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Functional Oxide Thin Film Fabrication by Flash Light Irradiation for Solid State Energy Devices

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Abstract

Here we report a photonic annealing process for metal oxide electrolyte/electrode thin film for solid state energy devices. Ceramic films prepared by precursor solution based simple wet-chemical method and nanoscale powder based screen printing method are directly heat-treated within very short process time by the flash light irradiation system. Under standard pressure and temperature conditions, the residual organics in films were almost completely decomposed in the pre-annealing step, and the crystalline phase and good ionic, electronic conductivities were developed during the main annealing step. These films showed material properties comparable to those of thermally annealed films, and therefore, a long process time of several tens of hours in conventional thermal sintering can be significantly reduced within tens of seconds. The significance of this work includes the novel heat treatment methodology for various solid-state functional materials and the demonstration of solid-state energy devices such as solid oxide fuel cells and all-solid-state batteries.

1. Introduction

With the increasing global energy issues, solid-state energy conversion and storage devices such as all-solid-secondary batteries, photovoltaics and fuel cells have attracted many attentions because of their high reliability, efficiency, safety and flexibility of the systems[1,2]. Generally, the ceramic components in these systems are mainly produced by non-vacuum powder and solution based process to ensure high productivity. However, a high-temperature post-heat treatment process is usually required to achieve desired material properties. This long and high temperature sintering process significantly increases entire processing time and production costs, which impedes commercialization of these systems. Therefore, in order to reduce the manufacturing cost of the systems, it is essential to improve the fabrication process especially the high temperature sintering process.

Studies attempting to overcome this problem have been actively conducted to the development of alternative sintering methods such as laser sintering, microwave sintering and spark plasma sintering. However, the existing alternative sintering methods are problematic in terms of the availability, high system cost and low scalability[3]. For example, spark plasma sintering is difficult to use in practice owing to its non-uniformity and the constraints it imposes on the shape of the sample. In addition, although the sinterbility of laser sintering has recently been confirmed, it remains problematic with a narrow spot size and scalability that hinder its use in practical applications.

Therefore, in this study, the flash light irradiation process was adopted to fabricate functional oxide thin film within very short process time. The microstructure, film composition, crystallinity and electric/ionic conductivity were analyzed and compared with conventionally annealed functional oxide films. The results of this study will provide significant impact for the fabrication of functional oxide films and therefore, expedite commercialization of solid-state energy devices.

2. Experimental

To obtain oxide electrolyte/electrode thin films, metal organic precursor solution was synthesized by using metal acetates precursor with complexing and polymerization agent. The synthesized precursor solution was spin coated to deposit thin films. The detailed fabrication method is described in our previous reports[4].

The flash light irradiation process was conducted with a system consisting of a power supply, xenon lamp, a reflector, pulse controller and bottom heater. The controllable variables in the flash light irradiation process are the input voltage, irradiation time, pulse numbers, pulse duration and substrate temperature. The detailed system configuration is described in the following section.

3. Results and Discussion

The flash light irradiation system is composed of a xenon flash lamp, reflector, beam guide, power supply and controller. Figure 1(a) shows a schematic diagram of the flash light irradiation systems. The generated flash light from the arc plasma discharge has a broad wavelength range (380-980 nm) as shown in Fig. 1(b). In this study, the flash light irradiation process was divided into two steps; pre-annealing and a main-annealing step. The pre-annealing step involved total 30

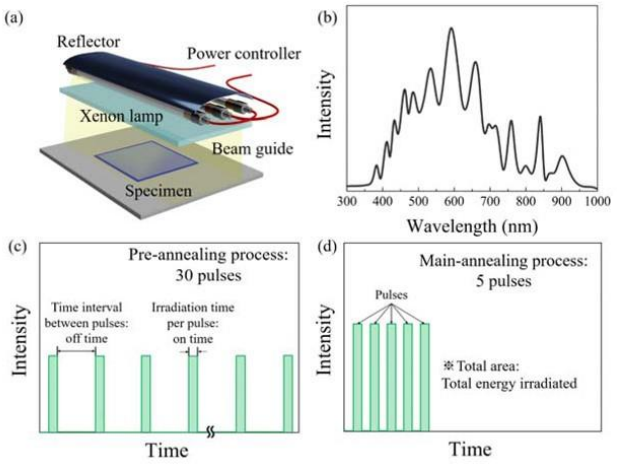

Fig. 1 (a) Schematics of the flash light irradiation systems. (b) spectrum of the flash light irradiation pulse from the xenon lamp. pulse characteristics for the (c) pre-annealing step and (d) main-annealing step.

pulses with relatively long pulse duration and low applied voltage (on-time: 10ms, off-time: 100ms) to completely remove the residual organics. On the other hand, the main-annealing step composed of 5 pulses (on-time: 10ms, off-time: 10ms) with high applied voltage and short pulse duration to sufficiently increase surface temperature for crystallization. The schematics of the applied pulse conditions are described in Fig. 1(c) and (d).

The residual organic in the as-deposited films should be completely removed during annealing process because the residues can cause secondary contamination and deteriorate the film properties. From the FT-IR results shown in Fig. 2(a), the as-deposited sample clearly shows the existence of organic functional groups, such as carboxylic acids at the wavenumber range of 1100-1800 cm^{-1}. On the contrary, it is confirmed that no trace of characteristic organic bands was observed in the samples thermally and pre-annealed by flash light irradiation. The irradiated light absorbed by the film sufficiently increases surface temperature to decompose remnant organics. The X-ray diffraction (XRD) patterns of yttria-stabilized zirconia(YSZ) solid oxide electrolyte films shown in Fig. 2(b). The result clearly shows that the crystallinity of the flash light sintered sample with main annealing condition ($80J/cm^2$) has similar crystalline development comparable to those of the thermally annealed samples (800 and 900°C). This result revealed that the energy required for the crystallinity development from the amorphous phase of as-deposited films was sufficiently supplied by the flash light irradiation process.

The ionic conductivity of the YSZ thin films sintered by thermal and flash light annealing were measured and shown in Figure 3. The ionic conductivity of the flash light sintered films showed similar ionic conductivity with the thermal sintered films due to the sufficient crystalline development. Although this flash light irradiation process has extremely short processing time, the sintered YSZ electrolyte films can achieve comparable material properties to conventionally annealed samples.

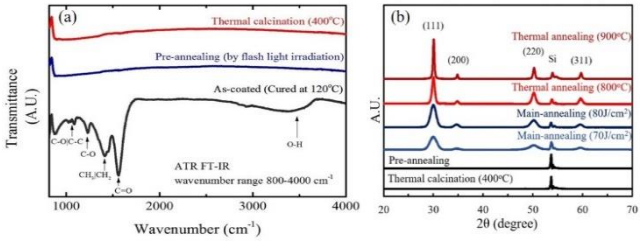

Fig. 2 (a) FT-IR spectra of as-deposited, flash irradiated (pre-annealing) and thermally calcined samples. (b) X-ray diffraction patterns of YSZ thin film; thermal annealed, flash light irradiated (main-annealing).

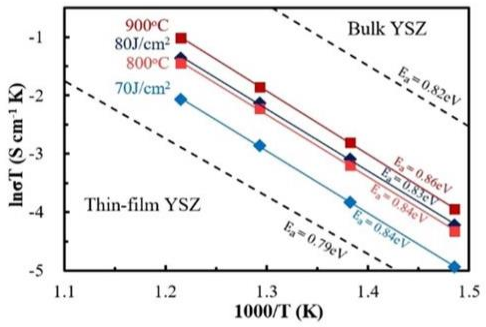

Fig. 3 Arrhenius plot of ionic conductivity of flash irradiated (main-annealing) and thermally sintered YSZ thin films.

4. Conclusions

In this study, the flash light irradiation process was demonstrated to fabricate functional oxide electrolyte/electrode thin film within very short process time. The flash light sintered films showed material properties comparable to those of thermally annealed films and therefore a long process time can be significantly reduced to a few seconds. The results of this study will provide significant impact for the fabrication of functional oxide films and therefore, expedite commercialization of solid-state energy devices.

Acknowledgements

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