



# Flash light sintering of nickel nanoparticles for printed electronics



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## ABSTRACT

In this paper, a sintering process for nickel nanoparticles using flash light irradiation was investigated for multi-layer ceramic capacitors, magnetic devices and printed electronics. The existence of a small amount of 10 nm size nanoparticles in the ink significantly improved the flash light absorption and induced the melting and sintering of the nanoparticles. Due to this phenomenon, uniform diameter nickel nanoparticles (50 nm) could not be sintered, while the nickel nanoparticles with varying diameters (5–500 nm) were sintered by flash light irradiation.

In order to acquire high electrical conductivity in the sintered nickel nanoparticles, several flash light sintering parameters such as light energy and pulsed light patterns were optimized. Also, the nickel nano-ink was optimized by changing the weight fractions of organic binder in the ink.

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## 1. Introduction

Nickel has been used extensively in various applications, including multilayer ceramic capacitors (MLCCs), magnetic devices and electrodes for supercapacitors in the electronics industry [1–13]. In MLCCs, the development of base-metal electrodes (BMEs) played an important role in expanding the potential applications. For BME applications, nickel electrodes recently attracted a lot of attention as they can reduce fabrication cost at least 10-fold compared to materials conventionally used for BME inner electrodes such as palladium and gold [1–5].

In magnetic devices, transition metal magnetic nanoparticles such as Fe, Co and Ni are widely used owing to their ferromagnetic properties. Also, these materials have been studied extensively because of their size- and shape-dependent physical, chemical and magnetic properties. Among these, nickel nanoparticles are mainly used due to their low cost and favorable melting temperature [6–9].

Recently, printed electronics have been studied as an alternative to the conventional photolithography process for electronic devices as printing offers many advantages such as substrate flexibility, low cost and ease of processing [10]. Most researchers have used gold and silver nanoparticles to fabricate electrical circuits due to their low melting temperature, low electrical resistivity, and thermodynamic stability (no oxidation phenomenon) [10–13]. However, the cost of silver and gold are about \$17 and \$1100 per ounce, while the cost of copper and nickel are about 20 cents and 53 cents per ounce, respectively [11]. Unfortunately, copper and nickel nanoparticles are easily oxidized upon contact with air. This

makes the sintering process very complicated as an inert or reductive gas must be supplied to prevent oxidation or to reduce the oxide layers and transform them to a pure metal. Therefore, in our previous studies, we proposed a flash light sintering method to overcome the problems of thermal sintering methods [12–15]. The flash light sintering method has merit in that the process is carried out at room temperature under ambient conditions at ultra-fast speed (~ms). In the sintering of copper nanoparticles, the flash light irradiation was shown to reduce the copper oxide shell and transform it to pure copper when used in combination with a poly(N-vinylpyrrolidone) polymer coating on the copper nanoparticles [12]. In a separate work, in-situ reactive reducing and sintering of copper nanoparticles at room temperature and ambient conditions was demonstrated [14]. Furthermore, flash light sintering can prevent damage to various flexible substrates that may have a low melting or glass transition temperatures such as polyethylene and polyethylene terephthalate [14]. Light-assisted sintering of metal particles has been employed by several researchers. The summary of these studies are shown in Table 1. However, a study of the flash light sintering of nickel nanoparticles has not been conducted yet.

As mentioned above, the sintering of nickel nanoparticles is crucial for fabrication of MLCCs, magnetic devices, and printed electronics. However, sintering of nickel nanoparticles by a thermal sintering method is impossible without an inert or reducing gas chamber due to the fact that the melting temperature (300–600 °C) is higher than the oxidation temperature (135 °C); as such, nickel nanoparticles oxidize prior to melting or sintering [3,4,29]. The flash light sintering method mentioned above would be a promising alternative to alleviate these problems.

Therefore, in this work, we investigated the flash light sintering process for nickel nanoparticles. Specifically, we investigated the effect of

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**Table 1**

The summary of the reported light-assisted sintering with respect to applied metal, light source, substrate, and conductivity.

Metal	Light source	Substrate	Conductivity	Author
Silver	Infrared ray (IR)	Glass	3 $\mu\Omega$ cm	Tobjörk [16]
		Paper	6 $\mu\Omega$ cm	
Silver	Flash light	Glass	24 $\mu\Omega$ cm	Galagan [17]
Silver	Flash light	Glass	9.6 $\mu\Omega$ cm	Abbel [18]
Silver	Flash light	PI	6.2 $\mu\Omega$ cm	Yung [19]
Silver	Flash light	PET	1.5 $\Omega$ /sq	Hosel [20]
Silver	Flash light	PET	4.9 $\mu\Omega$ cm	Kang [21]
Silver	Flash light	PET	0.95 $\Omega$ /sq	Chung [22]
Silver	Flash light	PI	–	Lee [13]
Silver	Flash light	PI	3.6 $\mu\Omega$ cm	Park [15]
Silver	Laser	Glass	–	Peng [23]
Copper	Flash light	PI	5 $\mu\Omega$ cm	Kim [24]
Copper	Flash light	PI	5 $\mu\Omega$ cm	Ryu [12]
Copper	Flash light	PI	173 $\mu\Omega$ cm	Han [25]
Copper	Flash light	PI	72 $\Omega$ /sq	Hwang [14]
Copper	Flash light	PET	388.3 $\mu\Omega$ cm	Wang [26]
Copper, Ni	Laser	–	–	Agarwala [27]
Copper, Sn, Ni	Laser	–	–	Khaing [28]

particle size distribution on the flash light sintering by comparing two nanoparticle systems (one with a uniform diameter of about 50 nm, the other with various particle diameters from 5 nm to 500 nm). In order to achieve high electrical conductivity in the sintered nickel nanoparticles, several flash light sintering parameters such as light energy and pulsed light patterns were optimized. Also, the nickel nanoink was optimized by changing the weight fractions of organic binder in the ink.

## 2. Experiments

Two types of nickel nanoparticles were used: QSI nickel nanoparticles (QNI, 99.9% > purity, Quantum Sphere Inc.) and Sigma-Aldrich nickel nanoparticles (SNI, 99.9% > purity, Sigma-Aldrich Co.). QSI nickel nanoparticles had a uniform diameter of about 50 nm, while Sigma-Aldrich nickel nanoparticles had various particle diameters ranging from 5 nm to 500 nm. The nickel nanoparticle ink was prepared as follows. First, 2.5 g of diethylene glycol (DEG, 99% > purity, Sigma-Aldrich Co.) and polyvinylpyrrolidone (PVP, wt.% 40,000, Sigma-Aldrich Co.) were mixed together under ultrasonication for 2 h. In order to study the influence of PVP on the flash light sintering, we varied the amount of PVP from 0.2 wt.% to 0.4 wt.% in the flash light sintering process. Second, 0.5 ml of N-dimethylformamide (99.8% purity, Sigma-Aldrich Co.) was added to a PVP/DEG solution under ultrasonication for 1 h. Finally, 2.0 g of nickel nanoparticles were added and dispersed in an ultrafast mixer (AJ300V, Ajintech) for 1 h. The viscosity of resulting nickel nanoink was in the range from 165 cSt to 350 cSt depending on the wt.% of PVP.

A polyimide (PI) film with a thickness of 225  $\mu\text{m}$  was prepared as a substrate material and was ultrasonicated in ethanol and distilled water for 10 min to remove surface contamination. Nickel nanoparticle inks were coated onto the PI substrate using a spin coater (SC-200, Nanotech Co.) at 800 rpm for 30 s. The coated films were 2 cm  $\times$  2 cm. Once coated, the resulting films were thoroughly dried on a hot plate (HSD 180, MISUNG SCIENTIFIC Co.) at 150  $^{\circ}\text{C}$  for 20 min before flash light sintering. For the flash light generation, a xenon flash lamp (PerkinElmer Co.) was used. This lamp generates flash light via arc plasma generation with wavelengths ranging from 380 nm to 1.0  $\mu\text{m}$  [12]. Manipulating the pulse number and intensity of the arc plasma enables control of the energy from 0.001 J/cm<sup>2</sup> to 100 J/cm<sup>2</sup>. Transformations of the microstructure were observed by scanning electron microscopy (SEM, S4800, Hitachi, 15 kV operating voltage) and focused ion beam (FIB, LYRA FEG 1, TESCAN, 15 kV operating voltage), and conductivity was measured using a four-point probe method with a source meter (2015 THD, Keithley). Also, we conducted X-ray diffraction

(XRD, D/MAX RINT 2000, Rigaku) using the Bragg–Brentano geometry with Cu K $\alpha$  radiation to analyze the particle size and phases of materials. UV–vis (S-4100, Scinco) test were conducted to characterize the optical properties of the nickel nanoparticles, respectively. The samples of UV–vis test were prepared by 1–10 wt.% of nickel nanoparticles mixed with ethanol. The mixed solution with ethanol and nickel nanoparticle was treated with ultrasonication in 10 min to acquire the uniform dispersion of the nickel nanoparticles in ethanol.

Flash light sintering was divided into two-steps, preheating and the main sintering step, to acquire a fairly packed sintered film structure [15]. The preheating step was conducted to gradually remove the organic binder surrounding the nickel nanoparticles before the main sintering step (Fig. 1) [15]. Energy used for the preheating and the main sintering step was varied from 7.5 J/cm<sup>2</sup> to 17.5 J/cm<sup>2</sup>. In the preheating step, 15 rectangular pulses (5 ms duration with a 30 ms pulse gap between pulses) were applied. On the other hand, a single pulsed light was irradiated in the main sintering step. The energy of the irradiated light was measured using a power meter (Nova II, People Laser Tech.).

## 3. Results and discussion

Fig. 2 shows the XRD patterns of the two types of dried nickel films. XRD patterns of all the nickel films exhibited diffraction peaks of the (111), (200), (220), and (311) planes of face-centered cubic nickel at 2 $\theta$  values of 44.5 $^{\circ}$ , 51.8 $^{\circ}$ , 76.5 $^{\circ}$ , and 92.9 $^{\circ}$ , respectively [30–32]. However, the peaks of the nickel oxide were not observed in SNI and QNI films. Also, the peaks of the SNI film were sharper than those of the QNI film due to their mean size. Based on the peaks in the spectrum of each nickel film, the mean size of the nickel nanoparticles was calculated using Scherrer's formula as follows:

$$L = (0.9\lambda)/(B \cdot \cos \theta) \quad (1)$$

where L is the mean size of the alloy particles,  $\lambda$  is the X-ray wavelength (Cu K $\alpha$ ,  $\lambda = 0.1541$  nm),  $\theta$  is the maximum angle of the peaks, and B is the half-peak width for in radians. For accurate calculation, the Lorentzian fitting was conducted when the mean size of QNI nanoparticles was calculated due to the broadening of the XRD peaks of QNI nanoparticles. Meanwhile, the resolved K $\alpha$ 1 lines were used for calculating the mean size of SNI nanoparticles due to the very sharp XRD peaks of SNI nanoparticles. Thus, the mean size of QNI was calculated to be 7.16  $\pm$  1.6 nm, and that of SNI was 35.45  $\pm$  7.5 nm.

To determine the morphology of the nickel particles, SEM images were taken with (see inset Fig. 3). The SEM images show the spherical shape and size distribution of each type of nickel nanoparticles. The SEM image of dried QNI nanoparticles showed a regular size of about 50 nm (see inset figure in Fig. 3a). However, the SEM image of the dried SNI nanoparticles showed various diameters from 10 nm to 400 nm (see inset figure in Fig. 3b).

To quantify the size distribution for the types of nickel nanoparticles, we conducted a size distribution analysis using a size distribution analyzer (Scatteroscope I, K-ONE LTD). As shown in Fig. 3, the QNI nanoparticles showed a log-normal distribution with an average diameter of 52.4 nm. Meanwhile, the SNI nanoparticles showed three major diameter peaks at 8 nm, 50 nm and 200 nm. These results are similar to SEM images (insets in Fig. 3). From the relative particle number distribution, the relative weight fraction of the nanoparticles was calculated using the density of nickel and the volume a sphere as follows:

$$m = (4/3) \cdot (\rho\pi r^3) \quad (2)$$

The blue line in Fig. 3 is the relative weight fraction of nanoparticles with respect to their diameter. It is noteworthy that 0.01 wt.% of the nanoparticles with diameters smaller than 10 nm (34.27% in particle number portion) were present in the SNI nanoparticles. The discussion

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