23P010-O

Effect of Intermittent Supply of the Raw Material and Quenching Gas during Nanopowder Synthesis using Pulse Modulated Induction Thermal Plasmas

PMITPを用いたナノ粒子生成における原料・クエンチングガスの間歇投入効果

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This paper describes the effect of intermittent feeding of the raw material and quenching gas using the pulse modulated induction thermal plasma (PMITP), during TiO_2 nanopowder synthesis. The fast solenoid valves were adopted to control quenching gas flow and raw material feeding intermittently being synchronized with the pulse modulation. Synthesized nanopowder was analyzed by SEM and XRD. The mean particle diameter of synthesized nanoparticles were compared with/without the pulse modulation, intermittent feeding of raw materials and intermittent supply of quenching gas.

1. Introduction

Titanium oxide (TiO_2) nanoparticles in anatase phase are continuously receiving attention for use as photocatalysts, photonic crystals, photovoltaic cells, gas sensors, and a strong deoxidation material. It is also used for producing hydrogen gas from water for fuel cells. The authors have investigated a pulse modulated induction thermal plasmas (PMITP) system to be used for TiO_2 nanoparticles [1,2]. The PMITP are established by the modulated coil current, which perturbs the thermal plasma temperature. From those results we found that adoption of the pulse modulation provided smaller nanoparticles because evaporated material was rapidly cooled [1,2].

In the present work, the fast solenoid valves were used to control intermittent feeding of both quenching gas flow and raw material. The intermittent feeding was synchronized with the pulse modulation. Using this system, nanoparticle synthesis was made to study intermittent feeding effects.

2. Experimental setup

Fig.1 shows a schematic diagram of experimental setup for nanopowder synthesis. This setup has four parts: a plasma torch, an upstream chamber, a downstream chamber, and a collection filter. Inside the plasma torch, an inductively coupled thermal plasma is formed. The coil current sustaining the thermal plasma can be modulated into a rectangular waveform. $Ar\text{-}O_2$ gas was supplied as a sheath gas along the inside wall of the torch. Ar/Ti powder was injected through a water-cooled tube with Ar carrier gas from the top of the torch. The Ar quenching gas was also supplied in radial direction from 8 holes located downstream of the torch. These Ar/Ti powder and Ar quenching gas can be intermittently supplied by switching fast response solenoid valves installed, which was synchronized to the coil current modulation for sustaining thermal plasmas. The experiment condition is shown in Table 1. Note that the total Ar quenching gas flow rate was 40 slpm for intermittent supply of quenching gas, while it was lower than 50 slpm for continuous supply of quenching gas. Synthesized particles were collected from the filter.

3. Experimental results

Fig.2 presents examples of FE-SEM micrographs of particles five conditions: (a) non-modulation, continuous feeding of raw material and continuous feeding of quenching gas (NM/CWFM/CWFQ), (b) non-modulation, intermittent feeding of raw material, and continuous feeding of

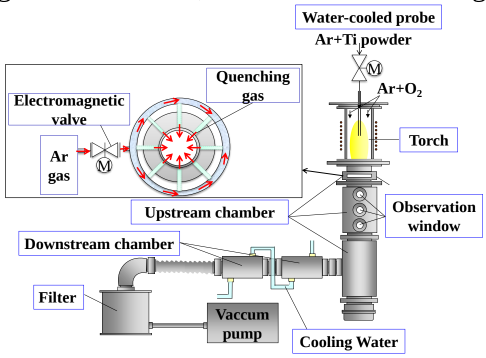


Fig.1. Experimental setup for nanopowder synthesis.

quenching gas (NM/IMFM/CWFQ), (c) non-modulation, continuous feeding of raw material, and intermittent feeding of quenching gas (NM/CWFM/IMFQ), (d) pulse modulation, continuous feeding of raw material, and continuous feeding of quenching gas (PM/CWFM/CWFQ), (e) pulse modulation, continuous feeding of raw material, and intermittent feeding of quenching gas(PM/CWFM/IMFQ). For any condition, nanoparticles were confirmed to be synthesized. Using the micrographs, the particle size distribution and then the mean particle diameter were obtained from observation of 200 randomly sampled particles. The XRD analysis was also made for the synthesized particles, indicating they contained 85%-90% anatase TiO_2 for any condition.

3.1 *Effect of pulse modulation*

Fig.3 shows the estimated mean diameter of nanoparticles synthesized under different five conditions. The condition NM/CWFM/CWFQ corresponds to the conventional one with continuous feeding of gas and raw material. We have had results that use of the pulse modulation with no quenching gas decreased the mean particle diameter around 50-65 nm [1,2]. In that case, rapid cooling of evaporated material might occur mainly in off-time of the pulse modulation. On the other hand, use of quenching gas in the present work provides much smaller particles around 30-40 nm as shown in Fig.3. In this case, the pulse modulation offers a larger mean particle diameter by comparing results NW/CWFM/CWFQ and PM/CWFM/CWFQ. This may be because the pulse modulation causes a large disturbance in the plasma, which may decrease the temperature gradient strongly formed by quenching gas.

3.2 *Effect of intermittent supply of raw material*

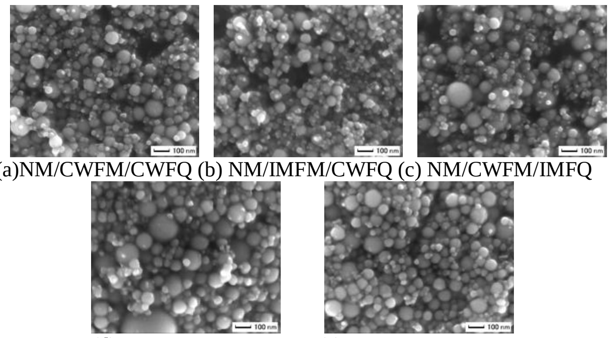
Effect of intermittent supply of raw material can be seen by comparison between the results of NM/CWFM/CWFQ and NW/IMFM/CWFQ in Fig.3. The mean diameter of nanoparticles produced with intermittent feeding of raw material is a little smaller than that with its continuous feeding. This may be because the raw material was evaporated or cooled efficiently in the intermittent feeding. It will be possible to produce smaller nanoparticles and to improve the production efficiency by adjusting timing of the intermittent feeding of raw materials [2].

3.3 *Effect of intermittent supply of quenching gas*

We compared results between NM/CWFM/ CWFQ and NM/CWFM/IMFQ or between PM/CWFM/CWFQ and PM/CWFM/IMFQ. Note again that total gas amount for intermittent supply of quenching gas is lower than that for continuous supply. In spite of that, nanoparticles with similar mean diameter could be obtained. This means that use of intermittent supply of quenching gas efficiently produces area with a high temperature gradient where evaporated material is cooled down.

Table 1.Experimental condition.

Input power	**20 kW**
Pressure	**300 torr**
Gas composition	**90%Ar+10%O_2**
Carrier gas flow rate	**Ar:4 slpm**
Quenching gas flow rate	**Ar:50 slpm (40 slpm for intermit. supply)**
Quenching gas supply direction	**Radial**
Immersion length of water-cooled probe	**134 mm**
Modulation cycle	**15 ms**
Shimmer current level SCL	**80%**
Duty factor	**80%**
Ti powder feed rate	**3～4 g/min**



(a)NM/CWFM/CWFQ (b) NM/IMFM/CWFQ (c) NM/CWFM/IMFQ
(d)PM/CWFM/CWFQ (e) PM/CWFM/IMFQ

Fig.2. SEM images of synthesized TiO_2 nanoparticles. PM means the pulse modulation of the coil current, whereas NM means non-modulation. CWFM means the continuous feeding of raw material, CWFQ means the continuous feeding of quenching gas, IMFM means intermittent feeding of raw materials, and IMFQ means intermittent feeding of quenching materials.

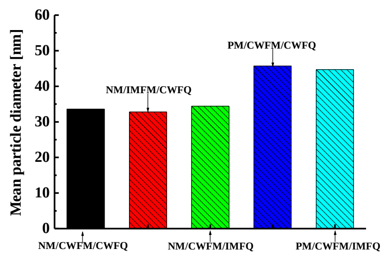


Fig.3. Mean particle diameter of produced particles.

References

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