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Development of a new cellular solid breeder for enhanced tritium production

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HIGHLIGHTS

- A new cellular solid breeder is presented with 2 to 3× the thermal conductivity and substantially higher density ($\sim$90%) compared with pebble beds.
- The cellular solid breeder contains an internal network of interconnected open micro-channels ($\sim$50 –100 μm diam.) for efficient tritium release.
- Cellular breeders are made by melt-infiltrating Li-based ceramic materials into an open-cell carbon foam followed by removal of the foam.
- High temperature (750 °C and 40 °C/mm) cyclic compression tests demonstrated good structural integrity (no cracking) and low Young's modulus of of <5 GPa.
- Deuterium absorption–desorption release rates were comparable with those from pebble beds with similar characteristic T-diffusion lengths.

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ABSTRACT

A new high-performance cellular solid breeder is presented that has several times the thermal conductivity and is substantially denser compared with sphere-packed breeder beds. The cellular breeder is fabricated using a patented process of melt-infiltrating ceramic breeder material into an open-cell carbon foam. Following solidification the carbon foam is removed by oxidation. This process results in a near 90% dense robust freestanding breeder in a block configuration with an internal network of open interconnected micro-channels for tritium release. The network of interconnected micro-channels was investigated using X-ray tomography. Aside from increased density and thermal conductivity relative to pebble beds, high temperature sintering is eliminated and thermal durability is increased. Cellular breeder morphology, thermal conductivity, specific heat, porosity levels, high temperature mechanical properties, and deuterium charging-desorption rates are presented.

1. Introduction

Lithium-containing ceramic breeder materials have been under development for several decades as a tritium breeding material and to convert energy into heat in solid breeder blankets. Early blanket conceptual design activities considered bulk ceramic breeder materials in form of sintered blocks [1–3]. However, the limited tritium recovery and thermal and radiation stability of sintered blocks, soon led to an alternative breeder configuration that of a pebble bed, where the breeder is in the form of small pebbles (0.5 to $\sim$1.5 mm diam.) and mixed with even smaller pebbles (0.1–0.3 mm) in order to maximize packing fractions (PF). The fundamental issue of using a sphere-pack configuration is the low thermal conductivity and the inherently low PF (<72% dense) results in low lithium to structure volume fractions, which result in marginal tritium breeding ratios (TBRs).

A new cellular breeder material is presented here with substantially higher densities and thermal conductivities relative to pebble beds. The cellular breeder contains a network of interconnected micro-channels, which are engineered during the fabricated process and which facilitate tritium release rates similar to those of pebble beds. Aside from increased density and thermal conductivity, high temperature sintering is eliminated and thermal durability

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is increased due to the morphology of the cellular breeder. Cellular breeders can be fabricated using any lithium-containing ceramic material, however here we report on the development of lithium zirconate (Li_2ZrO_3) as the first cellular breeder with Li_2TiO_3 to follow shortly.

Previous analysis and experiments on sintered blocks identified thermal stress induced cracking, as a feasibility issue leading to physical and thermal integrity concerns [4]. However, the newly developed cellular material appears to be stronger in that it is capable of sustaining prototypical temperature gradients without cracking. Because tritium release is a critical aspect of the solid breeder, a substantial fabrication effort was focused on control of micro-channel size and we show that the diameters of the tritium micro-purge channels can be tailored to meet breeder density requirements. A series of deuterium thermal desorption tests were conducted at Idaho National Laboratory (INL), which showed that release rates are efficient and comparable to those of packed beds. These preliminary characterizations encourage further development of cellular breeder materials for solid breeder blanket applications.

2. Cellular breeder fabrication

2.1. Fabrication process

Fig. 1 shows the schematic of cellular breeder fabrication. The new breeder is constructed by casting breeder material into a highly porous open-cell carbon foam skeleton. The carbon skeleton is then removed by oxidation, leaving a three-dimensional (3D) network of finely spaced interconnected and open tritium purge channels. Processing begins with reticulated vitreous carbon (RVC) foam. Various pore sizes are available from 3 to 130 pores per linear inch (ppi). RVC foam is 3 vol.% dense (97% open porosity), independent of pore size. The thickness of the foam ligaments is then increased by infiltrated with carbon using chemical vapor infiltration (CVI) until the material reaches a desired density (~10 vol.%). This step allows to vary the purge channel diameter/volume. Next, the breeder material, in powder form, is heated above its melting point and melt infiltrated (cast) into the carbon foam. The final step is to burn out, oxidize the carbon foam, leaving behind an interconnected network of purge channels for tritium. Just a single infiltration step is needed to completely fill the void space in the foam. Components up to 30 cm square ×7 cm thick have been fabricated.

2.2. Foam protective interlayer

Prior to melt-infiltration of the lithium-containing ceramic, a thin metal or ceramic interlayer is first applied to the foam to protect ligaments from reacting with the molten ceramic. A thin tungsten metal layer was applied by chemical vapor deposition to protect the foam during melt infiltration and to promote wicking of the molten ceramic. Fig. 2A–C shows 45- and 65-ppi tungsten-coated carbon foams used to fabricate cellular breeder specimens and a cut section of an individual 65-ppi carbon foam ligament coated with tungsten. The tungsten interlayer must be thick enough to prevent reaction between the underlying carbon foam and the lithium compounds but thin enough to prevent excessive strengthening of the foam. If the foam is too strong, stresses caused by thermal expansion mismatch with the lithium compound occur, resulting in micro-cracking of the lithium compound during solidification. The foam must be sufficiently weak that micro-cracking during solidification occurs in the foam ligaments rather than in the lithium compound. An optimal amount of tungsten of a few vol.% was established to prevent micro-cracking in the lithium compound. Some of the tungsten is removed during foam removal and the rest uniformly diffuses throughout the structure.

This tungsten should not inhibit permeation of hydrogen. However, neutron irradiation of this tungsten will create bulk defects that will retain tritium. The impact of tungsten on tritium release and microstructure evolution in the Li_2ZrO_3 cellular breeder material should be investigated further via neutron irradiation tests.

2.3. Initial cellular breeder samples

Through melt process optimization (temperature, pressure, time) and use of timed vibrations, Li_2ZrO_3 cellular breeder specimens with high overall densities of ~90% were produced. Fig. 3A–D shows SEM images of a Li_2ZrO_3 cellular breeder sample after removal of the carbon foam by oxidation. Li_2ZrO_3 was melted infiltrated into a 1″ diameter × 0.180″ thick carbon foam, after a thin tungsten interlayer was first deposited on the foam. The cellular breeder has an overall density of ~92.9% or a total porosity of about 8%, which consists of the micro-channels and closed pores. The channel spacing shown in Fig. 3 is similar to that in current pebble beds. The micro-channel width is about ~100 μm and the average cell diameter (Li_2ZrO_3) is ~850 μm, which translates to a maximum tritium diffusion length of ~425 μm. The geometric features of the Li_2ZrO_3 cellular breeder shown in Fig. 3 are relatively similar to those of pebble beds.

3. Cellular breeder characterization

3.1. Tomography

Efficient tritium release, i.e. release by convection requires a stable network of interconnected micro-channels that lead to the free surface of the cellular breeder. To verify the network of interconnected micro-channels, several Li_2ZrO_3 cellular breeder samples were sent for micro-tomography scanning at the High-Resolution X-ray Computed Tomography Facility at the University of Texas at Austin [5], which has an X-ray energy of 15 kV with a resolution of 5 μm. The dimensions of the 88.7% dense specimen were 0.165 × 0.165 × 0.071″, with a weight of 0.1289 g, a bulk density of 4.069 g/cm^3, a tungsten density within the specimen of 0.386 g/cm^3, and a Li_2ZrO_3 density within the specimen of 3.683 g/cm^3. An SEM image of a specimen is shown in Fig. 4A–B along with a superimposed tomography scan showing the structure of the interconnected micro-channels within the cellular breeder and Fig. 5 shows details of a tomography scan highlighting the structure of the interconnected micro-channels.

The micro-channels shown in Figs. 4 and 5 were made visible by post processing of tomography scans. It is interesting to note that the structure of the micro-channels is a “negative print” of the original carbon foam used for melt infiltration. The low level of closed porosity can be seen as small, scattered brown spots in Fig. 5.

The maximum distance between tritium purge channels (micro-channels) in the cellular breeder is dictated by the pore size of the foam skeleton used during construction. To date, a foam pore size of 0.8 mm has been successfully used in cellular breeder fabrication, resulting in a maximum tritium diffusion distance of 0.4 mm (comparable to the current solid breeder pebble diameter). In future work, processing may be feasible using foam with a 0.5 mm pore size which is available, yielding a further reduced tritium diffusion distance of 0.25 mm.

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