Capture of compositional distribution for binary alloy during induction thermal plasma process

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Abstract: Quantitative investigation of the compositional distribution spatially of nanoparticles obtained through the thermal plasma process is carried out using a Ni-Cu and Sm-Fe system. In this experiment, the spatial distribution of composition was observed perpendicular to the plasma flow direction for the Ni-Cu system and parallel to the plasma flow direction for the Sm-Fe system. These results suggested that the ITP process has a possibility to utilize as a high throughput process for materials research.

Keywords: binary, compositional distribution, thermal plasma process, combinatorial

1. Background

Materials informatics and process informatics are recognized as powerful tools for discovering new compounds and developing new processes. In order to use these, a certain amount of experimental and analytical data is required. Therefore, how to obtain and collect these data efficiently is a very important theme. From the viewpoint of material synthesis, high-throughput synthesis using thin films can be reported[1]. However, for bulk materials, it is still difficult to obtain samples with many different compositions and conditions in a single experiment.

In alloy nanoparticle formation from a gas phase through the nucleation, condensation and coagulation processes in the thermal plasma process, the composition of the obtained nanoparticles has a distribution since the intrinsic properties of surface tension and vapor pressure strongly affect the composition distribution. If the compositional distribution, which seems to be a drawback, can be utilized to collect the powder with certain composition, the ITP process may be utilized as a high-throughput process. Especially in the ITP process, metastable phases can be obtained due to the high quenching process[2], so the scope of compound research is wide.

Calculations by Vorobev et al. suggest that the thermal plasma jet composition variation, may manifest itself as a spatial distribution of particle composition[3]. However, there are no reports of experimental verification of this prediction.

In this study, the correlation between the composition distribution and particle diameter of the obtained powder and the spatial distribution of composition during the process were clarified experimentally. We select Ni-Cu and Sm-Fe systems, which are expected to have a large composition distribution due to the large difference in vapor pressure and surface tension. In addition to the experimental evaluation, the formation mechanism of alloy nanopowder was estimated computationally based on a binary aerosol formation-growth model (for details, see Ref. [4, 5]) for Ni-Cu.

By making it possible to investigate the spread of the spatial composition in more detail experimentally and computationally, this process has high potential as a high-throughput powder process that can greatly contribute to future material research including the stable and metastable phase.

2. Experimental

The mixed powder of Ni (~3 μm and purity of 99.99 %) and Cu (~5 μm and purity of 99.9 %), and Fe (~3 μm and purity of 99.99 %) and Sm (~20 μm and purity of 99.9 %) were used as a starting powder for the ITP process. The atomic ratio were set to Ni : Cu = x : 1-x (x = 0.3 and 0.4) and Sm : Fe = 12.5 : 87.5 (at %), respectively. Because the Sm power was not commercially available, we prepared Sm powder using the gas atomize process. During the ITP process, the pressure in the chamber was controlled to 100 kPa. The flow rates were 35 and 3 L/min, respectively. In this study, a water-cooled copper bar with a diameter of 3/8 inch was introduced into the chamber to capture the nanopowder obtained by the ITP process.

As shown in Fig. 1, the water-cooled copper bars were placed 57 mm (upper) and 307 mm (middle) below the powder feeder port, respectively. The duration of the thermal plasma process was only about 10 s. After the process, the EDX profile was acquired for each position to quantitatively evaluate the spatial composition change.

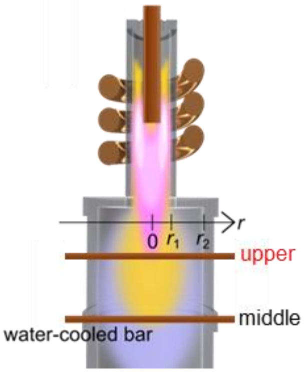

Figure 1. Schematic illustration of the geometry of the thermal plasma chamber with a water-cooled copper bar placed at the position near(upper) and far(middle) from the plasma.

3. Result and discussion

Figure 2(a) shows the Cu composition on a water-cooled bar as a function of radial position, *r*, from the center of the chamber (r = 0 mm) to the edge. The composition changed as a function of the radial direction of the chamber. The Cu concentration took mixinum of x = 0.5 at r = ~20 mm and mininum of x = 0.3 at r = ~40 mm for $Ni_{0.6}Cu_{0.4}$ sample, indicating that the composition distribution reached 20at%. Even when the composition ratio of the starting powder was changed, the same tendency of composition change as afunction of the position *r* was obtained. According to the numerical calculation of the formation mechanism of Ni-Cu alloy nanoparticles, the composition distribution can be understood to be the temperature difference between the nucleation and solidification processes of Ni and Cu[6].

However, the origin of the compositional oscillations along the radial direction is still unclear and a deeper understanding requires further experiments with different positions of the bar relative to the torch and simulations of the spatial compositional distribution in the chamber.

For the Sm-Fe system, the same experiment as the Ni-Cu system was conducted by changing the collection position. The results are shown in Figure 2(b). Unlike the Ni-Cu system, no compositional distribution of Sm-Fe was observed in the radial direction of the chamber. On the other hand, it was found that the composition obtained differs depending on the collection position. This is probably because the vapor pressure of Sm is so high that the coagulation of Sm occurs at a lower temperature than that of Fe. In other words, when the sample collection position is high, Sm is still in a gaseous state and cannot be collected with the bar. Therefore, it is presumed that the difference in the composition of Sm and Fe in the direction of plasma propagation (the vertical direction in the figure 1) is due to the difference in the coagulation rate of Sm. Based on these considerations, in order to utilize the thermal plasma process as a combinatorial process, it might be necessary to collect the sample in the vertical direction in a single experiment.

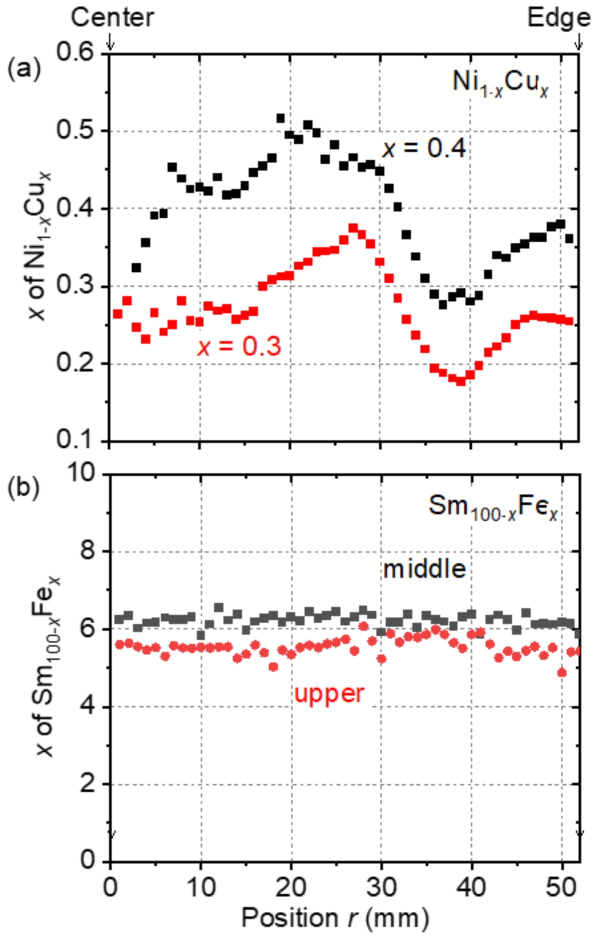

Figure 2 Cu and Sm concentration as a function of the radial position, *r*, from the center (r = 0 mm) to the edge of the chamber for (a)$Ni_{0.7}Cu_{0.3}$ and $Ni_{0.6}Cu_{0.4}$ and (b) Sm_7Fe_{93} nanopowders. For Ni-Cu case, the position of the bar was middle. Reproduced by permission from[6], copyright [2023, Elsevier] for (a).

4. Summary

The spatial distribution of the composition was quantitatively evaluated by an experimental approach. For the Ni-Cu system, the composition of Cu changed depending on the radial direction of the chamber. While, for the Sm-Fe system, the composition of Sm changed depending on the distance from the torch instead of changing depending on the radial direction of the chamber. These phenomena indicate that it is possible to obtain a powder with a continuous composition that can be sorted by location in one experiment.

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6. References

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