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Journal of Nuclear Materials 367–370 (2007) 1281–1286

journal of nuclear materials

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Status and perspective of the R&D on ceramic breeder materials for testing in ITER

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Abstract

The main line of ceramic breeder materials research and development is based on the use of the breeder material in the form of pebble beds. At present, there are three candidate pebble materials (Li_4SiO_4, and two forms of Li_2TiO_3) for DEMO reactors that will be used for testing in ITER. This paper reviews the R&D of as-fabricated pebble materials against the blanket performance requirements and makes recommendations on necessary steps toward the qualification of these materials for testing in ITER.

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1. Introduction

Worldwide research and development of ceramic breeder materials has been directed toward application in DEMO. ITER testing of the as-developed materials will offer performance checks and initial screening of their use in a fusion blanket. The quest to answer the questions of what confidence level we can have when extrapolating experimental results from ITER and in-pile fission tests to DEMO applications, and how to qualify materials for ITER testing to maximize this confidence, forms the central theme of this paper.

Ceramic breeder material is as important as the structure in a fusion reactor because it directly involves energy and tritium transport, both of which are critical to the functions of the blanket. Tritium release characteristics at high Li burn-up depend on the state of the interconnected pores and the grain size of the breeder material. The temperature gradient imposed in the breeder section can cause differential stresses, which may lead to cracking and fragmentation of breeder pebbles. If the breeder traps too much of the tritium produced, or the

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doi:10.1016/j.jnucmat.2007.03.240

cracking/fragmentation of pebbles blocks the purge gas flow line, breeder temperature control may be lost, causing the tritium inventory in the blanket to become a threat to safety, which may require early removal of the blanket. Moreover, the fabrication of such material needs to take into account the recycling process, not only to recover the expensive ^{6}Li for effective resource use, but also to remove radioactive isotopes.

As we proceed toward ITER testing, it appears there is a need to review and understand the development steps to qualify this material for testing in ITER. This is necessary not only to achieve experimental goals, but also to integrate testing results into the development line of DEMO. The status of fabrication processes being developed in various institutions and fabricated material properties are first reviewed. Experimental status of qualifying as-fabricated material with respect to functional requirements is then presented. The qualification focuses on the material's ability to fulfill tritium release requirements and withstand thermomechanical loading under large temperature gradients. The paper then presents a roadmap summarizing these qualification steps to guide continued materials development.

2. Ceramic breeder fabrication techniques

The main line of ceramic breeder materials research and development is based on the use of the breeder material in the form of pebble beds. The pebbles are quasi-spheroid, with small dimensions ($d < 1$ mm), because they have a better margin against thermal cracking, can easily fit into complex blanket geometries, and better accommodate volumetric swelling and expansion. EU and Japan have developed in the past several years three materials that are the candidates for their respective home DEMO reactors, which will be used for testing in ITER. They include the Li_4SiO_4 pebbles produced by melt-spraying [1] (EU-FZK with the collaboration of Schott Glas, Mainz), the Li_2TiO_3 pebbles produced by extrusion–spheronization–sintering [2] (EU-CEA with the collaboration of the industrial firm 'Céramiques Techniques et Industrielles'), and Li_2TiO_3 pebbles produced by a wet process [3,4] (JAEA, ENEA). In addition, SCICAS in China has produced small batches of Li_4SiO_4 as well as Li_2TiO_3 pebbles based on the extrusion–spheronization–sintering and binder-free wet methods, respectively [5].

Recent research on the extrusion–spheronization–sintering process focused on producing smaller Li_2TiO_3 pebbles in the range of 0.6–0.8 mm with a better sphericity, based on a revised formulation of the extrusion paste (binder and plasticizer content) as shown in Fig. 1. The study indicated that this diameter range is perhaps the lowest achievable by this process. The Li_2TiO_3 pebbles were sintered at 1100 °C and were characterized according to the size, shape, and microstructure using optical microscopy and scanning electron microscopy. Relevant characteristics, i.e., closed porosity, pebble bed density, grain size, and average crush load of the 2005 batches are listed in Table 1, showing that the open porosity decreases significantly and closed porosity decreases slightly for the latest smaller and higher-density pebbles.

'Wet methods' have been applied to the fabrication of Li_2TiO_3 breeder pebbles, making recycling

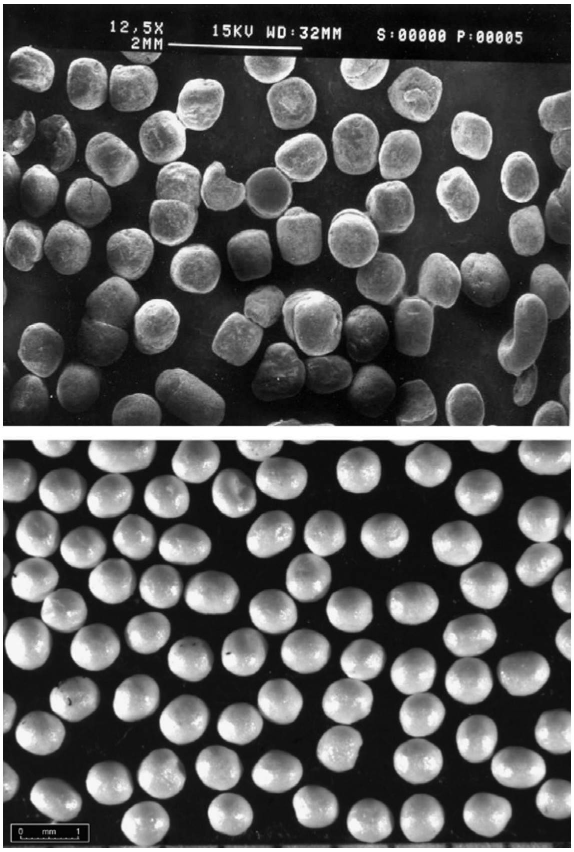

Fig. 1. Li_2TiO_3 pebbles produced by extrusion–spheronization–sintering process with size distribution ranging from 0.6 to 0.8 mm (top). A better sphericity of the pebbles has been achieved based on a revised formulation of the extrusion paste (bottom).

Table 1
As-fabricated pebble characteristics for CEA/CTI Li_2TiO_3 and FZK/Schott Li_4SiO_4

Pebble material	Pebble size (mm)	Pebble density (% TD)	Open porosity (%)	Closed porosity (%)	Grain size (μm)	Crush load (N)
Li_2TiO_3(CEA)						
CTI 273 batch	0.6–0.8	93.0	1.7	5.3	1–3	37 [14–65]
^{6}Li enriched	0.6–0.8	92.2	2.0	5.8	1–4	33 [25–52]
CTI 2964 batch	0.6–0.8	91.2	3.0	5.8	1–3	26 [15–42]
Li_4SiO_4 (FZK)					Specific surface area (m^2/g)	
OSi 03/2-8	0.25–0.63	93.7 ± 2.5	4.2 ± 0.7[a]	2.2 ± 2.0[a]	0.13	8.9 ± 1.8
OSi 03/2-9	0.25–0.63	94.0 ± 0.8	5.2 ± 0.3[a]	0.9 ± 0.8[a]	0.09	8.5 ± 1.9

[a] Data from Hg-porosimetry measurements.

of used breeder material more efficient. The direct wet process involves several steps: dissolving Li_2TiO_3 powder in H_2O_2 using citric acid as the chelating agent, condensing the solution, dropping the condensed solution into a solvent to form gel-spheres, drying and calcinations of the gel-spheres, and finally, sintering to improve pebble properties. In particular, the cracks in the gel spheres were almost entirely eliminated, while shrinkage occurred after sintering (1200 °C for 4 h) and surface morphology has been improved in comparison with the extrusion technique. Spherical pebbles of various sizes (0.5–1.5 mm) have been produced by these processes [3–5]; however, there is no published data on the characteristics of the pebbles fabricated.

At FZK, Li_4SiO_4 pebbles with a slight surplus of SiO_2 to enhance mechanical properties are produced by melting a mixture of LiOH and SiO_2 powders and then spraying the liquid material in air. The sprayed material solidifies during flight and is collected as pebbles with different sizes. The particle size distribution is strongly dependent on the flow rate of the melt, and only the size range of 250–630 μm is selected for blanket use. Due to the rapid quenching from the melt, most of the pebbles exhibit the typical dendritic microstructure at the surface, but there are some small glassy pebbles. For a similar pebble density of ~93%, the as-fabricated Li_4SiO_4pebble has a higher open porosity, shown in Table 1.

3. Out-of-pile thermomechanical testing of breeder pebble beds

Compressive crush load tests on multiple single pebbles provide statistical information on the mechanical stability of the pebble (Table 1); however, this does not assure their capability to withstand the combined temperature gradients and mechanical constraints in a prototypical blanket geometry. There is a need to verify the pebbles' mechanical stability by further thermomechanical tests on a larger scale as in the HELICA mock-ups for thermomechanical characterization of the reference EU lithium ceramic breeder pebbles [6]. The mock-up consists of a single welded cell made from ferritic-martensitic steel ASME SA 387 grade T91, which can be oriented to simulate vertical- and horizontal-blanket pebble bed configurations; the vertical configuration is shown in Fig. 2. The breeder cell, with two flat electrical heaters designed to reproduce the reference pebble temperature increase, is divided into three sub-cells 446 mm in width, 192 mm in depth, and 4.6 mm in thickness. Additional details can be found in Refs. [6,7].

The 2005 HELICA I test campaign was performed on Li_4SiO_4 pebbles with a narrower diameter range of 0.2–0.4 mm, as qualified by FZK [7]. The bed level is measured by a linear variable displacement transducer (LVDT) elastically pushed against the pebbles. The temperature distributions across the beds are measured by thermocouples located in various locations. The pebble filling was performed by pneumatic hammering at high frequency, reaching a very high packing factor (65.6% compared to a typical value of ~62%). However, an overall bed height reduction was measured in the vertical oriented mock-up caused by gravity. Even the increase in packing density of +0.2% was considered negligible after 20 thermal cycles, this irreversible bed height reduction has not yet reached an asymptotic value. Post examination of pebbles

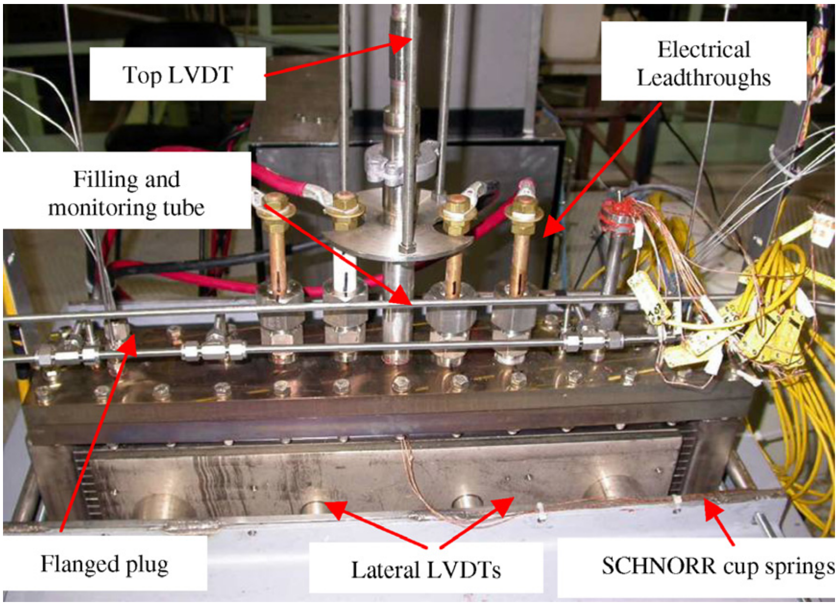

Fig. 2. HELICA I mock-up in vertical pebble bed configuration.

shows that about 0.7% of the fragmented pebbles (by weight) have sizes below 138 μm [7]. The implication of the broken pebbles for blanket performance is not yet addressed.

4. In-pile EXOTIC and PBA tests on tritium release and mechanical stability

The EXOTIC irradiation experiments in the high flux reactor (HFR) at Petten were designed to investigate tritium release properties, tritium residence time, and mechanical stability of the fabricated pebbles at different levels (up to DEMO) of Li burn-up. In the EXOTIC-8 experiment, lithium orthosilicate pebbles with 50% ^{6}Li-content irradiated up to about 11% ^{6}Li burn-up showed an increase in smaller cracks in the bulk and the presence of larger through-cracks [8]. The tritium release characteristics of the latest high density Li_2TiO_3 pebbles supplied by CEA were studied in the EXOTIC-9 experiment [9] by differential inventory from in-pile thermal transients and post-irradiation inventory measurements. Preliminary results of EXOTIC-9/1 showed that increasing pebble density in Li_2TiO_3 leads to a higher tritium inventory build-up as shown in Fig. 3 [9]. The new finding on its in-pile

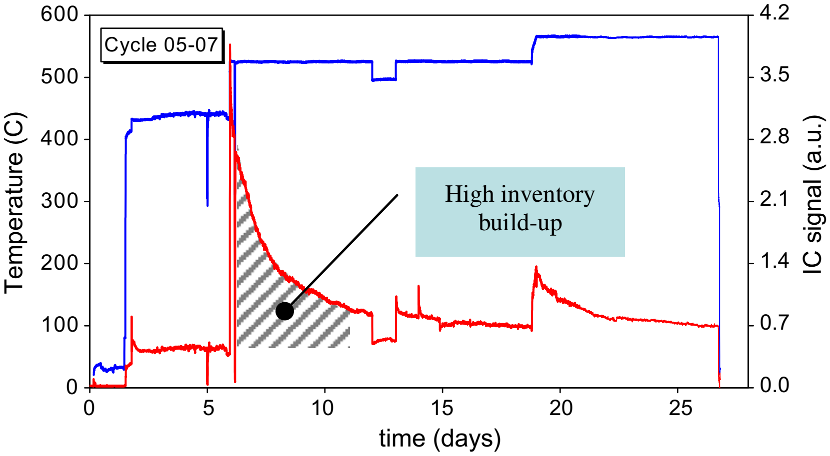

Fig. 3. Preliminary results of EXOTIC-9/1 showed that increasing pebble density in Li_2TiO_3 leads to a higher tritium inventory build-up.

tritium release characteristics suggests a design optimization between ^{6}Li density and tritium inventory.

In-pile pebble bed assembly (PBA) irradiation experiments were launched to study the effect of neutron irradiation on the thermomechanical behaviour of a ceramic breeder pebble bed while achieving DEMO-representative temperatures with defined thermomechanical loads [10]. This series of 12 cycle experiments was completed in November 2004, with an accumulated irradiation time of ~7200 h, which gave approximately 2 dpa in Eurofer and a total lithium burn-up of about 2–3%. To trace the thermomechanical state of the pebble bed during irradiation, neutron radiographs were made before the start of in-pile operation, after the first cycle, after the 8th cycle, and at the end of irradiation. The best image of neutron radiography shows no evidence of large gap formation (Delta > 0.05 mm); this will be confirmed in detail in the post-irradiation examination [11].

5. Testing in ITER toward DEMO development

In the development of ceramic breeders for use in a DEMO reactor, some issues have not yet been completely addressed, e.g., the question of the maximum allowable temperature at high fluence/high constraint for candidate ceramic breeder materials. The current knowledge base includes tritium release data under low constraint conditions and temperatures up to 700 °C. These data are at DEMO relevant burn-ups, i.e., up to 11% for Li_4SiO_4 and 17% for Li_2TiO_3. In addition, from a thermochemical stability point of view, the spinel phase ($Li_4Ti_5O_{12}$) present in stoichiometric Li_2TiO_3 pebbles can react with H_2 in the purge gas and decompose into a completely different Li-oxide at high temperature [12]. Tests for exploring high temperatures under a prototypical ratio of dpa/^{6}Li burn-up have already been planned as in high fluence irradiation ceramic breeder pebble stacks tests (HICU) [13] for all three reference materials.

The effects of pebble microstructure on tritium release and mechanical integrity and stability need to be verified. Some performance tests have been presented in this paper, but a large mock-up test program should be performed to reduce development risk. The results of out-of-pile performance tests have motivated some recent studies of heat treatment on amorphous Li_4SiO_4 pebbles to induce

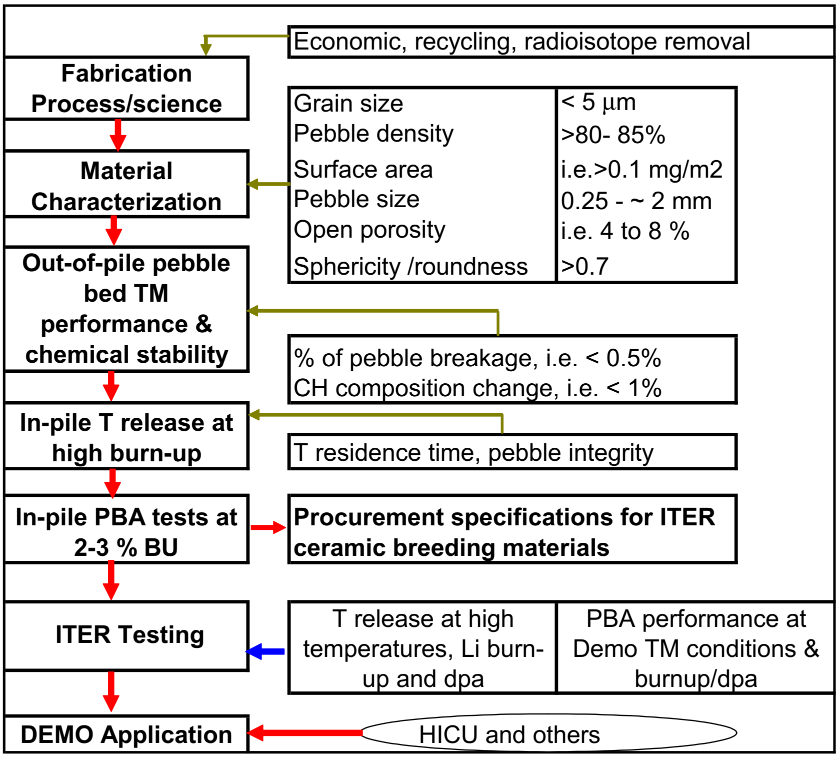

Fig. 4. Outline of a ceramic breeder material development roadmap prior to ITER testing is proposed.

crystallization of ortho- and meta-silicate without introducing pores and cracks [14], and the development of oxide-doped Li_2TiO_3 for better grain size and chemical stability [15]. Their impacts on blanket performance, such as the occurrence of a small amount of fragmented particles, have yet to be quantified.

The test blanket module (TBM) irradiation in ITER is an important step to investigate beginning-of-life (BOL) performance aspects of the ceramic breeder behavior in a fusion environment. Nevertheless, the licensing requirements for the ceramic breeder may not be critical because its malfunction or inadequate performance will not disturb ITER operations. However, since ITER testing is an extremely costly exercise and is most likely the only opportunity to test a fusion blanket prior to DEMO, the questions of the choice of materials for the ITER TBM and the definition of a set of requirements (and the related qualification program) to assure safety, reliability, and test performances become particularly important. Accordingly, this paper proposes a roadmap (Fig. 4) outlining the necessary development steps for qualifying and accepting the pebbles for ITER and fusion applications. For each development step, a set of criteria is presented as a means for initial screening before proceeding to the next evaluation tests in order to reduce development costs. However, it is important to recognize that ITER conditions (e.g., lower fluences) are far from sufficient to qualify any specific breeder material to be used in DEMO. Thus, parallel with ITER and subsequent to ITER testing, tests such as HICU or in fusion relevant neutron sources like IFMIF for any candidate ceramic breeders under typical reactor blanket conditions with relevant nuclear environment are necessary for this purpose.

6. Summary

Ceramic breeder material R&D has been geared toward DEMO application. A ceramic breeder material development roadmap has been described in this paper based on the accumulated progress, especially R&D of Li_4SiO_4 pebbles. The roadmap provides a systematic and economical approach to guide fabrication processes and materials characterization. It identifies specific needs for out-of-pile and in-pile experiments prior to the final design of ITER TBM and the ceramic breeding material specification for TBMs in order to reduce programmatic risks for not achieving its experimental mission such as any unanticipated system performance leading to reduced or unquantifiable operating conditions. Further development of this roadmap involves completing the criteria matrices for each development step.

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