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Rapid preparation of tritium breeder material Li_2TiO_3 pebbles by thermal plasma

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ABSTRACT

A route to obtain tritium breeder material Li_2TiO_3 pebbles in one step was investigated using a direct-current (DC) thermal plasma system. For this purpose, irregular and agglomerate powders were used as raw powders which were injected into DC thermal plasma torch with a plasma power level of ~18 kW. The morphology, particles size distribution and phase change of the prepared Li_2TiO_3 pebbles were characterized by field-emission scanning electron microscope and X-ray diffraction (XRD). The results showed that prepared Li_2TiO_3 pebbles presented excellent spherical degree with average size about 100 μm and the spheroidization rate was close to 100%. The XRD patterns demonstrated that higher degree of crystallinity of Li_2TiO_3 pebbles could be obtained by thermal plasma processed. Formation mechanism of Li_2TiO_3 pebbles in DC thermal plasma system was discussed.

1. Introduction

As a key Li-ceramics material, Li_2TiO_3 has been considered as candidate for tritium breeder materials in **ITER** test blanket module, because of its reasonable lithium atom density, low activation, excellent chemical stability, good compatibility with structural materials and good tritium release characteristics at low temperature [1–4]. This tritium breeder material can be solidified into column, circular or spherical shape. Among these, spherical Li_2TiO_3 are considered as the preferred materials due to the large specific surface, more channels between the pebbles, good permeability which is conducive to the diffusion and release of tritium.

Some methods for the preparation of Li_2TiO_3 pebbles have been proposed in the past two decades, such as extrusion-spheronisation-sintering [5], sol-gel process [6], wet process [7,8], and melting granulation method [9]. Although these methods have been adopted to prepare Li_2TiO_3 pebbles with good degree of sphericity and higher yields, there are still some difficulties during the process of preparation, such as long reaction time, complex procedure, tough conditions, even bringing to pollution, such as waste water and toxic gases.

In recent years, some people reported the preparation method of spherical powder by thermal plasma, including a variety of refractory metallic and ceramic powder, such as nickel [10], tungsten [11], alumina [12], silicon carbide [13] and so forth. This preparation method of spherical powder is based on high processing temperature in plasma core area (up to 1.0×10^4 K), and fast quenching rate ($\sim 10^{5-6}$ Ks^{-1}) at the plasma tail [14,15]. Any powders which are injected into the thermal plasma are heated up and melted down immediately, and then become spheres due to surface tension; the melted droplets solidify into spherical particles due to rapidly quenching when they spurt out of the plasma. The sphericity and flowability, and mechanical properties of prepared spherical powder have been significantly improved. In addition, the highlight advantage of this method is one-step-preparation without any additional pollution in the preparation process. In particular, the preparation of tritium breeder material Li_2TiO_3 pebbles by thermal plasma has not been reported.

Thermal plasma can be generated by various methods of discharges, i.e. radio frequency (RF) inductively coupled discharge, and DC arc discharge. Although RF inductively coupled thermal plasma has advantages, such as absence of electrode pollution, lower velocity resulting in long residence time of powder in plasma, the flow instability and lower electric-thermal conversion efficiency limit it to be widely used. DC thermal plasma has excellent flow stability and higher electric-thermal conversion efficiency which can reduce the preparation cost effectively.

In this work, the application of nitrogen plasma for preparation Li_2TiO_3 pebbles is presented firstly. The ultra-fine and

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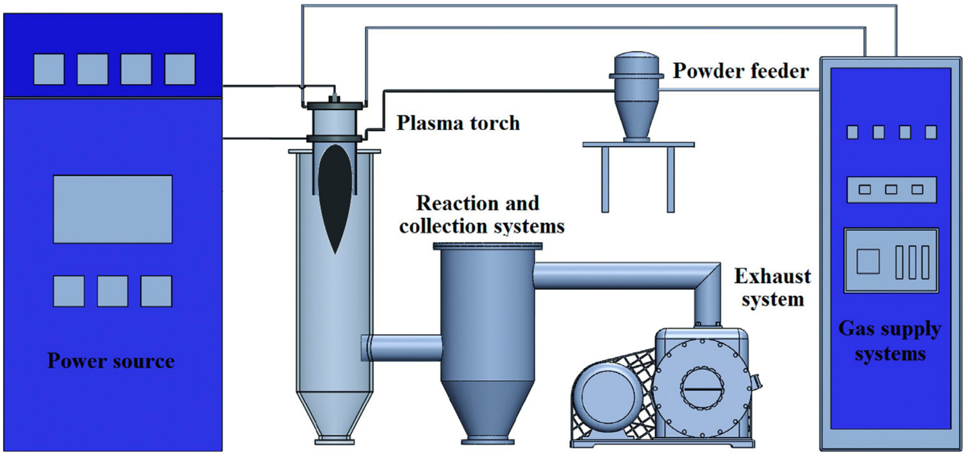

Fig. 1. Schematic illustration of the experimental setup.

agglomerated Li_2TiO_3 powder as raw materials is introduced into the reactor and mixed with thermal plasma jet. The morphology and crystallography of prepared Li_2TiO_3 pebbles are characterized. The forming mechanism of Li_2TiO_3 pebbles is discussed in the end.

2. Experimental setup and methodology

The experiment aiming at preparing Li_2TiO_3 pebbles was performed by DC thermal plasma operating with a homemade plasma torch. The plasma torch was energized by a DC power supply with a maximum output capacity of 50 kW, and nitrogen was used as the plasma gas. The plasma working gas introduced between the anode and trigger electrode is used to produce a stable plasma, the protective nitrogen gas injected between cathode and trigger electrode is used as a shroud gas to protect the cerium-tungsten cathode, and the role of the carrier gas is to transport the raw powder into the plasma torch. The raw powder was injected into the plasma torch using a homemade rotary powder feeder with the nitrogen carrier gas. A tubular reaction chamber with cooling water outside was closely connected to the plasma torch. Followed the reaction chamber, there is a powder collection chamber connected with a two-stage water-ring vacuum pump which was used to maintain a negative pressure. The gas supply systems were used continuously to provide nitrogen for plasma and powder feeder. A schematic description of the experimental setup is shown in Fig. 1.

The experiment is conducted as follows. Firstly, a small amount of nitrogen gas was inject into the plasma torch in order to ignite the plasma by applying a high-frequency voltage, then increase the input power and gas flow rate to maintain stable plasma. At this time, the plasma temperature is higher than the melting temperature of Li_2TiO_3 material and the length of plasma flow is about 240 mm. Next, the raw powder was delivered to high temperature zone of plasma through the feeding mouth located between the anode and trigger electrode using the carrier gas. The injected powder will be heated up, melted down and even a small part of particles were vaporized when they flew into the high temperature region of the plasma torch, and the melted droplets would be quenched and solidified into spherical pebbles under the effect of surface tension when they spurt out of the plasma region. Finally, prepared Li_2TiO_3 pebbles were collected from the bottom of the reaction and collection chamber. During the operation, the plasma power was 18 kW, calculated from the product of DC arc voltage (180 V) and the arc electric current (100 A). The plasma working gas, protective gas and carrier gas was supplied at the flow rate of 1.2, 3.0, and 0.6 m^3/h, respectively. The flow rate of the raw powder was controlled at 10.0 g/min.

Morphology and phase of the prepared samples was characterized by field-emission scanning electron microscope (FE-SEM, Model S-4800, Hitachi, Tokyo, Japan) and X-ray diffraction (XRD, X'pert Pro MPD, Philips, Netherlands), respectively. The size distribution of the Li_2TiO_3 pebbles was subsequently determined from the FE-SEM micrographs on an image analyzer, with 350 pebbles counted in each batch.

The d50 value for each batch of Li_2TiO_3 pebbles, that is the pebble size at the cumulative fraction of 50%, was used to denote the average pebble size, which was proposed by J-G Li [16].

3. Results and discussion

3.1. Morphology

The FE-SEM in Fig. 2(a) shows the morphology of raw Li_2TiO_3 powders at different magnifications in this work. It is clearly seen that the raw powders are ultra-fine particles without any liquidity. In magnification image Fig. 2(b), it can be seen that the raw powder are very fine, irregular and easy to agglomerate. After processed by plasma, spherical Li_2TiO_3 pebbles are formed and the FE-SEM image is shown in Fig. 3(a). It demonstrates that the prepared Li_2TiO_3 pebbles present excellent spherical degree and spheroidization rate close to 100%, which is better than that achieved using the wet chemistry method [7]. This indicated that the raw Li_2TiO_3 powders are melted completely through absorbed plasma energy at the feed rate of 10 g/min. The experiment found that at increased feed rate, some particles will not be sufficiently melted to become spheroidized and remain irregularly shaped. From magnification of ×1000 of Fig. 3(b), it can be seen that the Li_2TiO_3 pebbles are composed of many small particles. Under the action of plasma flow, melted or partly melted droplets collide with each other, some of them even bond together, and then grow into larger droplets; finally the droplets are solidified and stuck to a larger particles. The same phenomenon was found in spheroidization of alumina [17,18].

Fig. 4 presents the energy dispersive spectrometer (EDS) spectrum of raw and prepared Li_2TiO_3 pebbles. It shows that the powders almost exclusively composed of titanium and oxygen which the atomic ratio is close to 1:3, and there are no significant changes in weight and atomicity before and after plasma treatment. However, the EDS of the matrix could not show the presence of lithium because of its light weight. In addition, a traces of Al impurity were found in the EDS analysis of Li_2TiO_3 pebbles. As shown in Fig 4(a), this Al impurity should be mainly introduced by