G Model
FUSION-8452; No. of Pages 5

ARTICLE IN PRESS

Fusion Engineering and Design xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Fusion Engineering and Design

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

Pebble fabrication and tritium release properties of an advanced tritium breeder

Tsuyoshi Hoshino [a,*], Yuki Edao [b], Yoshinori Kawamura [c], Kentaro Ochiai [d]

[a] *Breeding Functional Materials Development Group, Department of Blanket Systems Research, Rokkasho Fusion Institute, Sector of Fusion Research and Development, Japan Atomic Energy Agency, 2-166 Obuch, Omotedate, Rokkasho-mura, Kamikita-gun, Aomori 039-3212, Japan*
[b] *Tritium Technology Group, Department of Blanket Systems Research, Rokkasho Fusion Institute, Sector of Fusion Research and Development, Japan Atomic Energy Agency, 2-4 Shirakata, Shirane, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan*
[c] *Blanket Technology Group, Department of Blanket Systems Research, Rokkasho Fusion Institute, Sector of Fusion Research and Development, Japan Atomic Energy Agency, 801-1 Mukoyama, Naka, Ibaraki 311-0193, Japan*
[d] *BA Project Coordination Group, Department of Fusion Power Systems Research, Rokkasho Fusion Institute, Sector of Fusion Research and Development, Japan Atomic Energy Agency, 2-166 Obuch, Omotedate, Rokkasho-mura, Kamikita-gun, Aomori 039-3212, Japan*

HIGHLIGHTS

- Li_2TiO_3 with excess Li ($Li_{2+x}TiO_{3+y}$) pebble as an advanced tritium breeders was fabricated using emulsion method.
- Grain size of $Li_{2+x}TiO_{3+y}$ pebbles was controlled to be less than 5 μm.
- $Li_{2+x}TiO_{3+y}$ pebbles exhibited good tritium release properties similar to that of Li_2TiO_3 pebbles.

ARTICLE INFO

Article history:
Received 31 August 2015
Received in revised form 6 January 2016
Accepted 12 January 2016
Available online xxx

Keywords:
Advanced tritium breeder
Pebble fabrication technique
Tritium release properties
Li_2TiO_3
Li_2TiO_3 with excess Li

ABSTRACT

Li_2TiO_3 with excess Li ($Li_{2+x}TiO_{3+y}$) has been developed as an advanced tritium breeder. With respect to the tritium release characteristics of the blanket, the optimum grain size after sintering was less than 5 μm. Therefore, an emulsion method was developed to fabricate pebbles with this target grain size. The predominant factor affecting grain growth was assumed to be the presence of binder in the gel particles; this remaining binder was hypothesized to react with the excess Li, thereby generating Li_2CO_3, which promotes grain growth. To inhibit the generation of Li_2CO_3, calcined $Li_{2+x}TiO_{3+y}$ pebbles were sintered under vacuum and subsequently under a 1% H_2–He atmosphere. The average grain size of the sintered $Li_{2+x}TiO_{3+y}$ pebbles was less than 5 μm. Furthermore, the tritium release properties of $Li_{2+x}TiO_{3+y}$ pebbles were evaluated, and deuterium–tritium (DT) neutron irradiation experiments were performed at the Fusion Neutronics Source facility in the Japan Atomic Energy Agency. To remove the tritium produced by neutron irradiation, 1% H_2–He purge gas was passed through the $Li_{2+x}TiO_{3+y}$ pebbles. The $Li_{2+x}TiO_{3+y}$ pebbles exhibited good tritium release properties, similar to those of Li_2TiO_3 pebbles. In particular, the released amount of tritiated hydrogen gas for easier tritium handling was greater than the released amount of tritiated water.

1. Introduction

The demonstration power reactor (DEMO) reactors require advanced tritium breeders which have higher stability at high temperatures. To develop these advanced tritium breeders, fabrication studies together with characterization of breeder materials have been carried out over the Broader Approach (BA) period, by preparing the necessary experimental equipment in the BA Rokkasho site.

Then, the investigation of the tritium release properties for the temperature, types of tritium purge gas and tritium breeder materials including advanced tritium breeders was conducted in the fusion neutronics source (FNS) facility of JAEA. The experimental researches give important databases for DEMO blanket design.

Lithium metatitanate (Li_2TiO_3) has been recognized as a prominent candidate material [1–3]. However, the mass of Li_2TiO_3 components has been found to decrease with time because of Li

* Corresponding author.
E-mail addresses: hoshino.tsuyoshi@jaea.go.jp, hoshino.tsuyoshi@jaea.jp (T. Hoshino).

http://dx.doi.org/10.1016/j.fusengdes.2016.01.012
0920-3796/© 2016 Elsevier B.V. All rights reserved.

evaporation under a H_2 atmosphere at high temperatures and Li burn-up [4]. To inhibit this mass decrease, Li_2TiO_3 with excess Li ($Li_{2+x}TiO_{3+y}$) has been developed as an advanced tritium breeder [5–7]. $Li_{2+x}TiO_{3+y}$ is a non-stoichiometric compound, as modeled by Kleykamp [8]. An advanced tritium breeder incorporating excess Li in single-phase $Li_{2+x}TiO_{3+y}$ is expected to be stable under the prevailing operating conditions—specifically, in a neutron environment at high temperatures.

A technique for the fabrication of $Li_{2+x}TiO_{3+y}$ pebbles using the emulsion method has been reported [9,10]. The emulsion method is one of the candidate methods for mass pebble production. The pebble diameter is well controlled in the target ranges by this method. However, the average grain size on the surface and cross section of the sintered Li_2TiO_3 with excess Li ($Li_{2+x}TiO_{3+y}$) pebbles was <5 μm and 5–10 μm. With respect to the tritium release characteristics of the blanket, the desired grain size after sintering is less than 5 μm [11]. Therefore, we need to optimize the granulation conditions to reach the target value.

Moreover, although the tritium release properties of tritium breeders are documented in databases for the DEMO blanket design, no in situ examination under fusion neutron (deuterium–tritium (DT) neutron) irradiation has been performed. In this study, a preliminary examination of the tritium release properties of advanced tritium breeders was conducted.

In the past, DT neutronics experiments have been conducted using assemblies that simulate the Li_2TiO_3 blanket at the Fusion Neutronics Source (FNS) facility of JAEA to investigate the accuracy of the predicted tritium production rate [12–16]. On the basis of the experiments, the accuracy of the predicted tritium production rate for the Li_2TiO_3 blanket was determined to be within 10%. Some DT neutronics experiments to test the accuracy of the predicted tritium production rate have been performed with lithium ceramics in various countries; however, no investigation concerning the tritium release properties of the Li_2TiO_3 blanket has been conducted with DT neutrons. Experiments concerning the tritium release properties of the Li_2TiO_3 blanket are therefore an urgent technical issue.

For the investigation of the tritium release properties of $Li_{2+x}TiO_{3+y}$ breeder pebbles, research and development using DT neutron irradiation experiments were started as new tasks of the DEMO R&D component of the IFERC Project in 2013. Investigations of the tritium release properties with respect to the temperature, type of tritium purge gas, and the tritium breeder material, including advanced tritium breeders, are needed because experimental research provides important data for the design of DEMO blankets and tritium plant systems.

In this study, an examination of pebble fabrication and the tritium release properties of advanced tritium breeders was performed.

2. Experimental

2.1. Pebble fabrication by emulsion method

In this application of the emulsion method, gel particles were produced from a single-phase $Li_{2+x}TiO_{3+y}$ slurry and oil in a granulator. As such, ensuring that the slurry contains single-phase $Li_{2+x}TiO_{3+y}$ is important. During solution-phase synthesis using Li and Ti alcohol solutions, the excess Li was observed to be incorporated into Li_4TiO_4, which is chemically less stable than Li_2TiO_3.

Consequently, the processing steps of a new solid-state reaction using the raw material powders of $LiOH{\cdot}H_2O$ and H_2TiO_3 were developed [17]. During a mixing of the powders, the mixtures gradually transformed into a gel by solid-state reaction. After being dried, the gel was calcined at 773 K for 5 h in air and sintered at 1373 K for 5 h under N_2. The sintered $Li_{2+x}TiO_{3+y}$ powder was then used as the raw material in the emulsion method.

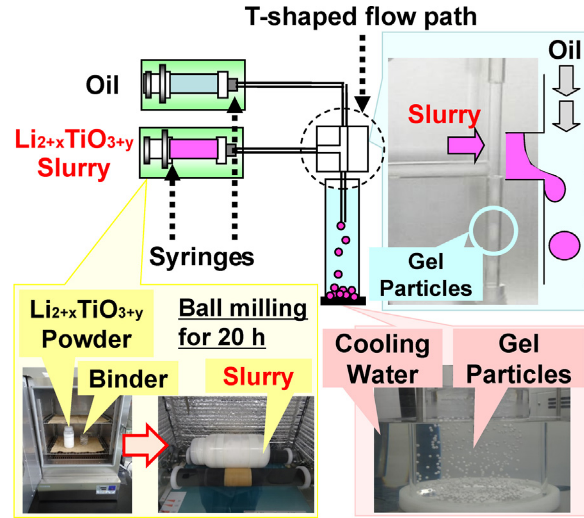

Fig. 1. Illustrations of $Li_{2+x}TiO_{3+y}$ pebble fabrication by the emulsion method.

The emulsion method is illustrated in Fig. 1. One syringe was filled with oil and the other with the $Li_{2+x}TiO_{3+y}$ slurry. The $Li_{2+x}TiO_{3+y}$ slurry was prepared from $Li_{2+x}TiO_{3+y}$ powder, binder (agar), and water. These mixtures were mixed in a ball mill at 323 K for 20 h. The $Li_{2+x}TiO_{3+y}$ slurry flow was cut by the oil flow in T-shaped flow path, and the $Li_{2+x}TiO_{3+y}$ gel particles were deposited in an oil-filled container. After granulation, the gel particles were cooled down in the container around a 283-K cooling water pool for 3 h. The obtained gel particles were subsequently calcined in air at 873 K for 5 h and sintered under several sets of conditions in an effort to control the grain size.

2.2. DT neutron irradiation

Beryllium blocks were used to enhance the tritium production rate at the region of $Li_{2+x}TiO_{3+y}$ pebbles in the container (Fig. 2). The DT neutron source of the JAEA-FNS target can generate neutrons at a rate on the order of 10^{11} n/s. The tritium production rate of the DT neutron irradiation was calculated using a Monte Carlo code (MCNP).

$Li_{2+x}TiO_{3+y}$ pebbles were placed inside a stainless steel irradiation container (Fig. 3). The temperature of the tritium breeder was maintained at 573 K, 723 K, or 873 K using a wire heater. To remove the tritium produced by neutron irradiation, 1% H_2–He purge gas was passed from the inlet to the outlet through the $Li_{2+x}TiO_{3+y}$ pebbles. The purge gas was passed through the pebble area in the irradiation container at a flow rate of 100 cm^3/min and the gas carried the released tritium to the water bubblers to allow the amount of tritium released from the $Li_{2+x}TiO_{3+y}$ pebbles to be measured.

The tritium gas consisted of tritiated hydrogen (HT) and tritiated water (HTO) gases. The HTO gas was collected in the first water bubbler. The HT gas was then oxidized to HTO using an oxidation reactor, and this HTO was collected in the second water bubbler. Our previous data obtained with the system combined a CuO bed and water bubbler indicated a questionable result HT was continuously released for a while even after neutron irradiation stopped [18,19]. We have determined that the delayed HT release after irradiation stopped owing to the hydrophilic property of CuO. The mass transfer resistance for the desorption of tritiated vapor from CuO contributed significantly to the overall tritium release behavior even though the CuO was heated up to 773 K.