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Manufacturing Challenges Of Fusion Pebbles Bed Material

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ABSTRACT

The thermal stress on breeding blanket structure is one of the main design driver; consequently, an open issue for fusion power reactor is the choice of breeding blanket material.

The mostly promising and worldwide-investigated solution refers to the ceramic material, e.g. $Li4SiO4$, in form of pebble beds instead of a bulk form such as a block or a disk. This would introduce some advantages, especially for what concerns the heat transfer phenomena, low activation characteristics, low thermal expansion coefficient, high thermal conductivity, etc.

In this paper, particularly the reliability/availability of a methodology capable to produce stable and well-sized ceramic pebbles is a major challenge as well as the evaluation of the thermal conductivity, that is a necessary input data for the understanding of blanket behavior (support the thermal-structural and thermo-hydraulic analyses).

As for this latter, moreover, a sample holder considering the heat transfer mechanism through the pebble-bed was duly designed such to determine the pebble-bed thermal conductivity by means of a hot rig with guard resistance method.

This paper introduces (and analyses) preliminary results of the effective thermal conductivity on the pebble-bed as well as a description of the adopted methodology.

This research activity was developed in the framework of PRA2016 project.

1 INTRODUCTION

Many blanket designs (considered actually still a challenge) for fusion reactors foresee the use of lithium as promising tritium-breeding materials with inherent safety and potential higher efficiency and rely on the development of advanced structural materials compatible with high temperature operation (very severe working condition).

Specifically, this material following the capture of a neutron by the natural isotope Li-6 allows the production of tritium, 6Li (n, t) 4He, new fuel for nuclear fusion reaction.

In this paper we focus on the orthosilicate (Li4SiO4) in form of pebble beds (Figure 1).

Despite the uncertainty about the release of tritium and effects induced by irradiation, the structure of the Li4SiO4 is very interesting because of the high density of lithium, low thermal expansion coefficient, high stability and thermal conductivity [1][2]. This latter in particular is widely investigated ([3][4][5][6]) because it may greatly influence the power generation. In addition, the thermo-mechanical behavior of the packed pebble beds is different from that as bulk material and depends to other parameters, such as pebble size, packing density, etc.

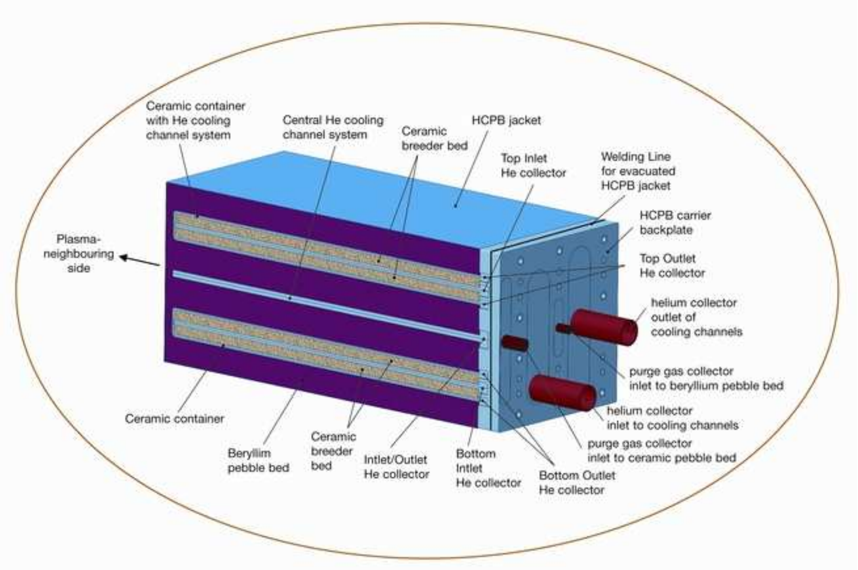


Figure 1: One blanket scheme with main components; the layers in grey are the pebbles bed.

Ceramic breeder pebbles have been long investigated in the recent years [6][7][8]**Errore. L'origine riferimento non è stata trovata.** and are currently a focus for R&D, particularly for what concerns the physical-chemical and the thermo-mechanical properties of pebbles as well for the overall blanket module behaviour.

In this paper we will describe the actual fabrication process and methodology used for the fabrication of stable and well-sized ceramic pebbles and the new as well as methods for the characterization of the thermal conductivity.

2 MANUFACTURING METHODS

A variety of industrial processes, even if far from industrial standard, has been proposed for making 0.1-1 mm orthosilicate pebbles diameter and of desired properties [10][11]. Nevertheless some manufacturing problems were observed like e.g. the chemical stability (agglomeration phenomena) and material degradation during storage. This because recently, the hygroscopic nature of lithium ceramics has been identified as a critical factor conditioning the manufacturing processes.

In what follows we will first provide insight on the main pebbles production methods, with advantages and drawbacks and then we will present the fabrication process that the research group of the University of Pisa is developing jointly with Industrie Bitossi.

2.1 Pebble bed fabrication process

The melt-spraying process [10][11], developed at KIT (EU) and in China, is based on a rapid quenching of liquid droplets of melt LOS (1400 °C) in air, produced by using either lithium hydroxide (LiOH) or lithium carbonate (Li2CO3) as raw material, together with silica (SiO2). This method allows to obtain high density (>90% TD), low porosity and spherical shaped pebbles of 0.25-0.63 mm size, with the possible direct re-melting as an option for reprocessing.

The melt-spraying process demonstrated to be unsatisfactory because the rapid quench was responsible of the presence of a second phase (Li2SiO3) and dendritic formations on the surface of the pebble. A further modification of this process aimed at increasing the crush load was made by introducing lithium metatitanate phase (up to 20% mol)[13].

Wet methods of production without melting raw materials was set up by Gao et al. [14]who used Li_2CO_3 and SiO_2 to produce LOS powder by the well-known solid state reaction in aqueous solution at 700 °C for 12 h. After polyvinyl alcohol addiction (binder), spheres formed during the dropping into liquid nitrogen and subsequently sintering: the best results were obtained at 1020 °C (density 80% TD, average crush load of 20 N and averaged grain size of about 5 µm) for 0.5-0.8 mm pebbles size. The main problem of this process was the pebble size and roundness to guarantee.

A different water-based sol-gel technique was proposed by Wu et al. [15][16] consisting in mixing SiO_2 and LiOH in water, adding citric acid as chelating agent to obtain a gel which is subsequently dropped, dried, calcined (675 °C for 4 h) and sintered (900 °C for 4 h). Citric acid was used to guarantee a reasonable value of pH (8.5), otherwise the pH level grows up to 12-13 (too basic for sol/gel applications).Despite pebbles had 0.8 to 2.0 mm diameter, with up to 74% TD, they were high porous (and thus low resistant) because of the citric acid effects.

Other processes consider sol-gel method at least starting from solid state reaction at room temperature to obtain 1.18 mm diameter pebbles of 89% TD [17].

In materials fabricated using methods of Table 1, it was observed the phenomenon of lithium loss through volatilisation, even if in different way, caused by the hydroxide precursor. This loss and the excess of lithium necessary to correct for this, as in [17], are dependent on pebble parameters and firing conditions.

Table 1: Pebble fabrication processes with characteristics.

Fabrication Process	Raw materials	Main process phases	Diameter and density
Melt-spray [10] [12] [13]	LiOH + SiO_2	1. Melting 2. Dropping 3. Quenching 4. Annealing (2h@1000°C)	Φ: 0.25÷0.63 mm (50%) ρ > 0.90 TD
	Li_2CO_3 + SiO_2	1. Melting 2. Dropping 3. Quenching 4. Annealing (2h@1000°C)	Φ: 0.8÷1.0 mm (50%) ρ > 0.90 TD
Sol-gel [15][16]	LiOH in citric acid suspension ($C_6H_8O_7$) + SiO_2 (aerosol)	1. LiOH + citric acid + water 2. Silica addition 3. Vaporizing @70 °C 4. Dropping into acetone 5. Calcining + sintering	Φ: 1.2 mm ρ = 0.74 TD

		(4h@900 °C)	
Capillary-based microfluidic [17]	$C_2H_3O_2Li \cdot 2H_2O$ + SiO_2 lithium acetate dihydrate	1. Stirring raw materials. 2. Inlet T junction with silicon oil 3. Vaporization and drying 4. Silicon oil removal (72 h@120 °C) 5. Calcining + sintering (4h@750 °C)	Φ: 0.84 mm ρ = 0.82 TD
Wet process with substitution [10]	LMT powder	1. Dropping LMT + sodium alginate + 4HF in zinc chloride 2. Calcining 3. Sintering	Φ: 0.238 mm ρ = 0.89 TD

2.2 University of Pisa pebble bed fabrication process

The innovative character of the new methodology under development at the Department of Civil and Industrial Engineering (DICI) of the University of Pisa jointly with Industrie Bitossi is based on the drip casting method (main phases synthetized in Figure 2): pebbles are produced by dripping the ceramic suspension through a nozzle plate into a wet solution where droplets coagulats before calcination and sintering.

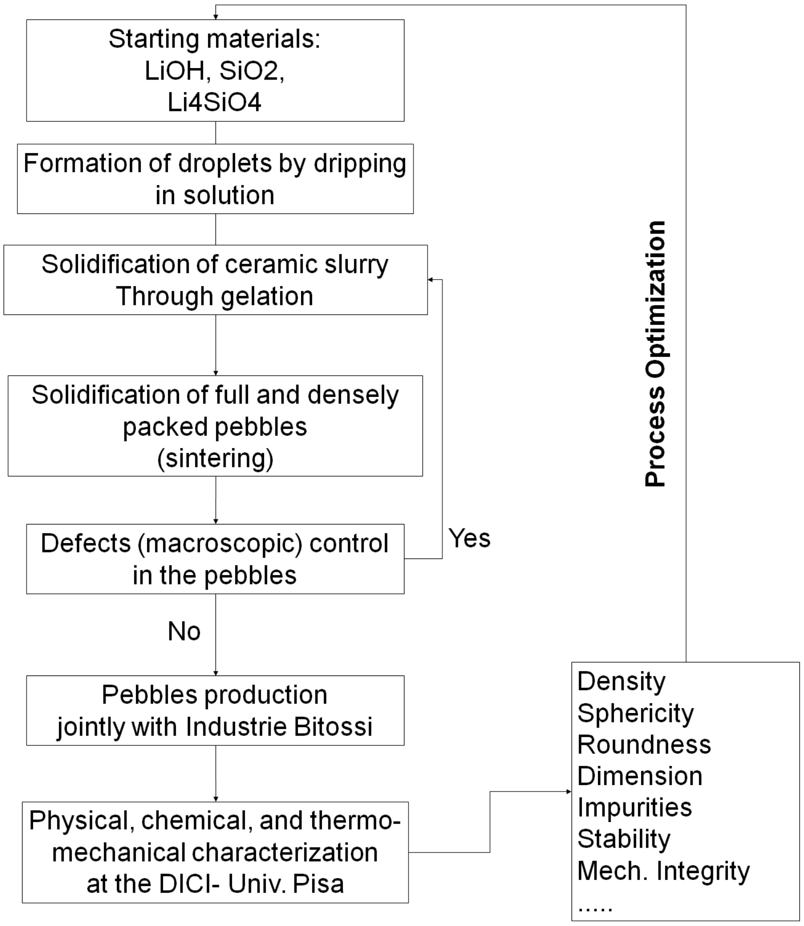


Figure 2: Flow diagram with the main phase of the proposed fabrication process.

It is a wet process with substitution reaction of the liquid mixture (made starting from orthosilicate powder) and alginate solution as the binder, allowing to generate gel-spheres (Figure 3). Calcination and sintering allow then pebbles to reach the requested high density.

Figure 3: Drip casting process: cross linking in alginate gels involves cooperative bonding of divalent metal ions.

Drip casting process is alternative to the “melting” process; we may obtain thus stable Li4SiO4 with more accurate and controlled spheroidal particle sizes.

This process will allow to control accurately not only the geometry of pellets, by the viscosity of the liquid mixture, nozzle diameter, and wettability between the material of nozzle and liquid mixture, but also their density, coherence and chemical stability so as to limit the surface interaction H2O-Li4SiO4 (responsible of detriment of pebbles properties [1]). Another aspect not less relevant is the possibility to control and minimize impurities, that in some kind could jeopardise the advantages of using low-activation materials.

2.3 Pebble characterization

The characterization of Li4SiO4 pebbles is scheduled so to determine density, microstructure and crystal form (by SEM and X-ray diffractometer), and morphology. After that, the thermo-mechanical characterization needs to be carried out.

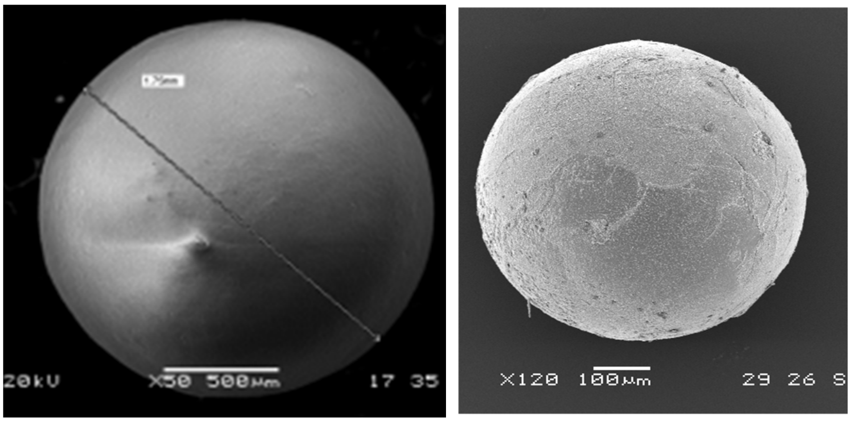


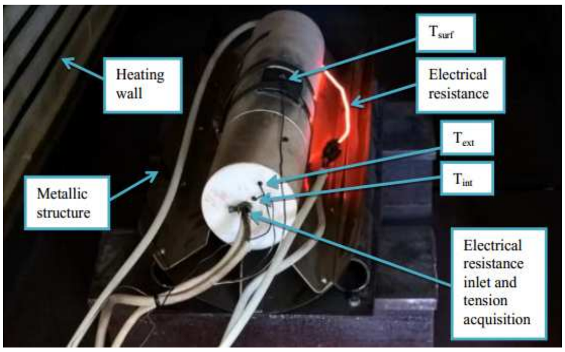
Figure 4: Shape of an orthosilicate pebble by SEM

Since ceramic materials, by their nature, are brittle and prone to cracking under external mechanical loads, the evaluation of the collapse load is felt important in consideration of the strong mechanical loadings to undergo in the blanket structure (due to the differential thermal expansion between breeder pebbles and retaining structure).

Such an understanding of the stress-displacement behaviour, obtained by performing cyclic compression tests and crushing tests with Instron apparatus, is necessary to providing confidence in the performance and lifetime of pebbles (and in turn for breeder blanket design) as well as to evaluate the influence of the chemisorption products on the mechanical properties, such as stress-displacement, ultimate load, and failure modes of pebbles.
Experimental data will be also used for the development of reliable modeling tools.

The knowledge of the thermal conductivity is an essential key issue to be investigated for a proper thermo-mechanical blanket design and for assessing heat transfer processes. It is also influenced by the deformations due to the different thermal expansions between bed and the other structural materials.

Several numerical simulations, as pre-tests, have been carried out to simulate the mechanisms of heat transfer in pebble beds and optimize the operational test conditions. The thermal conductivity is instead measured by performing either hot wire test (bulk material) and hot rig test. To better characterize the effective thermal conductivity of the pebbles bed, experimental tests will consider a wider range of temperatures and pebbles diameters.



a)

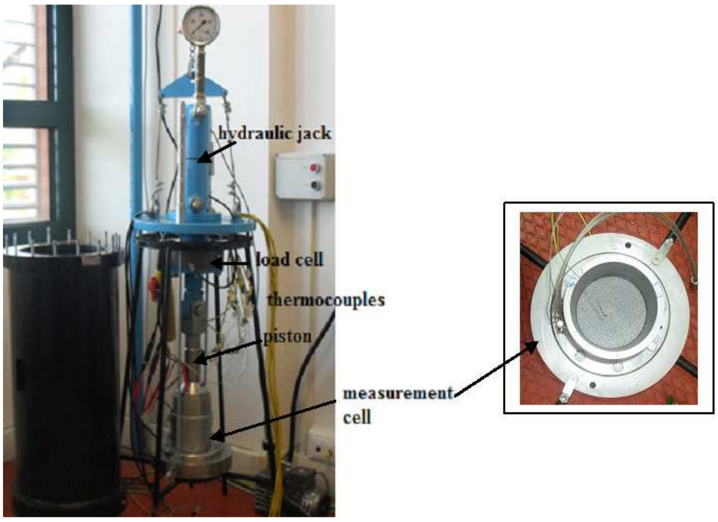


Figure 5 a, b: a) Hot Wire apparatus and b) hot rig facility

The experimental test will be carried out on the same diameter pebbles so to relate the thermal conductivity to the geometrical characteristics. In addition it is foreseen to perform tests with different loading (with and without compression) at same temperature conditions (from ambient temperature till 800°C). The procedure may be summarized as follows:

a. Heating measurement cell containing pebbles bed until steady state condition is reached;

b. Switch on the cooling system to generate an axial heat flow through the bed thickness;

c. Compensation of the heat flux radial losses (depending on the radial distribution of temperature);

d. Application or not (depending on the type of tests) of the compression load;

e. Evaluation of the bed thermal conductivity based on measured reference temperatures.

The activity related to the fabrication and characterization of pebbles is not finished yet. Results will be presented and discussed as soon as possible.

3 SUMMARY

The thermal stress on breeding blanket structure is one of the main design challenges to face; consequently, the choice of the most appropriate breeding blanket material becomes extremely important

In this paper, we presented some methodology used to fabricate ceramic pebble beds and the new fabrication method under development at the DICI - University of Pisa jointly with Industrie Bitossi based on the drip casting method (see Figure 2 for diagram with the main process phase).

The novelty relies in adapting this method, already used for conventional purpose, to the needs of the critical working condition of pebble inside the fusion reactor blanket module. The drip casting will offer advantages related to the control of critical parameters affecting and influencing blanket operation, such dimensions, roundness, sphericity, chemical stability, density, impurities etc.

As for the thermo-mechanical characterization we presented the experimental campaign necessary for a broad understanding of the pebbles performance, which consist of compression test (cyclic and collapse) and thermal conductivity evaluation tests (by means of a hot rig with guard resistance method).

Experimental test results will be presented soon the research activity related the manufacturing and characterization of fusion pebbles bed material will be completed.

ACKNOWLEDGMENTS

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