Fusion Engineering and Design 167 (2021) 112388

Contents lists available at ScienceDirect

Fusion Engineering and Design

journal homepage: www.elsevier.com/locate/fusengdes

Preliminary investigation of Li4SiO4 pebbles structural performance

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A R T I C L E I N F O

Keywords:
Blanket
Breeder material
Lithium orthosilicate
Sol-gel method
Compression tests

A B S T R A C T

One of the main purposes of a breeder blanket is to remove the heat produced in the blanket by the fusion reaction neutrons, and to breed the tritium required to sustain it. To achieve these requirements, several breeder materials (solid or liquid lithium-bearing ones) have been investigated in the past decades.

To date it has not yet been possible to identify a stable material, with high thermal conductivity and melting point.

This paper deals with the mechanical characterization of the lithium orthosilicate (Li_4SiO_4) in form of pebbles, produced at the University of Pisa at room temperature by a drip casting forming technique, starting from an aqueous suspension of Li_4SiO_4 precursor prepared by a sol-gel synthesis method.

To investigate also numerically, by means of FE code, the breeder blanket behaviour, it is of meaningful importance the mechanical characterization of such pebbles.

To the purpose, either static or cyclic uniaxial compression tests, without radial constraints, have been performed on several produced pebbles of about 1.5 mm diameter in order to determine the collapse and crushing loads and the stiffness. Moreover, the carried-out post-test SEM examination allowed to evaluate the failure mode and the crack shapes on the contact surface.

Results show the influence of the elastic properties and matrix flaw population on the crushing load.

The pebbles produced by the sol-gel method showed also a high strength, the value of which is comparable to that of the pebbles obtained by melting process.

1. Introduction

In the last decades, ceramic tritium breeding materials have attracted remarkable attention from both experimentalists and theoreticians. This is evidenced by the growing number of research articles on them [1–10]. Lithium-containing ceramics such as Li_2TiO_3 and Li_4SiO_4 have been proposed as the main tritium breeder candidates [11]. However, the concepts developed by China, Europe and South Korea feature lithium orthosilicate as the designated solid breeder material [7].

There are many methods used for the fabrication of 0.1−1 mm diameter pebble the main aim of which is to combine favorable material properties such as the lithium density and the mechanical stability. Additionally, the pebbles offers several advantages such as the adaptability to complex structures, acceptable tritium release property and suitable effective thermal conductivity.

Hoshino et al. proposed to fabricate Li_2TiO_3- Li_4SiO_4 composite ceramics by solid state reaction [6,8,9]. Togashi et al. observed that Li_4SiO_4 can easily absorb CO_2 to form Li_2CO_3 and Li_2TiO_3 [12]. Lo Frano et al. fabricated successfully pebbles by hydrolytic sol-gel method starting from LiOH and $Si(OCH_2CH_3)_4$ (Fig. 1), as described at length in [14].

The higher density of pebbles may improve the mechanical stability and reduce thermal stresses. Nevertheless it is really challenging to densify Li_4SiO_4 (theoretical density-TD- more than 60 %) with good crushing load. As shown in [13], at low temperatures (200–400 °C) the lithium atom density and tritium release of Li_4SiO_4 were higher for in comparison to Li_2TiO_3.

In this paper we investigate experimentally and numerically the mechanical performance of Li_4SiO_4 pebbles, which were produced by drip casting technology at room temperature.

It is worthy to remark that the mechanical properties of pebbles are strongly dependent on the fabrication process; a high crushing strength is desirable in order pebbles may withstand the significant thermo-mechanical forces generated during the fusion reactor operation.

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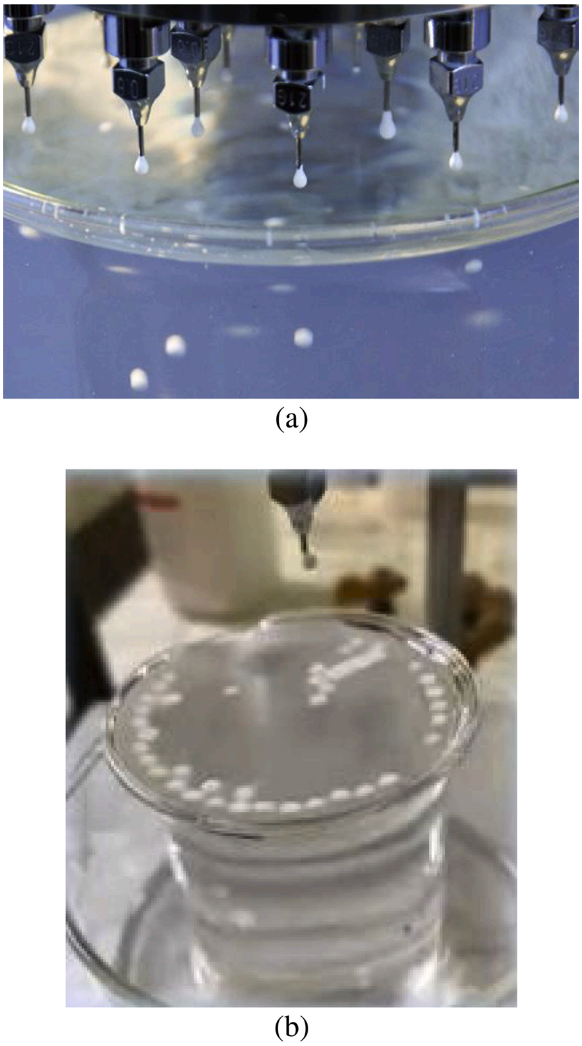
https://doi.org/10.1016/j.fusengdes.2021.112388
Received 4 December 2020; Received in revised form 11 February 2021; Accepted 19 February 2021


(a)

(b)

Fig. 1. a, b: Drip casting process phases: a) diffusion through bottom feeder needles and b) formation of cross linking (due to the bonding of divalent metal ions) and gelification.

2. Li4SiO4 pebbles mechanical characterization

In our previous work [14] we presented the feasibility of Li_4SiO_4 pebbles, which were obtained by hydrolytic sol-gel method starting from lithium hydroxide and tetraethyl orthosilicate.

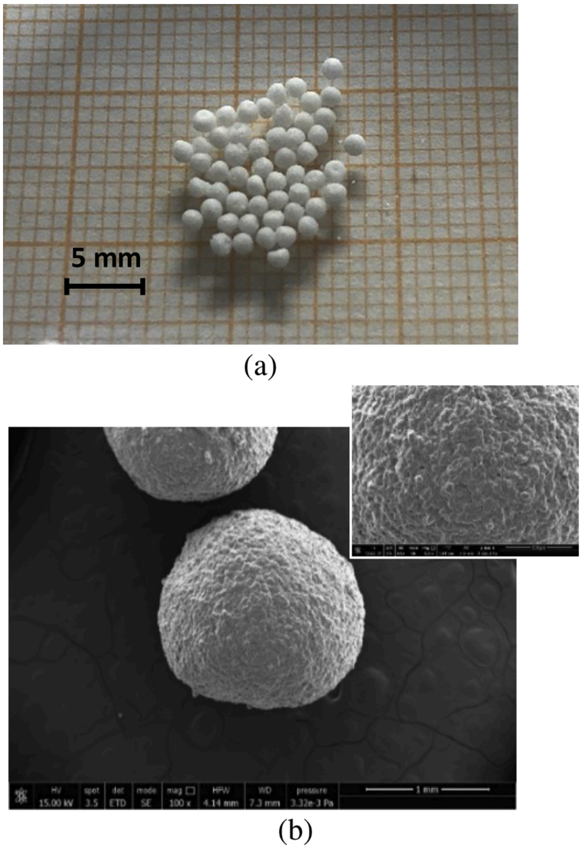
Fig. 2 shows the sintered pebbles having about 1.5 mm diameter, regular roundness, and good sphericity (aspect ratio: 0.9–1) that were subjected to compression test.

The mechanical interaction of the pebble with a wall apparatus (pebbles-breeder blanket interaction) is studied numerically by means of finite element (FE) code.

To develop simulation tools the knowledge and understanding of the produced pebbles properties become of meaningful importance to predict the pebbles bed behaviour occurring during the plant operation.

Therefore, firstly a FE analysis of the single pebble subjected to compression load at ambient temperature and atmospheric pressure, was performed, based on the elastic–plastic behaviour of a single pebble, implemented in an FEM code. Fig. 3 a shows the model of the individual pebble in contact with a wall (and bonded by adhesion forces).

The mathematical model implemented in the simulation was based on the Hertzian theory (Fig. 3b), which is capable to describe the characteristic of contact deformation during uniaxial loading. It was assumed that the pebble behaves like a soft spherical element in contact with a smooth flat surface (the flat element purple colored in Fig. 3a) [15]. Moreover, assuming a continuous material, contact theory and structure mechanics can be used to describe the mechanical behaviour of the pebble via the experimentally observed (and calculated) mechanical properties such as the Young's modulus, the yielding value, and the contact stiffness. The pebble shape is assumed fully spherical before loading with uniform inner structure. The material of the sphere is assumed to be isotropic and elastic until the first yield or failure is reached. Fig. 4 shows examples of the F-d curve from which these data are determined.



(a)

(b)

Fig. 2. a) Li_4SiO_4 pebbles (obtained after the thermal treatment at 900 °C) and SEM images of surface appearing homogeneous and quite uniform.

It is worthy to note that by varying the suspension characteristics and process parameters of pebbles fabrication it is possible to directly modify the properties of pebbles, including form, theoretical density, strength, thermal properties, etc.

In addition the numerical model implements a simplified damage model (for elastic and elastic-plastic materials): the damage growth begins approximately after an equivalent strain threshold (ε_d) experimentally defined. The coupling of the damage variable to the elastic properties of the material is obtained through the effective Young's modulus. The pebble-wall surface contact is defined through a suitable relationship of contact bodies' detection. To characterize the structural behaviour of pebble nonlinear transient analysis was performed.

3. Result and discussion

Analyzing the obtained results, we observed that the rupture occurs when the ultimate load in circumferential directions is reached.

Tensile forces, generated along the pebble circumferential perimeter, are responsible for bending effects and when yielding is reached failure begins. Under uni-axial loading, the geometry of pebble changes: the "reduced" volume of the compression path migrates to the lateral

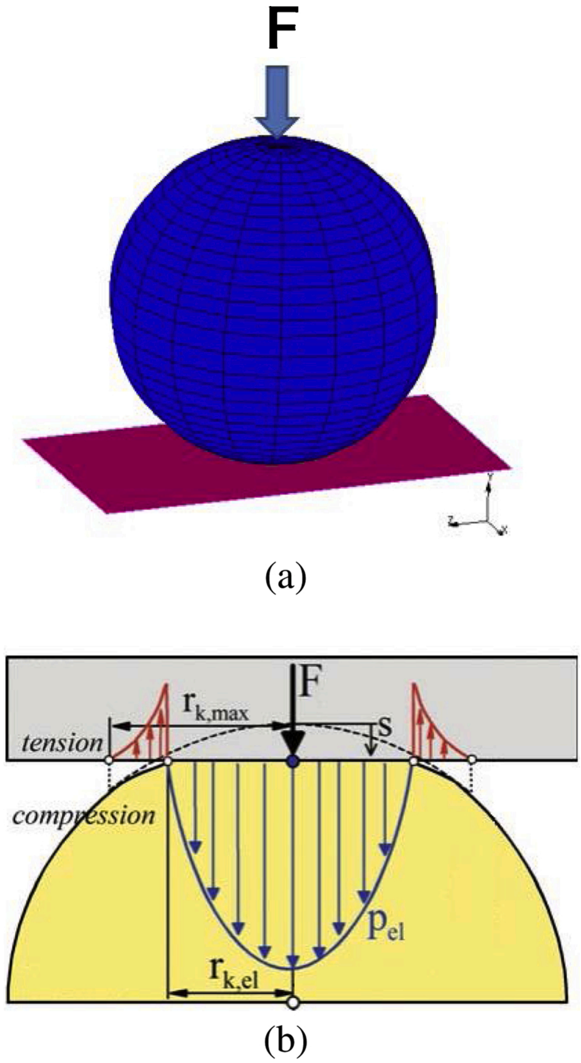


Fig. 3. a, b: a) FE model of a Li_4SiO_4 pebble; b) Characteristic pressure distribution during elastic deformation.

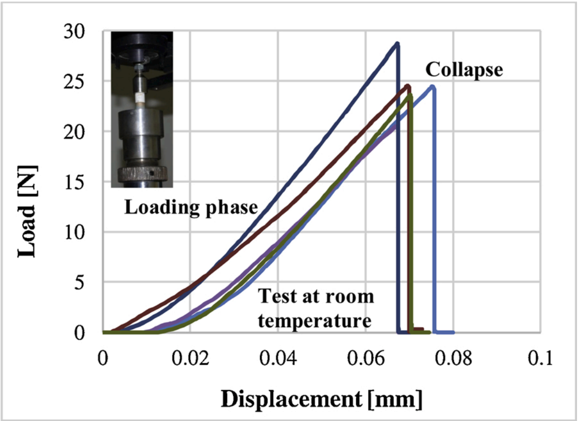


Fig. 4. Force-displacement curve for contact model. From the trend emerges that the elastic phase ends when breaking starts.

surface side of pebble, which is free to expand. As a consequence of that the radius of the totally deformed area is larger than the contact radius (i.e. 0.9 mm against 0.75 mm). The shape of the compressed particle remains axisymmetric, accordingly displacement of any axisymmetric position is identical (Fig. 5). Moreover, the contact area appeared to be deformation rate-dependent as shown in Fig. 7. When the pebble diameter decreases, the contact pressure increases, this means that for constant force the smaller pebbles behave softer than larger particles [12]. The maximum deformation was located along the vertical (y) axis at the center of the contact area where the compression force is applied (see Fig. 3a). Graphically this is represented by the narrow inward crushing, as shown in Fig. 5. Furthermore, as the actual density decreases the deformation area increases (and enlarges). The collapse occurs at an average ultimate crushing load of about 14 N.

Fig. 6 shows the comparison between the numerical and experimental displacement. The numerical displacement at contact is not represented by a linear behaviour due to the material behaviour assumptions. A trend line was so calculated in order to represent the pebble behaviour and for comparison with experimental data. The best-fit straight line (grey colored in Fig. 6) which sort of fits the numerical data has R-squared value of 0.75. The experimental data fit quite well with this trend line. Nevertheless improvement in the numerical modelling is necessary to represent better the effect of defects, as pores and cracks, on the pebble performance.

Fig. 7 shows the post-test examination performed by SEM (FEI Quanta 450 ESEM FEG with magnification range: from 6 x to 1,000,000 x). The left figure shows that the cracks that form in the contact area of the pebble propagate transversely and radially on the surface. The right figure shows the total crushed pebble at collapse.

Numerically it was not possible to simulate completely the pebble fragmentation, as experimentally observed (rupture three or four slices as shown in Fig. 7 right) due to the assumption of continuous material. Nevertheless the rupture phenomenon is controlled in the numerical simulations through the residuals method based on relative values: when simulation fails to converge to tolerance rupture is get started.

Fig. 8 shows the Von Mises stress distribution in the pebble cross section: after an initial phase characterized by some deformation not observed predominantly, the accumulation of such deformation reached at about 8 s the critical length. The cracks so propagate faster through the rest of the pebble at very high speed (of about 0.5 msec) determining the crushing.

4. Summary

The crushing load, that is an essential technical requirement for the tritium breeder pebbles, was determined performing experimental compression tests of single pebble ($\varphi = 1-1.5$ mm) in air at ambient temperature and atmospheric pressure and simulating numerically by means FE method the structural pebble performance.

In the numerical modelling it was assumed that the pebble behaves like a soft spherical element in contact with a smooth flat surface (Hertzian theory).

The maximum deformation was located along the vertical (y) axis at the center of the contact area where the compression force is applied. The contact area appeared to be deformation rate-dependent with a radius of the totally deformed area larger than the contact radius (0.9 mm against 0.75 mm).

Both the numerical displacement (0.04 mm) and pressure distribution seem to agree with experimental values (phase of compression of the sphere).

The pebbles (obtained by drip casting method [14]) exhibit an average crushing load of about 14 N.

The FE model demonstrated to be capable to reproduce the brittle pebble behavior and therefore may be used for performance assessment under different operational loads.

Additional characterizations of pebble properties (e.g. thermo-mechanical) are necessary however to provide a deeper understanding of pebbles-breeder blanket wall interaction.

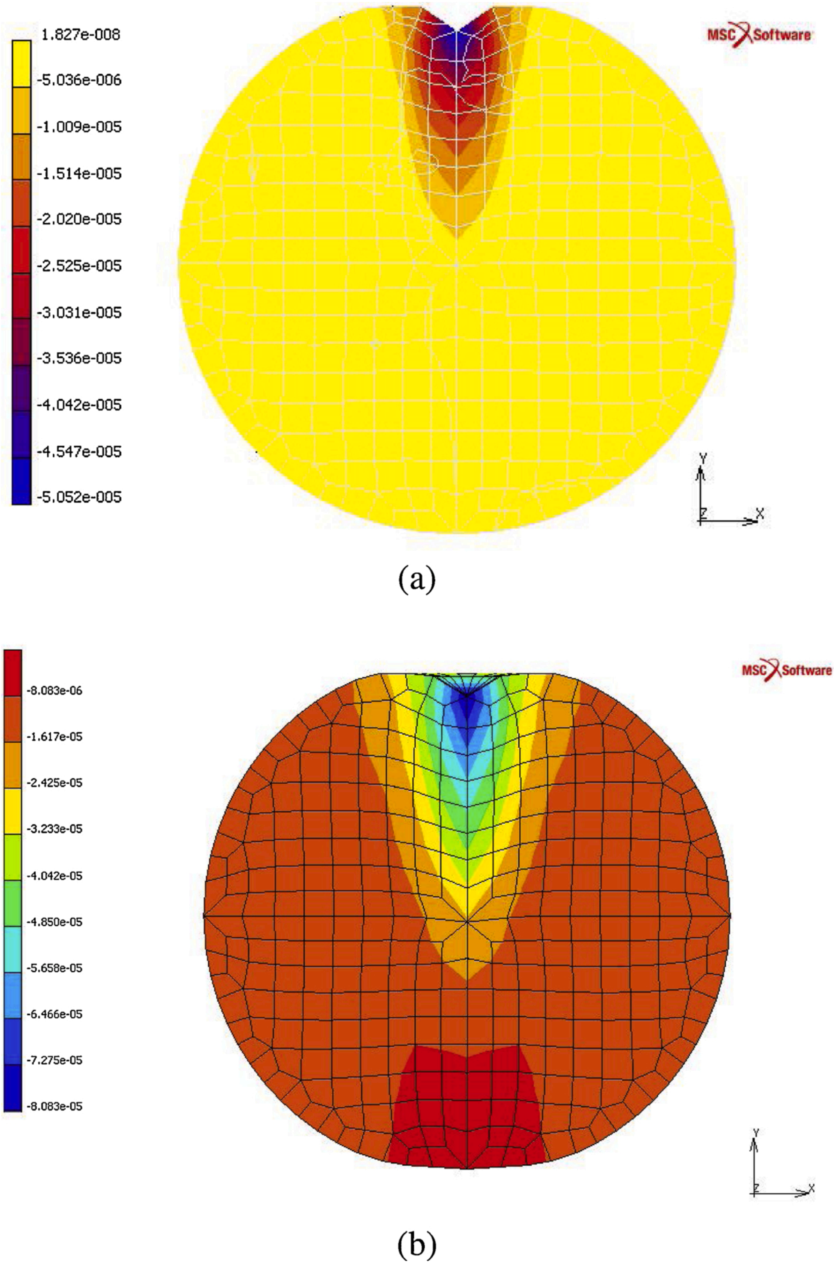


Fig. 5. a,b: Vertical displacement of pebble of about 60 % TD (a) and 40 % TD (b). In this latter case, also the reaction force contributes to the deformation.

Funding

No funding was received for this work.

Intellectual property

We confirm that we have given due consideration to the protection of intellectual property associated with this work and that there are no impediments to publication, including the timing of publication, with respect to intellectual property. In so doing we confirm that we have followed the regulations of our institutions concerning intellectual property.

Research ethics

Not applicable.

Authorship

We confirm that the manuscript has been read and approved by all named authors.

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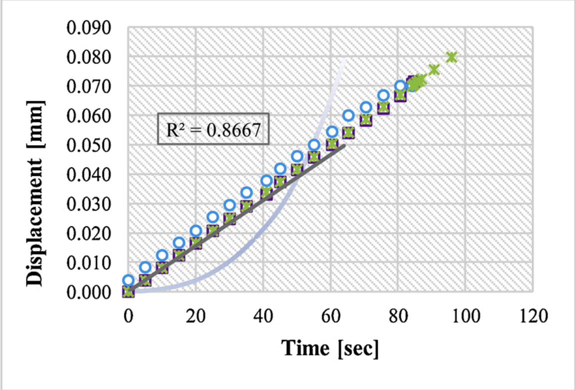

Fig. 6. Experimental vs numerical vertical displacement of pebble. The solid greys line represent the numerical data; the points indicated with circles, squares and crosses represent the experimental data. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

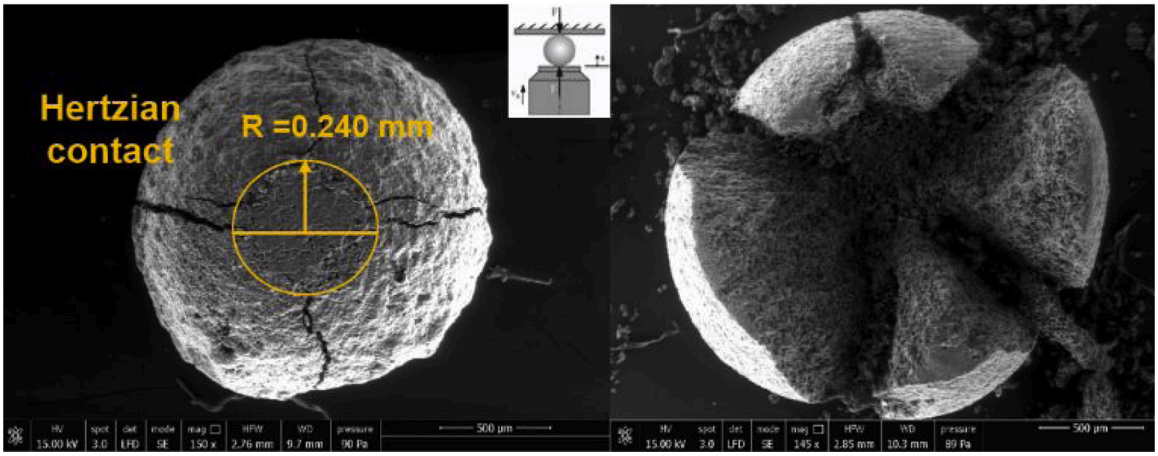

Fig. 7. Pebble fragmentation, as experimentally observed.

Fig. 8. Failure at low strain and high stress.

Declaration of Competing Interest

The authors report no declarations of interest.

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