Optics and Lasers in Engineering 101 (2018) 78–84

Contents lists available at ScienceDirect

Optics and Lasers in Engineering

journal homepage: www.elsevier.com/locate/optlaseng

Low-cost optical fabrication of flexible copper electrode via laser-induced reductive sintering and adhesive transfer

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A R T I C L E I N F O

Keywords:
Flexible electrode
Copper oxide nanoparticle
Reductive sintering
Adhesive transfer
Laser-assisted fabrication

A B S T R A C T

Fabricating copper electrodes on heat-sensitive polymer films in air is highly challenging owing to the need of expensive copper nanoparticles, rapid oxidation of precursor during sintering, and limitation of sintering temperature to prevent the thermal damage of the polymer film. A laser-induced hybrid process of reductive sintering and adhesive transfer is demonstrated to cost-effectively fabricate copper electrode on a polyethylene film with a thermal resistance below 100 °C. A laser-induced reductive sintering process directly fabricates a high-conductive copper electrode onto a glass donor from copper oxide nanoparticle solution via photo-thermochemical reduction and agglomeration of copper oxide nanoparticles. The sintered copper patterns were transferred in parallel to a heat-sensitive polyethylene film through self-selective surface adhesion of the film, which was generated by the selective laser absorption of the copper pattern. The method reported here could become one of the most important manufacturing technologies for fabricating low-cost wearable and disposable electronics.

1. Introduction

A variety of alternative fabrication methods with the potential to realize flexible and wearable smart devices that are not only marketable but will also dramatically increase future demand are being studied [1,2]. The key to realize these devices is to cost-effectively manufacture a high conductive electrode on a low-cost flexible film [3–5]. However, an expensive facility and highly complicated production processes are inevitably involved in the fabrication of flexible conductors since photolithography based on indirect pattering regime additionally requires a series of pre- and post-vacuum depositions, use of photoresists, and toxic chemical etching [6,7]. Selectively depositing metal nanoparticles (NPs), such as gold and silver, on heat sensitive polymer films, which is represented by ink-jet and roll printing, would be the most promising alternative to resolve these issues [8,9]. Organic suspensions of such noble metals are usually used as precursors because they show good chemical stability during high-temperature sintering and possess a high electrical conductivity as well [10,11]. However, the overall production cost will increase as these noble metals are more expensive than other low-cost conductive metals such as copper (Cu). Although Cu electrode has a comparable conductivity to that of noble metals, it rapidly oxidizes at high temperatures in air [12]; thus, either an inert environment or a vacuum chamber is additionally required to prevent the unwanted oxidation of Cu NPs during sintering so that Cu NPs can be applied to the printing method [13–15]. In addition, even though bulk Cu is cheaper than the noble metals, the cost of Cu NPs rises in the same range as that of the noble metal nanoparticles. Moreover, the long-term storage of Cu NPs without aggregation and oxidation is difficult. For these reasons, direct printing of Cu NPs has no great merit over the typical printing methods that use noble metals [16–18]. Therefore, Cu NPs should be replaced with a more cost-effective precursor. As a conventional approach, in order to achieve high conductivity via thermal aggregation of NPs, the printed NPs should be sintered at high temperatures above 200 °C in the typical printing methods. Heat-resistive polymer films such as polyimide, which can withstand heat up to approximately 300 °C without thermal damages, should be employed as a substrate to avoid heat-induced warpage that occurs during sintering [9,13–15]. However, since conventional heat-resistive films are expensive as compared to thermoplastic films such as polyethylene terephthalate (PET) and most of these low-cost films are vulnerable to damages at temperatures above 100 °C, an alternative sintering method that enables the use of low-cost flexible films is required for the general use of wearable devices. In terms of the cost-effectiveness of material and substrate, a laser-induced sintering method has several advantages, such as the concurrent process of patterning and sintering, rapid sintering by intensive light, wide choice of substrates from glass to polymer films, easy design change due to a digital process, and better resolution than that of typical printing methods [19,20]. This method was applied to fabricate the metal electrode from noble metals such as gold [21–25] and silver [26–34], to cheap copper [35,36]. Recently, it has extended to fabricate the stretchable electronics through the selective junction welding of Cu nanowires [37,38]. However, the laser-induced sintering method needs pure Cu NPs suspensions, which are more expensive and unstable

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https://doi.org/10.1016/j.optlaseng.2017.09.027
Received 26 March 2017; Received in revised form 4 September 2017; Accepted 29 September 2017
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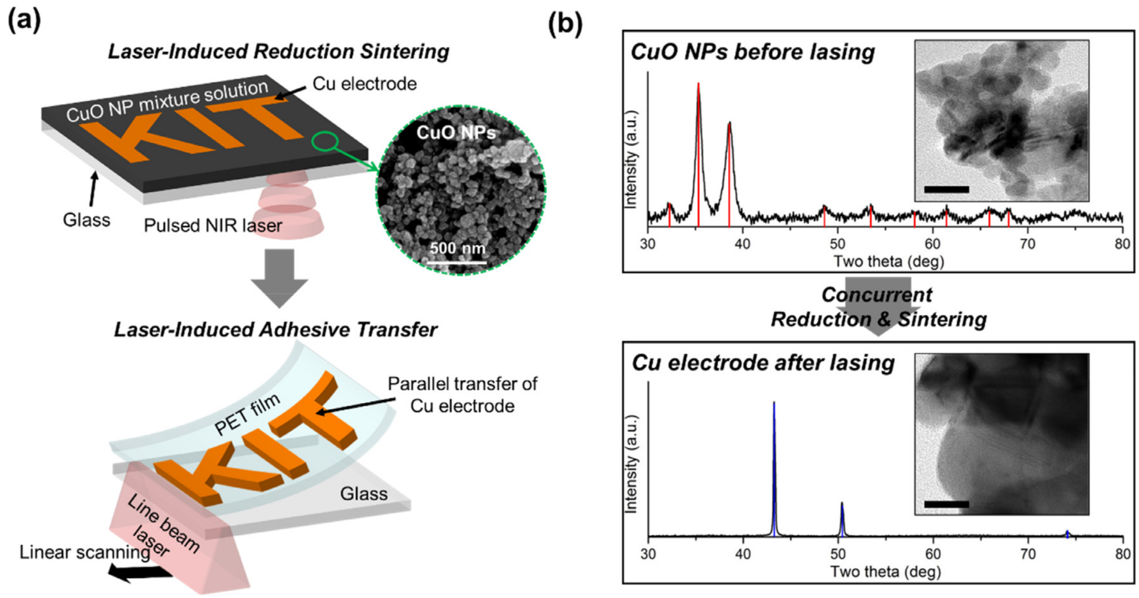

Fig. 1. (a) Schematics of overall fabrication procedure: laser-induced reductive sintering and laser-induced adhesive transfer. (b) XRD data obtained from CuO NPs before lasing and deoxidized Cu NPs after lasing. Insets show the corresponding transmission electron microscope (TEM) image. Scale bar is 20 nm.

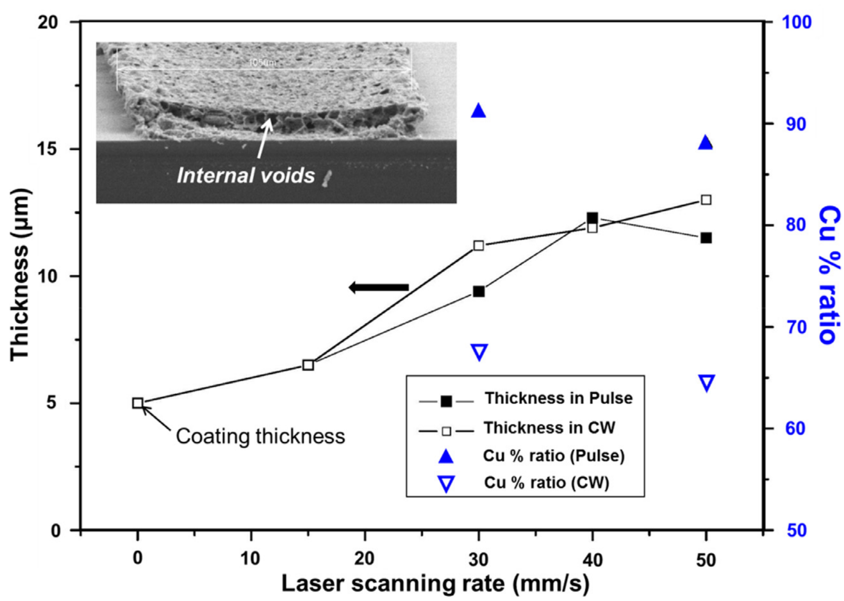

Fig. 2. Variation of pattern thickness and copper ratio with respect to laser scanning rates and laser irradiation modes (pulse and continuous wave).

than bulk Cu, and inevitably causes a large wastage of Cu precursor because the precursor in the un-irradiated region should be removed from the substrate [35–38]. And the electrical conductivity is also limited to avoid thermal damage of the low-cost flexile film since the temperature required to aggregate the NPs during laser sintering is higher than the glass transition temperature of the film. Thus, the specific resistance of Cu electrode fabricated on the low-cost film (PET) is at least 100 times higher than that of the Cu electrode fabricated on the PI films [36].

In this paper, a novel laser-induced hybrid method to fabricate Cu electrode on a heat-sensitive polymer film is reported to cost-effectively produce flexible and reliable electrodes under ambient conditions without the use of expensive precursors, vacuum facility, and chemical etchants. This laser-induced fabrication method has two steps: a laser-induced reductive sintering and a laser-induced adhesive transfer, as shown in Fig. 1(a). At first, the reductive sintering process directly fabricates a high-conductive Cu electrode from copper oxide (CuO) NP suspension on a glass substrate via photo-thermal reduction and agglomeration of CuO NPs. Then, the Cu pattern is transferred in parallel to a thermally delicate PET film by means of the self-selectively generated interface adhesion from the PET surface by the laser absorption of Cu pattern.

2. Experiment

2.1. Specimen preparation

CuO NPs of 20–50 nm (97.5% grade) were prepared as a precursor in this study, which is supplied by Alfa Aesar Co. The CuO NPs paste (60 wt%) was obtained by dispersing CuO NPs in the solution mixture composed of 13 wt% polyvinylpyrrolidone (PVP, mole weight 10,000), 27 wt% ethylene glycol (EG), and 60 wt% DI water. The PVP acts as a surfactant and the EG is used as a reduction agent of the CuO NPs. A viscosity of 5000 cps was obtained by adjusting the DI water so that it is appropriate for spin coating. The individual surface coating of CuO nanoparticle was not employed since a high-viscosity CuO nanoparticle solution is required to fabricate the thick Cu electrode. As a result, the