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Ceramic pebble bed development for fusion blankets

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Abstract

Research on lithium ceramic breeders has been intensive since the late 1970s. The bulk material properties of several candidate lithium ceramics are essentially available, although there is still much work to be done on properties under irradiation and on the overall behavior in blanket modules. Based on these results, lithium ceramic breeder blankets have been selected in many fusion reactor design studies. These lithium ceramics are incorporated into blankets typically as monolithic pellets or packed pebble beds. There is substantial industrial experience with pebble beds made from other ceramics as catalyst supports, and in fabrication and testing of pebbles for advanced fission reactor fuels. In fusion blankets, the pebble bed form offers several attractive features, including simpler assembly into complex geometries, a uniform pore network and low sensitivity to cracking or irradiation damage. Ceramic breeder pebbles have been a focus for several research groups. In general, the database is similar to that of monolithic pellets for the materials studied; basic production and material property data are available, but the irradiation and engineering database remains sparse. In addition to the basic requirements on any ceramic breeder material (such as low tritium hold-up, compatibility with structure, irradiation stability, etc.), the main pebble bed requirements may be roughly summarized as follows: economic, high yield production rates; high average bed (smear) density; adequate bed thermal conductivity; acceptable purge gas pressure drop; adequate crush strength; tolerance to thermal cycling. In this paper, the international ceramic breeder pebble bed database is reviewed with respect to these pebble bed properties, and the R&D needs for reactor blanket development are assessed.

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1. Introduction

Lithium ceramic breeder blankets have been selected in many fusion studies [1–5]. These ceramics are incorporated into fusion blankets as pellets, or as packed beds of spheres or pebbles. Ceramic sphere beds have been widely used in chemical engineering as catalyst supports and grinding media, and have been developed for use in advanced fission reactor fuels. In fusion blankets, the packed bed form offers several advantages [6], including simpler assembly into complex geometries, a uniform porosity and low temperature sensitivity to cracking or irradiation damage, but there is little industrial experience with the relevant ceramics.

The material properties of several candidate lithium ceramic materials are essentially available, although there is still much work to be done on properties under irradiation and the ceramic performance in complex

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blankets. The packed bed form offers several advantages [6], including simpler assembly into complex geometries, a uniform porosity and low temperature sensitivity to cracking or irradiation damage, but there is little industrial experience with the relevant ceramics.

The material properties of several candidate lithium ceramic materials are essentially available, although there is still much work to be done on properties under irradiation and the ceramic performance in complex blanket modules [7]. In addition to the basic requirements on any ceramic breeder material (such as low tritium hold-up, compatibility with structure, irradiation stability, etc.), the requirements for pebble bed blankets may be summarized as follows [6]:

size range of about 0.1–1 mm diameter;
smear density over 50%
adequate crush strength;
tolerance to thermal cycling;
adequate bulk and interface bed thermal conductivity;
economic, high-yield production rates over 0.1 m^3 d^{-1};
purge gas pressure drop of less than 100 kPa.

In this paper, the international ceramic breeder pebble bed database is reviewed with respect to the key pebble bed properties, and the R&D needs for reactor blanket development are assessed. The specific properties discussed are as follows: fabrication, packing, thermal conductivity, purge gas permeability, thermal–mechanical strength, and tritium release.

2. Pebble production and physical characteristics

Blanket design considerations define the general production characteristics for reactor-relevant fabrication processes [6]. As a result of the large number of pebbles required to fill a reactor with a breeder volume of 30–100 m^3, a useful industrial production rate of about 0.1 m^3 d^{-1} is suggested. As a result of the expensive ^6Li content, the process must have a high yield of useful pebbles. The pressure drop, heat transfer and packing considerations mean that the acceptable pebble size is roughly 0.1–1 mm in diameter, although the size distribution may be narrow, wide or binary within this range. It is not necessary for the pebbles to be uniform or highly spherical—both the production yield and some bed characteristics (such as packing) seem to improve with a modest range of diameters and non-sphericity.

A variety of industrial processes are used for making spheres or pebbles 0.1–1 mm in size, but these are not necessarily suitable for fusion ceramic breeder materials. For better pebbles, the processes noted in Table 1 are the most developed. The physical characteristics of the resulting pebbles are given in Table 2.

The AECL process makes pebbles by extruding and rolling a paste of ceramic plus binder. The pebbles are subsequently sintered and separated by sieving. Out-of-size pebbles are recycled after the extrusion step. The process yields pebbles of moderate density. The process has been used with $LiAlO_2$, Li_2ZrO_3 and Li_2TiO_3, and

Table 1
Typical fabrication characteristics for best current processes

Group	AECL	JAERI	KfK
Process	Extrude, roll	Granule, agglomerate	Melt, spray, anneal
Diameter (mm)	1.2	1	0.1–0.2, 0.3–0.6
Size range (mm)	80%, 0.9–1.2	90%, 0.85–1.2	100%, 0.35–0.6
Batch size	3 kg product in 3 h (powder to green pebble)	—	5 kg product in 8 h (melt and spray)
Yield	90%	—	45%, pass 0.3–0.6; 10%, pass 0.1–0.2; 85% total
Material	Li_2ZrO_3, Li_2TiO_3	Li_2O, Li_2ZrO_3, Li_4SiO_4	Li_4SiO_4 + 2.2% SiO_2

Table 2
Typical pebble physical characteristics

Material	Diameter (mm)	Density (% theoretical density)	Grain size (μm)	Surface area ($m^2 g^{-1}$)	Crush strength (N)	Source
Li_2O	0.83–1.15	90–95	40	0.12	20 [a]	JAERI [8]
Li_4SiO_4	0.1–0.2	97	20–40	—	2 [a]	KfK [9]
	0.35–0.66	97	20–40	1.3	6–8 [a]	KfK [9,10]
	0.83–1.15	93–97	40	0.06	3 [a]	JAERI [8]
Li_2TiO_3	1–1.4	80	10–50	—	30–40 [b]	AECL [11]
Li_2ZrO_3	1–1.4	80–84	10	—	9–10 [b]	AECL [11]
	0.8–1.2	86	13	0.09	20 [a]	JAERI [8]

[a] Mean strength.
[b] Weibull strength (Weibull modulus typically 3–4).

also should be applicable to other materials. It provides a good yield at high production rates but cannot make smaller diameter pebbles. The pebbles were produced in collaboration with Ceramics Kingston.

The Japan Atomic Energy Research Institute (JAERI) process starts with small granules of breeder and binder, and agglomerates additional powder onto these until the desired diameter is reached. The pebbles are sintered and separated by sieving. The process has been used to make Li_2O, Li_2ZrO_3 and Li_4SiO_4 pebbles. The pebbles were produced in collaboration with Kawasaki and Mitsubishi.

The Kernforschungszentrum Karlsruhe (KfK) process produces pebbles by melting intimately mixed powders of Li_4SiO_4 and 2.2 wt.% SiO_2. The melt is sprayed into pebbles, with diameters of 0.02–1.8 mm, which are sorted according to their diameter, and the non-spherical and hollow particles are then separated by a wind-sifting machine. The pebbles are subsequently annealed to increase their mechanical strength. The rejected pebbles can be remelted and used again for an overall yield of 85%. The resulting pebbles are dense spheres with a smooth glass-like texture. However, the process is not readily adaptable to higher melting temperature lithium ceramics, such as lithium zirconate. The pebbles were produced in collaboration with Schott Glaswerke. It should be noted that the lower crush strength of these pebbles (Table 2) is at least partly because of their small size.

In summary, three different processes have been developed that are capable of producing interesting (but different) pebbles, with at least two processes capable of production rates suitable for fusion blankets. The present emphasis within the fabrication groups is on 0.1–0.2 mm and 0.2–0.6 mm for Li_4SiO_4 (KfK), 1.2 mm for Li_2TiO_3 and Li_2ZrO_3 (AECL), and 1.0 mm for Li_2O (JAERI).

3. Packing

A key characteristic for fusion breeder pebble beds is the overall effective solid or smear density. This is a combination of the internal density of the pebbles (discussed in previous section) and the packing density of the pebbles into the bed. The smear density needs to be high to maximize the lithium content of the breeder region (for good tritium breeding and minimum burn-up). Recent pebble bed blanket designs [1,3,5] have used breeder smear densities of 55%–62% for single-size breeder pebble beds and [3,9] breeder smear densities of 10%–14% in binary Be–breeder beds.

The pebble packing density (or packing fraction) is also important for the bed thermal conductivity and purge pressure drop. The general rule-of-thumb is that the minimum container dimension should exceed 10 times the pebble diameter, to ensure a reliable packing fraction of around 63%. Specific results with ceramic breeder pebbles have shown consistent or better packing behavior than that expected from this rule, except for Li_2O pebbles. Examples of packing experiences are summarized in Table 3.

Part of the good packing appears to result from geometrical arguments: packing is better if the most constraining container dimension does not fully enclose the bed. For example, breeder-out-of-tube geometry

Table 3
Packing fraction experience with single-size ceramic breeder pebbles

Pebble diameter (mm)	Packed bed diameter ratio, ID/OD (mm) [a]	Minimum diameter ratio	Smear density (% theoretical density)	Pebble density (% theoretical density)	Packing fraction (%)	Reference
Li_2O						
0.95	0/20	21	50	89	56	JAERI [12]
1.0	36/110	37	43	90	48	JAERI [13]
1.4	0/20	14	26	66	39	JAERI [14]
Li_4SiO_4						
0.35–0.6	16/102	94	62	93	67	KfK [15]
0.35–0.6	16/102	94	62	97	64	KfK [16]
1.0	0/60	60	45	97	46	JAERI 1991
Li_2ZrO_3						
0.5	—	—	52	86	60	TRIDEX-5
0.97	0/20	20	56	89	63	JAERI [12]
1.2	2.3/13.2	4.5	52	82	63	BEATRIX-II [17]
1.2	10/38	12	53	82	65	CRITIC-II [18]
1.2	0/3.8	3	43	84	51	University of Manitoba [11]
	0/6.7	6	52		62	
	0/9.6	8	57		68	
	0/11.6	10	56		67	
	0/12.6	10	59		70	
	0/25.2	21	57		68	
1.2	0/29	24	49	82	60	UCLA [19]
	0/24	21	58		70	
1.2	0/12.7	10	58	82	70	Spectrum [11]
Li_2TiO_3						
1.2	0/26	22	51	79	65	AECL [11]

[a] Packed bed length always much greater than pebble diameter.

may be more tolerant than breeder-in-tube geometry. Secondly, part of the packing behavior seems to result from the nature of the pebbles themselves. For example, the combination of the pebble shape, material and size distribution associated with the AECL fabrication process seems to allow the pebbles to flow readily together into packings that have significantly higher densities than those expected for randomly packed perfect spheres. Furthermore, the JAERI pebbles, fabricated by similar processes, show good packing for Li_2ZrO_3 but generally show poor packing for Li_2O. Thirdly, it should be noted that the pebble densities may not be known precisely, as a result of measurement limitations. Therefore, while the overall smear density is measured directly, the inferred packing fraction is uncertain.

Tests also have been conducted with binary beds, including large breeder–small breeder, large breeder–small metal and large metal–small breeder combinations. The metal pebbles tested were beryllium, aluminum and steel "simulants". It was observed that forming a binary bed is straightforward if the smaller pebbles are less than one-seventh (maximum diameter) to one-tenth (average diameter) of the diameter of the larger pebbles (see Table 4). Then, the large pebbles can be packed first, and the small pebbles dropped or vibrated down through the space between the larger pebbles.

In summary, packing of single-size or binary pebbles is straightforward; reproducible for a given product; and generally meets or exceeds theoretical expectations. Packing of Li_2O is surprisingly low, but this is probably compensated for by its high intrinsic lithium density.

Table 4
Packing fraction experience with binary pebble beds

Large pebble diameter (mm)	Small pebble diameter (mm)	Packed bed diameter ratio, ID/OD (mm)	Large pebble packing fraction (%) [a]	Small pebble packing fraction (%) [a]	Reference
2.0 Be	0.1–0.2 Li_4SiO_4	16/102	63	18 (97)	KfK [9]
2.0 Be	0.1–0.2 Li_4SiO_4 + Be	16/102	63	18	KfK [9]
2.0 Al	0.1–0.2 Li_4SiO_4	16/102	60	19 (97)	KfK [16]
1.2 Li_2ZrO_3	0.18–0.25	0/10.3	63 (84)	1 (100)	University of Manitoba [11]
	0.15–0.18		63	20	
	0.11–0.15		64	21	
	0.07–0.11		63	21	
	<0.07		64	22	
	Steel				
1.2 Li_2ZrO_3	0.2 Li_2ZrO_3	0/9.5	48 (84)	17 (86)	University of Manitoba [11]

[a] Pebble density indicated in parentheses.

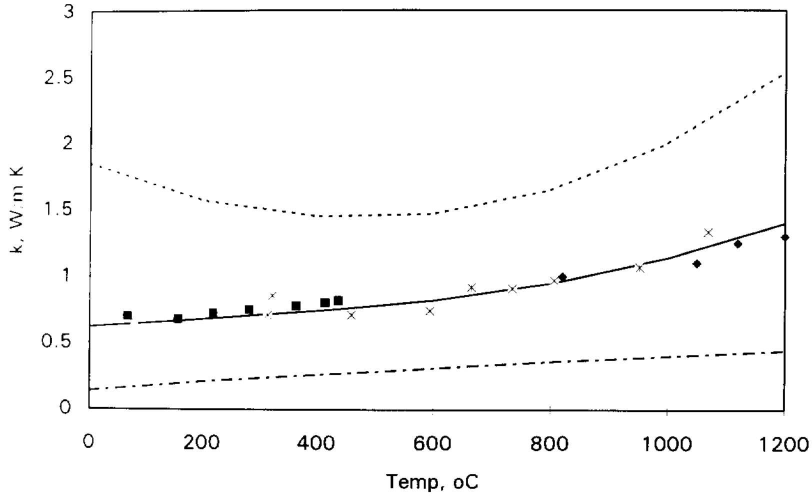

Fig. 1. Thermal conductivity of beds with Li_2ZrO_3 pebbles 1.2 mm in diameter in 1 bar He [11]: ■, Trent 1; ★, Trent 2; ◆, Trent 3; ○, UCLA; ---, 82% theoretical density Li_2ZrO_3; – · –, helium gas; ——, Schlunder model. (Conductivity of helium gas and of breeder material alone are also shown for comparison.)

4. Thermal conductivity

4.1. Bulk bed thermal conductivity

The thermal conductivity of pebble beds is critical for blanket heat transfer. The thermal conductivities of some ceramic breeder beds have been measured, alone or mixed with aluminum or beryllium pebbles. Figs. 1–3 summarize the breeder data under 1 bar He.

The bed conductivity for Li_2ZrO_3 pebbles 1.2 mm in size has been measured over the range 60–1200°C at Trent University (Canada) and UCLA. The tempera-

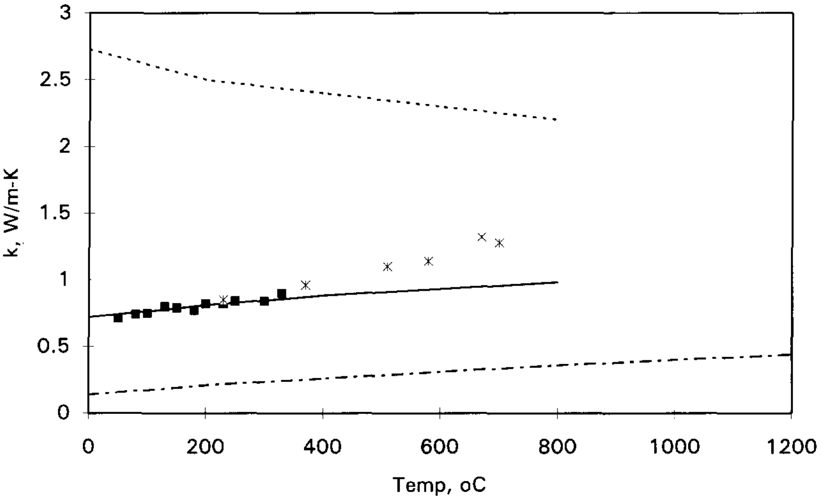

Fig. 2. Thermal conductivity of beds with Li_4SiO_4 pebbles 0.3–0.6 mm in diameter in 1 bar He [5,9]: ■, KfK 1; ★, KfK 2; ---, 97% theoretical density $Li_4SiO_4 + 13\%\ Li_2SiO_3$; – · –, helium gas; ——, Schlunder model. (Conductivity of helium gas and of breeder material alone are also shown for comparison.)

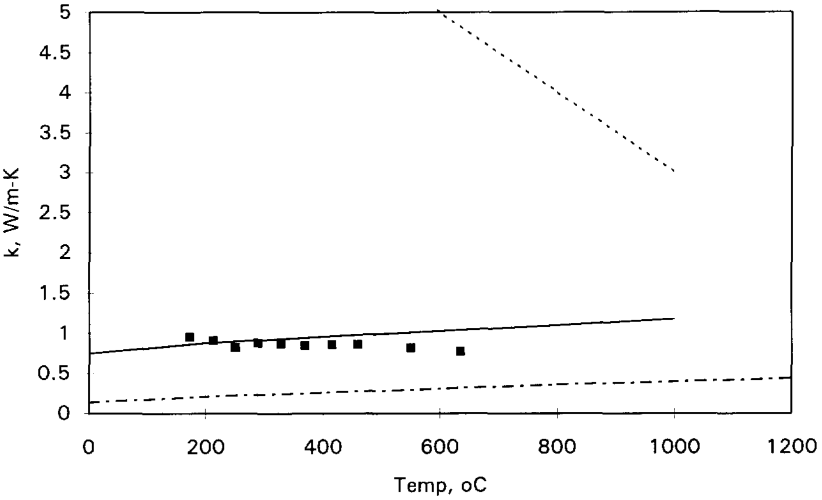

Fig. 3. Thermal conductivity of beds with Li_2O pebbles 1 mm in diameter in 1 bar He [13]: ■, JAERI; ---, 92% theoretical density Li_2O; – · –, helium gas; ——, Schlunder model. (Conductivity of helium gas and of breeder material alone are also shown for comparison.)

tures measured in the BEATRIX-II and the CRITIC-II in-reactor pebble bed tests are consistent with predictions based on these out-reactor data, and show no change up to 5% burn-up. Furthermore, the data are consistent with those for the bulk materials (ceramic and gas conductivity over this temperature range), and can be well-described by various semi-empirical models, such as the Schlunder model [15,20]. The Li_2ZrO_3 pebble conductivity also has been measured in 1 bar air and 0–3 bar He [11].

The bed conductivity for pebbles of Li_4SiO_4 (+2.2% SiO_2) 0.3–0.6 mm in size has been measured up to 700°C at KfK [5,9]. The measured conductivity increases faster above 400°C than is expected from semi-empirical models, such as those of Schlunder or Okazaki, or from the bulk material conductivity's temperature dependence.

Only preliminary data are available for Li_2O, because of experimental difficulties apparently resulting from the formation of LiOH in the apparatus [13]. The results are roughly consistent in magnitude with those expected from the Schlunder model.

The effect of mechanical loading on a pebble bed is an increase in the contact area between the pebbles and, therefore, in the bed conductivity. For the comparatively hard and low thermal conductivity ceramic breeders, this is not expected to be a significant effect. Measurements for Li_2ZrO_3 pebbles 1.2 mm in size confirm that there is no effect up to 1.3 MPa [19]. Similarly, data on Li_4SiO_4 show no loading effect at a differential radial thermal expansion of up to 0.2%.

The thermal conductivities of mixed metal–ceramic beds have been measured in support of blanket concepts utilizing mixed beds of beryllium and ceramic breeder. KfK has made such measurements with small Li_4SiO_4 pebbles mixed with larger aluminum and beryllium pebbles [9]. The mixed Be–Li_4SiO_4 beds were measured over the temperature range 30–500°C under 1 bar He gas. The thermal conductivity of unconstrained beds was found to increase from 2–3 $W\,m^{-1}\,K^{-1}$ for beryllium pebbles 2 mm in size to 4.7 $W\,m^{-1}\,K^{-1}$ with the addition of 17 vol.% Li_4SiO_4 pebbles 0.1–0.2 mm in size, and to 5.4 $W\,m^{-1}\,K^{-1}$ with 17 vol.% of Li_4SiO_4 and beryllium pebbles 0.1–0.2 mm in size added. In contrast to the pure ceramic pebble case, mechanical loading on the pebbles resulted in a strong increase in the overall bed thermal conductivity. In the KfK experiments, this occurred because of the differential thermal expansion between the pebbles and test cell of up to about 0.2%. Under this constrained condition, the mixed bed thermal conductivity increased to 8.5 $W\,m^{-1}\,K^{-1}$.

In summary, the single-size breeder pebble bed thermal conductivity is essentially predictable but is an important enough design parameter that experimental data should be obtained and cross-checked over the full temperature range. As expected, the bed conductivity is similar for the different ceramic breeders compared with the bulk material thermal conductivity. Information from in-reactor (Li_2ZrO_3) tests shows that the thermal conductivity can be stable up to at least 5% of burn-up, while out-of-reactor tests (Li_4SiO_4 and Li_2ZrO_3) show that the thermal conductivity is unaffected by mechanical loads, such as may arise from differential thermal expansion.

Binary beds of beryllium and ceramic increase the bed effective conductivity by a factor of at least 2, which is very attractive for reactor blankets. The database for the thermal conductivities for these beds is much more limited, and specific measurements are needed. The thermal conductivities of such mixed beds are also more sensitive to operating conditions, as illustrated by the variation in bed conductivity with pressure. However, although this may complicate the design, the conductivity increase with pressure naturally counteracts the effect of bed overheating or swelling (by reducing the bed temperature).

4.2. Near-wall bed thermal conductivity

Since the pebble packing and, therefore, the pebble–pebble contacts are different close to walls, the bed thermal conductivity is different in the near-wall region. This may be modelled as an effective interface conductance h, similar to the ceramic–structure "gap" conductance that is considered in thermal modelling of ceramic breeder blocks or fisssion fuel pellets. Alternatively, it can be modelled as an effective conductivity for the affected region near the wall, usually taken to be one pebble radius. The near-wall bed conductivity k_{nw} is related to the interface heat transfer coefficient h by

$$h = \frac{k_{nw}}{\{R(1 - k_{nw}/k_{bed})\}}$$

where R is the pebble radius (of larger pebbles if a binary bed) and k_{bed} is the bulk bed conductivity.

Table 5 summarizes the few experimental results available for ceramic breeder pebbles in terms of both h and k_{nw}. A few models have been proposed to predict this interface parameter but the database is still not sufficient to indicate which, if any, is best [20]. In fact, the effect is usually small in experiments, so it is difficult to measure accurately (the data quoted in Table 5 typically have large scatter or uncertainty bars). However, since the effect is comparatively small, the uncertainty is not of critical importance to blanket design. As with the bulk bed conductivity data, there is only a small effect of mechanical loading for pure ceramic beds (UCLA measurements on Li_2ZrO_3), but there is a significant effect with mixed Be–ceramic beds (KfK measurements on Be–Li_4SiO_4).

5. Purge gas permeability

A minimum purge gas throughput is required to keep the breeder tritium inventory low. The resulting purge

Table 5
Bed thermal conductivity data at the wall interface

Bed	Conditions	Interface h ($W\ m^{-2}\ K^{-1}$)	Near-wall k ($W\ m^{-1}\ K^{-1}$)	Reference
0.5 mm Li_4SiO_4	1 bar He, 100–400°C	2000–4000	0.3–0.4	KfK [15]
0.3–0.6 mm Li_4SiO_4	1 bar He, 200–400°C	6000	0.5	KfK [16]
2 mm Be + 0.1–0.2 mm Li_4SiO_4	1 bar He, 200–500°C, unconstrained	1000	0.8	KfK [9]
1.2 mm Li_2ZrO_3	1–2 bar He, 300–900°C	600–2000	0.2–0.4	Trent University [11]

Table 6
Range of purge gas pressure drop tests

Material and diameter	Packing fraction (%)	Packed bed size (mm)	Temperature (°C)	Reynold's number	Measured permeability (μm^2)
Li_2O 0.95 mm	56	20 diam. × 50	400–800	0.01–0.3, He	3300 [12]
Li_2O 1.4 mm	39	20 diam × 50	600	0.08, He	1900 [14]
Li_4SiO_4 0.3–0.6 mm	64	—	—	0.04–0.5, Ar, He	1110–1150 [24]
0.1–0.2 mm Li_4SiO_4 + 2 mm Al	79	—	—	0.002–0.07, He	630–700 [24]
Li_2ZrO_3 1.2 mm	43–70	3.9–25 diam. × 600	20	Low, He	880–2800 [11]
Li_2ZrO_3 0.97 mm	63	20 diam. × 50	600	0.04, He	1080 [12]
Li_2ZrO_3 0.2 mm	≈63	10 diam. × 50	20	Low, air	22 [11]
1.2 + 0.2 mm Li_2ZrO_3	64	10 diam. × 600	20	Low, air	14 [11]
1.2 mm Li_2ZrO_3 + 0.04–0.17 mm steel	82–85	10 diam. × 600	20	Low, air	0.6–5 [11]

gas pressure drop across the pebble bed is related to the packing fraction and the pebble size. For blanket purposes, this pressure drop sets a practical lower bound on the pebble diameter of around 0.1 mm, and also limits blankets to single or binary beds [6].

A number of correlations have been proposed to model the pressure drop across packed beds but these have not been studied systematically for fusion purposes. A convenient starting point in laminar flow is to separate the intrinsic bed factors from the blanket design parameters, i.e.

$$\Delta p = \frac{\mu v L}{k_p}$$

where Δp is the pressure drop, μ is the gas viscosity, v is the superficial gas velocity, L is the path length and k_p is the intrinsic bed permeability. This permeability is a characteristic of the packed bed, and may be obtained directly from experiments or estimated by a model such as the Carman–Kozeny models, i.e.

$$k_p = \frac{\varepsilon^3}{5S^2(1-\varepsilon)^2}$$

where ε is the bed porosity and S is the bed surface area per unit volume. For uniform spheres, we have

$$S = \frac{6(1-\varepsilon)}{D_p}$$

where D_p is the sphere diameter. For multi-size pebble beds, the formula is expected to be less accurate but can be used with a weighted average diameter, i.e.

$$\frac{1}{D_p} = \frac{\sum f_i}{D_i}$$

where f_i is the pebble volume fraction with diameter D_i.

The above expression for the pressure drop assumes laminar gas flow, i.e. a low bed Reynold's number

$$\mathrm{Re} = \frac{\rho v D_p}{\varepsilon \mu}$$

where ρ is the gas density and μ is the gas viscosity. This is appropriate for fusion blanket purge conditions. Energy length effects should extend only over regions a few times the inlet diameter, so should also not be significant. Gas flow along the walls rather than through the bed itself can occur for small columns but, in practice, this is only significant in columns that are less than several pebble diameters across; therefore, this is not relevant to blanket conditions [11].

Table 6 summarizes the range of data for lithium ceramic pebble beds, including data for binary beds of ceramic and mixed metal–ceramic pebbles [11,12,14, 24]. To illustrate the agreement with theory, Fig. 4 summarizes the measured permeability plotted against the ideal Carman–Kozeny permeability (i.e. uniform spheres),

$$k_{pCK} = \frac{\varepsilon^3 D_p^2}{180(1-\varepsilon)^4}$$

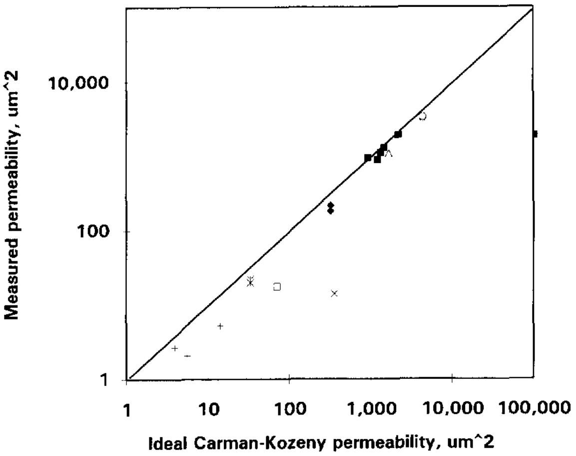

Fig. 4. Measured bed permeability vs. ideal Carman–Kozeny permeability for pebble beds of various diameters and materials (see Table 6 for references): ■, 1.2 mm, Li_2ZrO_3; △, 1 mm, Li_2ZrO_3; □, 0.2 mm, Li_2ZrO_3; ○, 0.95 mm, Li_2O; ●, 1.4 mm, Li_2O; ◆, 0.45 mm, Li_4SiO_4; ★, 2 mm, Al + 0.15 mm, Li_4SiO_4; +, 1.2 mm, Li_2ZrO_3 + 0.17 mm, SS; ×, 1.2 mm, Li_2ZrO_3 + 0.2 mm, Li_2ZrO_3.

The results are reasonably consistent with the ideal Carman–Kozeny model. The binary bed results show more scatter, as expected.

6. Chemical reactivity

Specific measurements on the compatibility of ceramic breeder pebbles with the coolant, multiplier and structure include the following:
- water reactivity with Li_2O, Li_2ZrO_3 and Li_4SiO_4 pebbles [8];
- out-of-reactor compatibility of beryllium, Li_4SiO_4, Li_2O and Li_2ZrO_3 pebbles [9,8];
- in-reactor (SIBELIUS) compatibility of beryllium and Li_4SiO_4 pebbles [21].

The compatibility of lithium ceramic pebbles is generally observed to be similar to that of ceramic pellets, for which a reasonable amount of literature exists [8].

7. Thermal–mechanical behavior

The largest static mechanical load on the pebbles is expected to be from differential thermal expansion. Theoretical models for elastic spheres indicate that completely constrained thermal expansion from 20 to

Table 7
Description of out-of-reactor thermal cycling test conditions

Pebbles	Temperature range (°C)	Ramp rate (°C s^{-1})	Pebble bed constraints	Number of cycles
KfK, 0.1–0.2 mm Li_4SiO_4 + 2.2% SiO_2	20–600 He	+5 to −54 peak	0.2 m × 18 mm diam. pin	⩽1000 [9]
JAERI, 1 mm, Li_4SiO_4, Li_2O, Li_2ZrO_3	400–800 He	±20	—	⩽2000 [8]
AECL, 1.2 mm Li_2ZrO_3, Li_2TiO_3	20–1000 air	±0.06	None	15–40 [11]
AECL, 1.2 mm, Li_2ZrO_3	50–700 air	±1 average	3 m × 13 mm diam. pin	65 [11]

600°C would impose loads of 0.2–2 N for pebbles 0.1–0.2 mm in diameter and 15–50 N for pebbles 1 mm in diameter. The load would be at the high end of these ranges for Li_2O and Li_4SiO_4, and at the low end for Li_2ZrO_3 and $LiAlO_2$, with Li_2TiO_3 at an intermediate value. The load increases with the pebble density and diameter.

Typical as-fabricated crush strengths of various ceramic breeder pebbles are summarized in Table 2. (Such measurements are often reported as simple mean values from a series of tests to failure, but should be presented in terms of the appropriate statistical average for ceramics, such as the Weibull distribution.) In comparison with the conservative load estimates above, it is clear that the pebble intrinsic strengths are close to these loads.

In a pebble bed, the bed thermal expansion and its effective modulus of elasticity are not necessarily simply related to the pebble intrinsic values. Therefore, tests in representative geometries are necessary. Also, real beds will be exposed to thermal cycling and temperature gradients, which will cause additional stresses. The maximum temperature ramp rates from rapid shutdown are about −20°C s^{-1} in the pebble bed region in the European breeder-out-of-tube DEMO blanket [2] and the JAERI ITER blanket [3].

Thermal cycling tests have been carried out on Li_2O, Li_4SiO_4, Li_2ZrO_3 and Li_2TiO_3 pebbles, as summarized in Table 7. In addition, pebble beds have been exposed to some thermal cycles during in-reactor tests—either from planned temperature changes or from reactor scrams (such as Li_4SiO_4 in DELICE, and Li_2ZrO_3 pebbles 1.2 mm across in BEATRIX-II and CRITIC-II).

The strength of the pebbles is a function of several factors, including the material and physical characteristics (such as the grain size and porosity), the fabrication process (such as the atmosphere) and the operating conditions (such as the temperature or the rate of change of the temperature). The effect of the pebble fabrication process is clearly indicated by comparing the thermal cycling results for JAERI and KfK Li_4SiO_4 pebbles; by KfK studies on the effect of SiO_2 addition and annealing on Li_4SiO_4 pebbles [10]; and by AECL tests on the effect of sintering conditions on Li_2ZrO_3 and Li_2TiO_3 pebbles [11].

The key design parameter for the mechanical integrity of the pebbles is a subject of current interest. Tests by KfK on Li_4SiO_4 and Be–Li_4SiO_4 pebble beds have been interpreted as indicating that the rate of change of the temperature is the most important parameter. Specifically, the KfK orthosilicate pebble beds with pebble diameters of 0.1–0.2 mm and 0.3–0.6 mm are resistant to cracking at temperature change rates of up to about −50°C s^{-1} at 400–500°C [9], which is quite sufficient for the worst shutdown scenario expected in the European breeder-out-of-tube DEMO blanket.

The effect of thermal cycling on causing settling and ratchetting of pebble beds also has been suggested as a concern, possibly leading to restricting the bed height as a design guideline. However, recent tests with pins 3 m high and with Li_2ZrO_3 pebbles did not show any settling at modest numbers of thermal cycles [11], while

Table 8
Pebble bed irradiation tests

	EXOTIC-6 [22]	EXOTIC-6 [22]	TRIDEX-5 [23]	BEATRIX-II [17]	CRITIC [18]	EXOTIC-7 1994
Material	Li_4SiO_4	Li_2ZrO_3	Li_2ZrO_3	Li_2ZrO_3	Li_2ZrO_3	Li_4SiO_4, Li_2ZrO_3
Pebble diam., source (mm)	0.3–0.5 KfK	0.3–0.5, KfK	0.5, KfK	1.2, AECL	1.2, AECL	0.15, KfK; 1.2, AECL
Mass (g)	—	8.2	4.8, 5.4	29.5	203	—
Grain size (μm)	10–30	10–40	10–40	≈10	≈10	—
Burn-up (at.% Li)	3	3	1.2	5.2	(1)	(10)
Temp. range (°C)	300–600	300–560	400	400–1100	200–900	—
Reference pure gas	He + 0.1% H_2	He + 0.1% H_2	He + 1% H_2	He + 0.1% H_2	He + 0.1% H_2	He + 0.1% H_2
Final H/T ratio	—	500	10000	140	180	—

tests with 0.2 m pins and Li_4SiO_4 pebbles did not show any dust transport after 1000 cycles at high helium flow rates [5].

In general, more work is required on the thermal–mechanical behavior of pebble beds, particularly under irradiation and in more realistic blanket module geometries and conditions.

8. Tritium release

Tritium release from ceramics is a complex phenomenon. Models to predict tritium release have had limited success so far and are not able to assess confidently the most important parameter, i.e. the tritium inventory in the blanket ceramic breeder. Therefore, experimental data are necessary. A useful irradiation database is now accumulating, and the results clearly depend upon the material, exposure history and microstructure. We anticipate that pebbles will have tritium release characteristics similar to those of monolithic materials with the same microstructure. Indeed, the data support this: for example, zirconate pebbles release tritium at low temperatures as do zirconate pellets. However, the most certain way to ensure the same microstructure—and the correct tritium inventory—is to test the as-fabricated pebbles themselves. Table 8 summarizes the results of tritium release tests using ceramic pebbles.

9. Conclusions

The extent of the database for lithium ceramic pebble beds has increased greatly in the past 5 years. The bulk of the experimental data applies to sizes of 1.2 mm for Li_2ZrO_3, 0.1–0.2 and 0.3–0.6 mm for Li_4SiO_4, and 1 mm for Li_2O, as produced by different groups. Mass fabrication and packing procedures have been established, and data are accumulating for the heat transfer, purge flow, mechanical behavior, compatibility and tritium release. Further measurements, expecially cross-checking of results from different manufacturers and test groups, will improve the database.

As a broad conclusion, the bed properties are generally as expected. Beds can be readily packed, and the resulting thermal conductivity and gas permeability are stable and reproducible. Irradiation data show stable bed temperatures up to 3%–5% of burn-up. The mechanical strength of the pebbles appears to be acceptable for KfK Li_4SiO_4 and JAERI Li_2O pebbles, but other materials need further improvement.

Based on these results, lithium ceramic pebble beds are suitable for use in fusion reactor blankets. The two most important remaining R&D needs are as follows: more irradiation data, in general, and more integrated or "engineering" tests to explore the bed behavior in representative blanket configurations and operating conditions.

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