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Original Research Paper

Preparation of silver coated nickel particles by thermal plasma with pre-treatment using ball milling

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ABSTRACT

Recently, reducing noble metals in electric devices has been identified as a key factor to lower product cost. Among these, noble metal coated particles are considered an alternative with the potential to dramatically reduce the usage of noble metals. A dense coating of noble metals over all surfaces is desirable for maintaining the properties of noble metal. However, our previous research showed that coated surfaces onto which the nanoparticles were attached were non-uniform because of evaporation of the raw materials. Therefore, in this study, we improved the coverage ratio of silver coated nickel particles using thermal plasma processing followed by ball mill pre-treatment. Silver and nickel particles were mixed using a ball mill, then injected into the thermal plasma jet. The silver particles were subsequently attached onto the surface of the nickel particles through ball mill processing, after which the silver attached nickel particles were melted through thermal plasma processing to produce silver coated nickel spherical particles. The cross section of the as-prepared particles showed a dense silver shell and nickel core, while the sintered body of the as-prepared particles showed the net-like silver covering over the nickel cores. These findings suggest that attachment of silver on nickel could lead to complete silver coatings by limiting the formation of nanoparticles.

1. Introduction

Recently, reducing noble metal use has gained increased attention in various industrial fields because of their close association with production costs. The methods to reduce noble metal use include development of alternative materials [1,2] and core-shell particles [3–5], reducing noble metal loads [6,7], and alloying with non-noble metals [8]. Among these, noble metal coated particles having a core-shell structure comprise one of the most promising alternative materials to reduce noble metal use. Silver, which is one of the most frequently used noble metals, is commonly applied as a conductive paste [9–13] that contains silver powder corresponding to over 60 wt% of its total weight. Therefore, the replacement of costly silver with an alternative material can considerably reduce the production costs of silver paste [14–16]. Performance of conductive film is depending on sheet resistance of that. It is expected that the acceptable level of sheet resistance is obtained with core-shell structure if the highly conductive channel is well-dispersed [17]. Therefore, core-shell particles with silver shells have attracted a great deal of attention as a promising alternative. Various materials including copper, nickel, and tin have been reported as core particles. Among these, nickel has great potential for use as a core material owing to its low solubility in silver (<0.1 wt% at room temperature), high melting point (1728 K) and high electrical conductivity (1.43×10^7 S/m). In a previous study, we prepared silver coated nickel particles but encountered several problems that hindered its application [18]. One was a low density of the coating layer owing to the presence of silver nanoparticles that covered the nickel core, inducing formation of a porous structure during sintering and finally lowering its electrical conductivity. Because of the higher thermal conductivity and lower boiling temperature of silver (429 $Wm^{-1} K^{-1}$ and 2435 K, respectively) than nickel (90.9 $Wm^{-1} K^{-1}$ and 3187 K, respectively), silver is easily vaporized in the plasma and then forms nanoparticles on the nickel particles, similar to what occurs during the vapor phase method. Another problem is that studies dealing with the preparation of micro-sized core-shell particles are insufficient because nano-sized core-shell particles have been frequently focused on. However, micro-sized silver particles of

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several micrometers are still frequently used as raw materials for conductive paste in industrial uses, even though advantages of applying nano-paste or nano-ink have been reported [19]. Nano-sized core-shell structures with dense shells have been successfully synthesized using various method including precipitation [20,21], electroless plating [22], microwave-assisted synthesis [23] and metal reduction [24]. However, it seems that there are some challenges to obtaining micro-sized core-shell particles possessing a dense shell [25,26].

To address the aforementioned problems, it is necessary to develop a micro-sized silver coated nickel particle with a dense silver layer. Although various methods to synthesize core-shell particles using plasma have been attempted, the formation of dense coating layers has been a problem, regardless of material type [18,27,28]. Therefore, it is necessary to control the vaporization of materials with low vaporization temperatures to obtain densely coated particles.

Here, we report the preparation of nickel particles covered by a dense silver layer using a ball mill process followed by the thermal plasma method. The ball mill process was employed to mix silver and nickel powder before plasma treatment since separate injection of each raw material was found to induce the evaporation of silver in our previous study [18]. The plasma-treated particles contained a spherical nickel core and dense silver external layer.

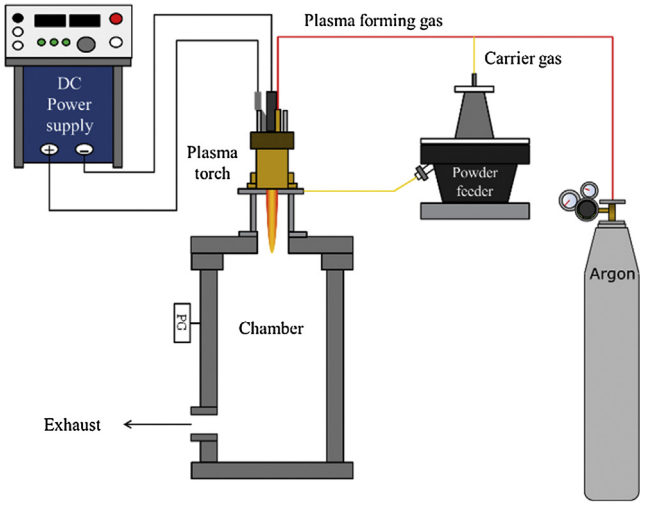

Fig. 1. Experimental set-up for the preparation of Ag coated Ni particles.

Table 1
Detailed plasma operating conditions.

Discharge gas	Ar 15 L/min
Current	300 A
Voltage	30 V
Power	9.0 kW
Carrier gas	Ar 5 L/min
Pressure	760 torr
Feeding rate	3 g/min

2. Experiments

Silver powder (50 g) and nickel powder (50 g) were mixed at a weight ratio of 1:1, then put into polytetrafluoroethylene (PTFE) bottles with zirconia balls. This Ag-Ni mixture powder was then milled at 150 rpm for 15 h.

Next, the Ag-Ni mixture powder was treated using a DC non-transferred thermal plasma system composed of a DC power supply, plasma torch, quenching tube, chamber and powder feeder (Fig. 1). Briefly, the mixture powder was put into the powder feeder and then injected vertically into the plasma jet with 5 L of argon carrier gas. The detailed operating conditions are summarized in Table 1. Following plasma-treatment, particles were collected from the bottom of the chamber and at a position near the plasma torch, respectively.

The powder samples were analyzed using an X-ray diffractometer (XRD, DMAX 2500, Rigaku Co.), a field emission scanning electron microscope (FE-SEM, S-4300, Hitachi Co.), an energy dispersive X-ray spectroscope (EDS) and a focused ion beam scanning electron microscope (FIB-SEM, JIB-4601F, Jeol).

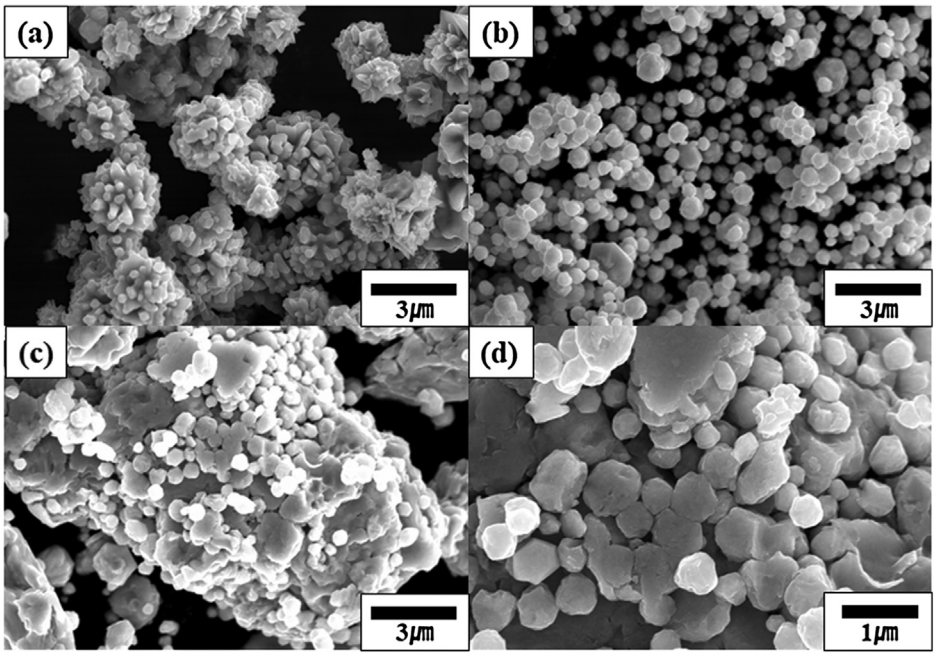

Fig. 2. SEM images of the raw materials; (a) Ni, (b) Ag and (c, d) Ag-Ni mixture powder after ball milling.

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