 °C (W/m K)
1	<50 µm	1000/2/15	82	1.6
2	<50 µm	1000/2/20	84	1.7
3	<50 µm	1000/2/25	87	1.9
4	<250 µm	970/2/15	70	1.3
5	<250 µm	1000/2/18	79	1.5
6	>250 µm	970/2/20	72	1.7
7	>250 µm	1000/2/20	75	1.9
8	400–600 µm	970/2/25	72	1.6
9	400–600 µm	970/6/25	75	1.9
10	400–600 µm	1000/2/30	81	2.1
11	400–600 µm	1000/2/50	89	2.2
12	Foam A	970/2/–	23	0.5
13	Foam B	970/2/–	24	0.7
14	Pebble bed	–	60	1.0

3. Results

3.1. Sample structures

The microstructure of the measured samples was examined by scanning electron microscopy. In Fig. 1, top left, the pebbles are shown in its original state. The other figures show the microstructure at fracture surfaces of sintered samples and two foams. Characteristic parameters of the samples are represented in Table 1.

At T_s = 970 °C the pressed samples are compacted by solid state sintering. Both foams exhibit different pore structures with fully open, partially interconnected, and closed porosity. At T_s = 1000 °C the material is partially melted. The microstructure of the <50 µm sample exhibits small particles with basically closed porosities, whereas fragments of pebbles, sinter necks, closed and open porosity can be observed for <250 and 400–600 µm particle sizes.

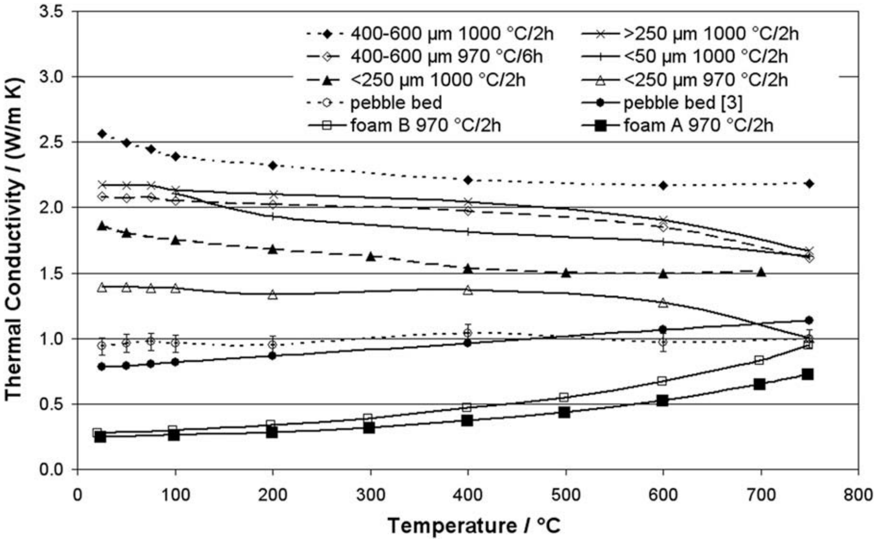

Fig. 2. Thermal conductivity of foams, pebble beds, and different pellets.

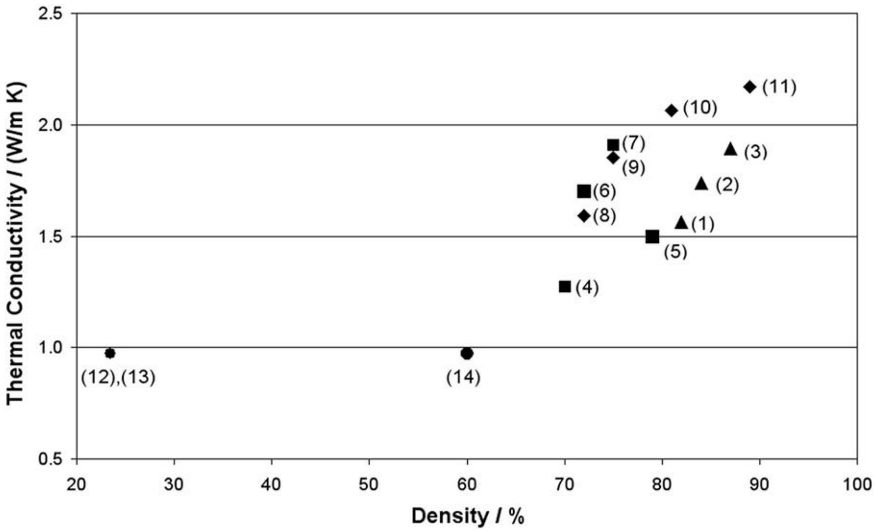

Fig. 3. Thermal conductivity at 600 °C versus sample densities, label reference see Table 1.

3.2. Thermal conductivity measurements

At first, the measurements were performed with samples with $T_s = 970$ °C. Fig. 2 shows that the pellet with particle sizes of 400–600 μm (75% TD) possesses the highest values which vary between 2.1 W/m K at 25 °C and 1.9 W/m K at 600 °C. The pellet with particle sizes <250 μm (70% TD) has lower values with 1.4 W/m K at 25 °C to 1.3 W/m K at 600 °C. The lowest values belong to foams with densities of 23% and 24% TD (600 °C: 0.5–0.7 W/m K). However, it should be noted that the foams were measured in an argon atmosphere. For the low density foams, the contribution of the gas to the effective thermal transport may be larger compared to the other samples, which also leads to a different temperature dependence of the thermal conductivity, i.e. $\lambda(T)$ increases with rising temperature. In order to estimate the foam conductivities in helium gas, the Schluender–Bauer–Zehner model (SBZ model) [6] was used. Compared to argon, the values are larger by about a factor of two. Results for the samples with $T_s = 1000$ °C are also shown in Fig. 2. The values are generally higher than those for $T_s = 970$ °C. Again, the highest values are obtained for the pellet with 400–600 μm particles with a density of 89%. The <250 μm samples (79% and 70% TD) exhibit the lowest values, whereas, the thermal conductivity values for the pellet with particle sizes <50 μm (84% TD) are intermediate. Fig. 2 also contains data for non-compressed OSi pebble bed from [3,5] and the corresponding results obtained by the laser method using the sample containing the non-compressed OSi pebbles. As the data differ not more than about 15%, the laser flash method can also be used as a screening method for non-homogeneous materials. The typical uncertainty for the thermal conductivity measurement of the pressed and sintered samples as well as for the foams is about ±5%.

The important result of the present study is, that compared to pebble beds, the thermal conductivity of the high density samples can be increased by a factor of larger than two, which is then only about 10% lower than the thermal conductivity of the solid material.

Peeters et al. [7] observed that the tritium release improves with increasing open porosity and that closed porosity does not contribute to tritium release. In pellets with a particle size <50 μm, the tritium release appears to be very difficult, whereas structures with large open pores are much better suited.

Fig. 3 shows the thermal conductivity values for all samples at 600 °C in helium atmosphere as a function of the density. Blanket pebble beds are characterized by densities of about 63–65% and a thermal conductivity of about 1 W/m K. This value changes only marginally if the pebbles are compressed under blanket operation [3]. For the sintered samples, the thermal conductivity increases with increasing density. The conductivity of the samples made of smaller particles is lower comparing samples with the same density. Such a tendency is also predicted by the SBZ model for pebble beds.

The data of the foams (assessed for helium atmosphere) are also given in Fig. 3. The thermal conductivity is similar to that of pebble beds although the density is much smaller. The reason for this is that foams have much larger conductivity bridges compared to pebble beds.

4. Conclusions

The results show that the thermal conductivity is affected by the density as well as by the microstructure of the ceramic samples. Therefore, variations in the manufacturing procedure have a clear impact on the thermal conductivity of lithium orthosilicate sintered plates.

Compared to OSi pebble beds, the sintered bodies consisting of relatively large particles have thermal conductivities up to two times higher. The OSi density also increases remarkably by up to 30%. These two improvements have a large impact on the blanket design: A larger thickness of breeder zones can be realized which reduces the inventory of the structural material. This leads to an increased breeding ratio which enables blanket designs with a smaller radial thickness or a reduced beryllium inventory. The open porosity in sintered plates consisting of large particles should be much more favorable for tritium removal.

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