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Original Research Paper

Preparation of indium oxide powders by microwave plasma dehydration of indium hydroxide powders

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ABSTRACT

Indium oxide was prepared from the dehydration of indium hydroxide using atmospheric-pressure microwave air plasma. Compared with the conventional thermal plasma processing that was performed with the vapor phase reaction, the solid-state reaction was attempted in this study because microwave plasma has an intermediate temperature that is comparable to the melting temperature of inorganic materials and between those of the electric furnace and the thermal plasma. The results were compared with those with the electric furnace and the thermal plasma. With both the microwave plasma and the electric furnace, the macro-morphologies of the raw material were maintained, which indicates successful dehydration. However, the micro-morphologies differed. The product of the microwave plasma had a smooth surface, whereas the product of the electric furnace had a cracked and rough surface. The cracks were regarded as the results of the poor diffusion of the dissociated water. In the microwave plasma, the high temperature and the fast heating rate enhanced the diffusion and controlled the formation of cracks. With the application of the thermal plasma, the nanoparticles were prepared due to the vaporization of the dehydrated material. Thus, the microwave plasma is considered applicable to the solid-state reaction accompanying degassing, without a change in the microstructure of the raw material.

1. Introduction

In recent decades, in-flight material preparation processes have attracted much interest because of their advantages of continuity and a short reaction time [1]. Most of these processes have focused on vapor-phase synthesis using an energy source such as flame or plasma [2–5]. Vapor-phase synthesis is appropriate for mass production, but the product is limited to submicron particles. Therefore, research on other types of reaction to obtain various products is required. As an example of such, the in-flight solid-state reaction with microwave plasma is suggested in this paper, and its performance is compared with those of the electric furnace and the thermal plasma. Microwave related techniques including microwave plasma have been widely used for material processing [6–8], its application to the in-flight solid-state reaction is not yet attempted.

Microwave plasma is strictly classified as non-equilibrium plasma. However, when it is discharged at atmospheric pressure, it comes close to the local thermodynamic equilibrium (LTE), and its gas temperature approaches some thousands of Kelvin due to the increase in the energy transfer from the electron to a heavy species [9]. By applying microwave plasma to the in-flight solid-state reaction, the reaction mechanism and the properties of the product are expected to be affected by the microwave plasma with the higher temperature and faster heating rate ($\sim 10^6$ K/s) than by the electric furnace, which is a conventional heat source for solid-state reaction. Although thermal plasma, which is the plasma that is conventionally used in the in-flight material process, also has a fast heating rate, the solid-state reaction is expected to be disturbed with it due to its high temperature of over 10,000 K in the hottest region.

As a reaction model, the dehydration of indium hydroxide [$2In(OH)_3 \rightarrow In_2O_3 + 3H_2O$], which is a favorable method of obtaining In_2O_3 [10–12], was selected because of some of its advantages. First, $In(OH)_3$ can be simply synthesized with a desirable morphology. Second, the macro-morphology of the as-prepared $In(OH)_3$ is maintained during its dehydration to convert it into In_2O_3. As a result, morphology-controlled In_2O_3 can be obtained. Similar results have been reported for other hydrates [13–15]. This process generally includes not only dehydration but also crystallization of the product. Therefore, the actual process is performed at a higher temperature for some hours in an electric furnace to obtain a high-crystalline product, although the dehydration of $In(OH)_3$ starts at nearly 480 K.

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Indium oxide (In_2O_3) is an n-type semiconductor with band-gap energy of 3.6 eV. It is optically transparent and has a high electrical conductivity by doping its impurities [16]. Tin-doped indium oxide (ITO) is the most widely used transparent conductive oxide (TCO) in the display field [17]. As In_2O_3 is the base material of TCO, techniques related to In_2O_3 have attracted much attention.

In the authors' previous study [18], ITO nanoparticles were prepared with thermal plasma, which has a maximum temperature of over 10,000 K. When $In(OH)_3$ particles that were as big as a few micrometers passed through the thermal plasma jet, they formed nanoparticles through the vapor-phase reaction.

In this study, $In(OH)_3$ was dehydrated with microwave plasma. The microwave plasma was applied as the energy source to replace the electric furnace, with the expectation that the $In(OH)_3$ particles can be dehydrated by passing through the microwave plasma jet for some milliseconds. This study was conducted to investigate the possibility of performing the in-flight solid-state reaction with microwave plasma and to reveal the differences in the products and the reaction mechanism in the microwave plasma and those in the electric furnace and the thermal plasma.

Table 1
List of the experimental conditions.

Classification	Sample name	Experimental conditions		
MWPs	MWP750 MWP1000 MWP1250	Microwave plasma	Power	750 W 1000 W 1250 W
EFs	EF773 EF1023 EF1273	Electrical furnace	Temperature	773 K 1023 K 1273 K
TPs	TP6 TP9	Thermal plasma	Power	6 kW 9 kW

2. Experiments

As mentioned, In_2O_3 was prepared with three different energy sources: microwave plasma, electric furnace and thermal plasma. In this study, the products were called by their experiment conditions, which are listed in Table 1.

2.1. Preparation of In_2O_3 using microwave plasma

The experiment setup is shown in Fig. 1. First, argon was tangentially introduced in a quartz tube (I.D.: 9 mm). Then microwave power (2.45 GHz, maximum power 1.3 kW, IDX Company, Ltd.) was coupled with argon to generate plasma in the quartz tube. After the plasma generation, the argon was entirely replaced with air to produce pure air plasma.

$In(OH)_3$ as the raw material was fed to the plasma jet in the direction coaxial to that of the air carrier gas, using a powder feeder (type: MF, Technoserve Co. Ltd.). The fed particles passed through the quartz tube and then were collected in a polytetrafluoroethylene (PTFE) filter with a pore size of 1 μm. In addition, cooling gas was introduced between the quartz tube and the filter to keep the PTFE filter from burning. The pressure was kept close to the atmospheric pressure. The detailed experiment conditions for the microwave plasma are summarized in Table 2.

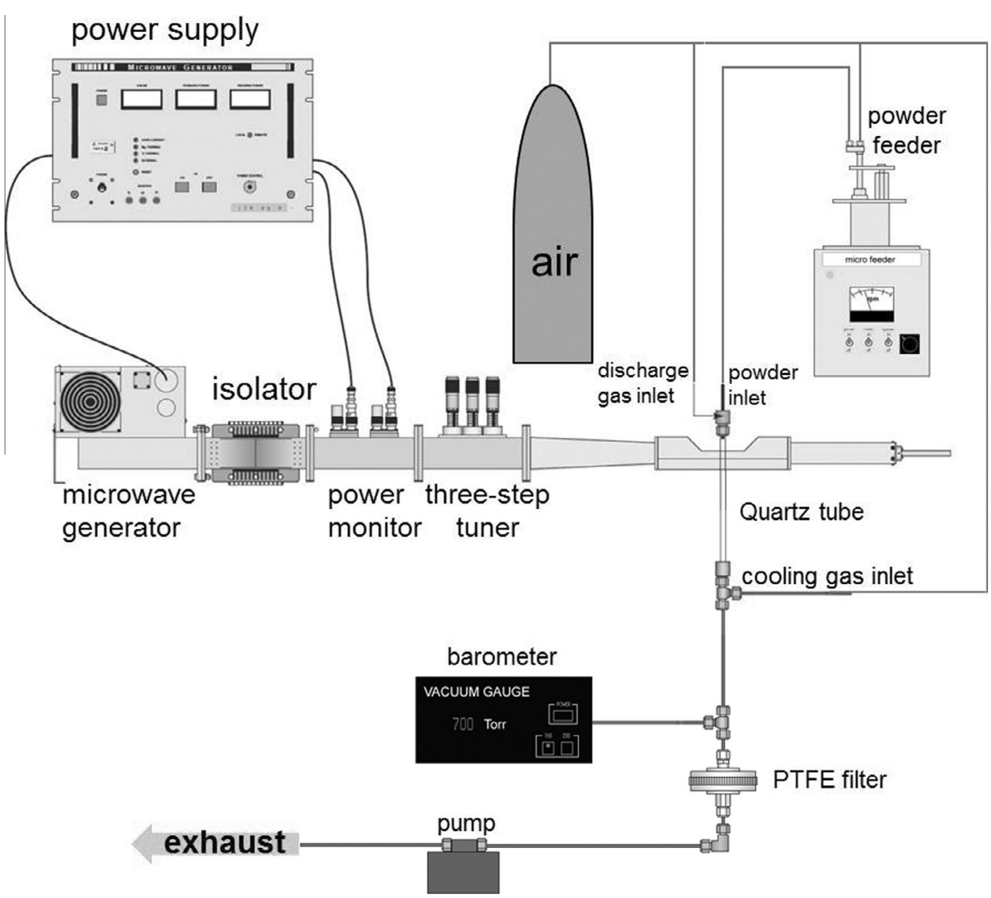

Fig. 1. Experimental set-up of the microwave plasma reactor.