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Original Research Paper

Decomposition of solution droplets under the influence of thermal convection over a heated horizontal plate

Ferry Faizal [a], Mitsuki Wada [a], Sosuke Koike [b], I. Wuled Lenggoro [a,b,*]

[a] Graduate School of Bio-Applications and Systems Engineering, Tokyo University of Agriculture and Technology, 2-24-16 Nakacho, Koganei, Tokyo 184-8588, Japan
[b] Department of Chemical Engineering, Tokyo University of Agriculture and Technology, 2-24-16 Nakacho, Koganei, Tokyo 184-8588, Japan

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ABSTRACT

Experimental approaches using an open-plate reactor were proposed to investigate the thermal decomposition and deposition processes of solution droplets sprayed over a heated horizontal plate. The temperature-dependent flow condition of the carrier gas may influence the properties of incoming sprayed aerosols. Zinc oxide (ZnO) particle layers were synthesized by spraying zinc acetate solution to the plate with deposition angle of 45°. The prepared particle layers and a numerical simulation verified the presence of thermally generated convection and fluid instability. Correlation between the size and morphology of deposited aerosols and the dynamical condition due to temperature distribution above the plate has been analyzed. A short distance (2 cm) between the nozzle and the plate will prepare finer (nanometer-scaled) ZnO particle layers. ZnO with higher crystallinity can be prepared using one-step route (directly spraying at 450 °C) compared with those of two-step heating procedures (150 °C – spraying then 450 °C – re-heating in 1 h), which consume higher energy.

1. Introduction

Spray route is one of the well-established methods to produce particle layer, by deposition of particles from solution/colloidal suspension. This method has been applied to synthesize various types of materials in atmospheric pressure, for application in sensing material, transparent conductive film, photo-catalyst and corrosion protection [1–7].

Considering the operation of spraying “cool” aerosol droplets onto a “hot” substrate, one of the issues is the difficulties of the aerosol droplets to deposit onto the substrate (i.e. the productivity). Previous studies discussed about thermophoresis as the main cause, but other studies claimed that thermophoresis was insignificant and the Leidenfrost phenomena (i.e., a heated object becomes insulated by a vapour layer and vapour generation may cause the levitation of aerosol droplets) was proposed [8,9]. It appears that the temperature is not only critical for driving the thermal decomposition of solution aerosol droplets but also influences the force balance over the heated zone due to the presence of thermal convection.

In the present study, an experimental system was designed to investigate the thermal decomposition and deposition processes of aerosol droplets in an open-plate reactor by taking ZnO as a model material. ZnO is a well-known metal oxide and its properties have been already widely studied [3,10]. The temperature distribution around the designed reactor was studied by numerical approaches.

The effect of experimental parameters such as substrate temperature, flow rate, spray distance and spray angle [3,5,6,11–15] on the formation of spray derived particle layer have been discussed in a number studies. However, the interaction between parameters such as flow-temperature which affect the dynamical condition inside the reactor was rarely discussed [8,16].

2. Method

The reactor was positioned in a simple semi-open chamber (dimension: 25 cm × 25 cm × 45 cm), with a heated zone (a circle with diameter: 15 cm) on the floor for positioning the substrate. A cylindrical pipes (inner diameter: 4 mm, outer diameter: 6 mm) is used as inlet, directed 45° to the horizontal plane [14,17]. This angle has been assumed to prevent the “unwanted” large droplets to deposit on the substrate. The designed chamber was combined with an ultrasonic nebulizer (NE-U17, Omron, Japan) through a

* Corresponding author at: Graduate School of Bio-Applications and Systems Engineering, Tokyo University of Agriculture and Technology, 2-24-16 Nakacho, Koganei, Tokyo 184-8588, Japan.
E-mail address: wuled.@cc.tuat.ac.jp (I.W. Lenggoro).

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simple droplet classifier (lab-made), driven by controlled air flow as a carrier gas (Fig. 1).

To deposit particle on the substrate, generated solution droplets (0.05 mol/l, zinc acetate ($Zn(O_2CCH_3)_2(H_2O)_2$)) was delivered in constant flow rate (1.5 l/min) to the 150 °C, 300 °C and 450 °C heated horizontal plate for 30 min. The temperature values were selected based on studies in thermal decomposition process of the ZnO from zinc acetate solution which has critical temperature around 280–340 °C [18–20]. In order to investigate the effect of force balance between carrier gas (forced convection) and buoyancy (thermal convection), the distance between the spray nozzle and the horizontal substrate (a silicon wafer) was varied at 2, 4 and 6 cm at constant temperature of the substrate. Numerical simulation has been conducted to investigate the presence thermal convection due to the parameters used in the experimental work (Appendix).

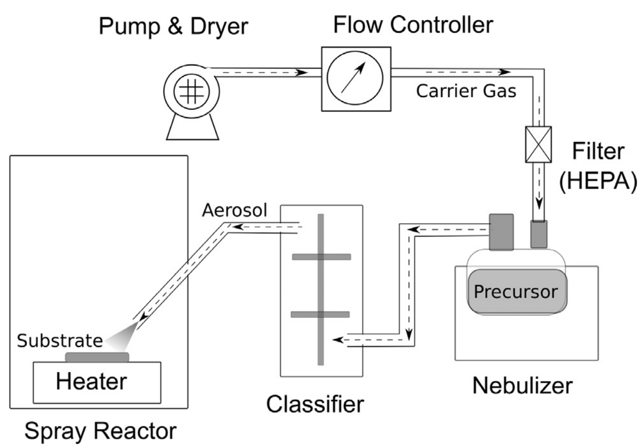

Fig. 1. Experimental setup of heated-plate spray pyrolysis experiment.

Powder X-ray Diffraction method (XRD, Rigaku, Japan) has been use to investigate the crystallinity of the particle layer. The size and morphology of the resulted layer was analyzed by two scanning electron microscopes (SEM and FE-SEM, JEOL, Japan). The optical vibrational properties of the deposited layer were investigated using Raman spectrometer (Micro-Raman, Thermo Scientific Nicolet Almega, USA).

3. Results and discussion

3.1. Crystallinity and Raman spectra of particle layers

The effect of different force balance due to thermal convection and carrier gas flow might also be indicated by XRD spectra,

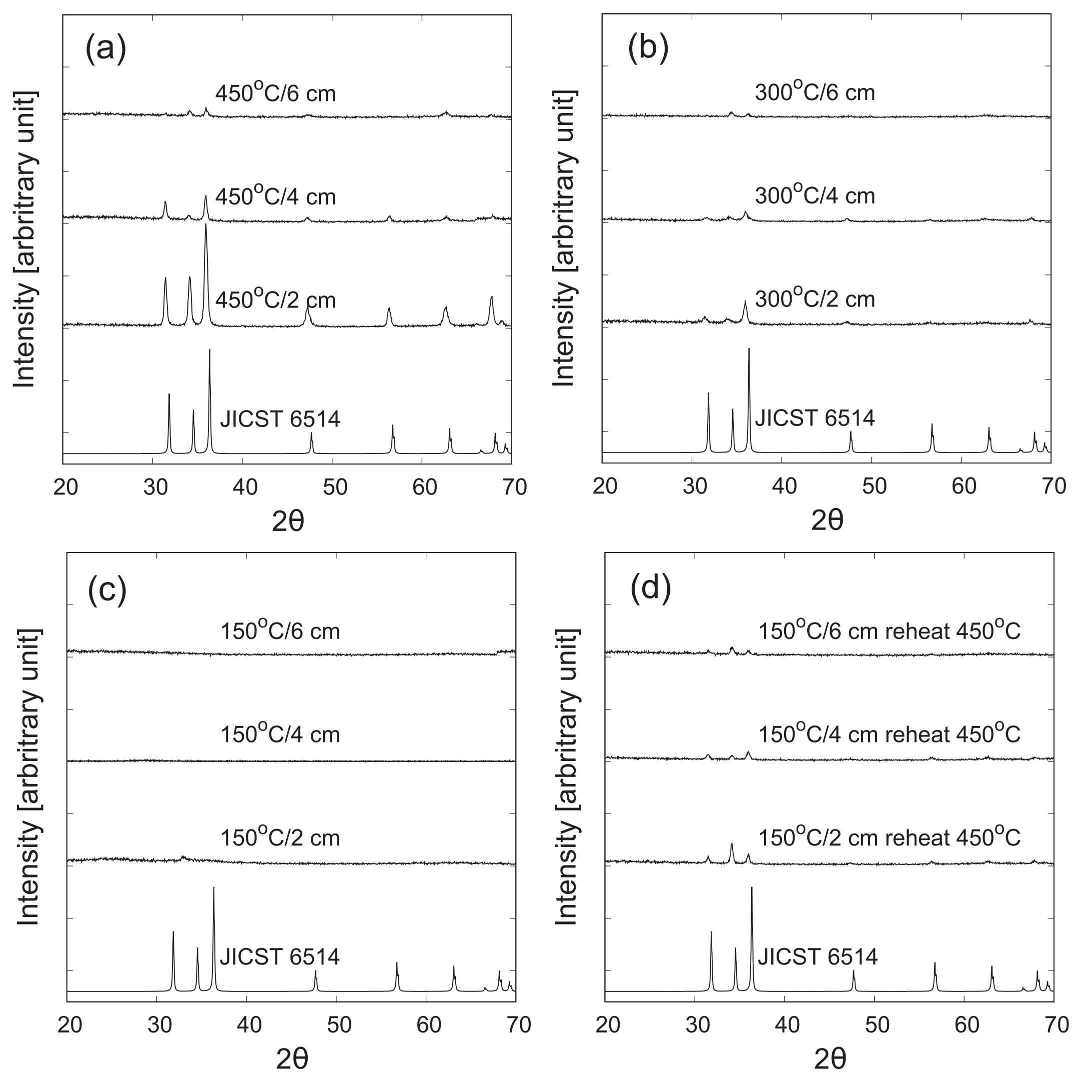

Fig. 2. XRD spectra of synthesized thin films. (a) at 450 °C. (b) at 300 °C. (c) at 150 °C. (d) at 150 °C followed with reheating for re-crystallization at 450 °C in one hour.

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