PHOTONIC CURING FOR ENHANCING THE PERFORMANCE OF ROLL-TO-ROLL PRINTED ELECTRONIC DEVICES

By

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ABSTRACT

The advent in printing technology promotes the possibility of roll-to-roll manufacturing process for low-cost, high throughput, and large area printing of the electronic devices in flexible substrate materials. However, the conductivity of the nanoparticle ink being utilized for printing process is the major challenge to compete with the existing silicon-based technology in terms of the device performance. A number of post-processing steps were proposed over the year to enhance the conductivity and among them thermal curing is the easiest solution so far. However, in high speed R2R processing system, thermal curing is not compatible owing to its long curing time or high temperature required to attain the desired conductivity. To overcome the issues of thermal curing, Intense Pulsed Light (IPL) photonic sintering has shown a promising capability to sinter the printed conducting patterns in milliseconds. Furthermore, photonic sintering can either be embedded together with R2R system during printing or in conjunction with the thermal curing is viewed as a viable approach to improve the conductivity of printed pattern. In this work, we studied different photonic sintering techniques and compared the results with the conventional thermal curing methodology in R2R printed near-field communication (NFC) antenna patterns. Experimental results showed that photonic sintering can reduce the resistance of the antenna more effectively than thermal curing even on polyethylene terephthalate (PET) with low melting point. The limitations of the present sintering techniques were highlighted from the prospect of future enhancement.

NOMENCLATURE

G	antenna gap between traces, mm
N	number of turns of antenna
Lx	length of antenna, mm
Ly	breadth of antenna, mm
W	width of the antenna trace, mm

INTRODUCTION

Roll-to-roll (R2R) printing is an emerging advanced additive for manufacturing flexible electronic devices with high throughput and low cost. As such, we have been implementing R2R gravure printing system (iPen Co. Korea) to realize several electronic devices such as RFID tag, transducers, sensors, thin film transistors (TFTs), antenna and so on. In order to obtain the acceptable result of those printed devices, the printing condition of the R2R gravure system were optimized by utilizing the printing speed of 6 m/min with a web tension of 5 kgf. Despite of those optimizations, the resistance of the conducting inks based on silver (Ag) or copper (Cu) nanoparticles is limiting the performance of those R2R gravure printed devices such as antenna[1,2], thin film transistors (TFTs) [3], diodes[4], and interconnecting lines. Therefore, the printed devices require a high operating voltage to compensate the power loss due to the high resistance. A convenient solution to reduce the resistance of the conducting ink based on Ag or Cu nanoparticles is to sinter at high temperature for gaining the best result but inexpensive substrates such as polyethylene terephthalate (PET) and papers cannot be used at high temperature (> 150°C) because of their low thermal stabilities.

In our printing system, the printing web (PET film) is passed through the drying chamber for thermal curing at 150°C for 5s which isn't sufficient for Ag or Cu nanoparticle to sinter for yielding low resistance. Thus, an additional thermal curing should be carried out outside the chamber for relatively long-time duration to obtain practical device performance. That extra curing process is impractical and inefficient for all R2R process and limits the commercialization of the R2R printed devices. In this regard, a photonic curing unit which can be implemented in R2R system independently or in conjunction with the thermal curing is viewed as a viable approach to improve the conductivity of printed Ag or Cu patterns. The photonic curing with a Xenon lamp can sinter the device at higher temperature for a couple of milliseconds so that the substrate like PET and paper can be utilized. Since R2R printed patterns are in few hundred nm to 1μm range in the thickness, only the top layer will be exposed to the light of Xenon lamp for short duration without damaging the underlying substrate.

PHOTONIC SINTERING

Among various sintering methods such as thermal sintering, chemical sintering, electrical sintering, plasma sintering, photonic sintering is more efficient and viable sintering technology that can be employed in R2R gravure printing system as it can sinter the printed patterns with different inks like Ag, Cu, nickel (Ni), and others selectively without affecting the substrate in short time period [5]. Among different photonic sintering techniques like IR sintering, UV sintering, laser sintering and intense pulsed light (IPL) sintering, IPL sintering is the most popular and effective where it can cover large sintering area on the moving web in milliseconds such that high throughput printing can be carried out [5].

In IPL photonic sintering, Xenon lamps are normally used which has typical emission spectrum of 200nm to 1200nm [5], covering entire visible spectrum with some emission lying in UV range and some in IR region. The actual wavelength of emitted spectrum depends upon the envelope quartz used in the Xenon lamp or the UV filter that may be used [6]. Most of the ink nanoparticles have absorption peak in visible region whereas the cost-effective PET substrate tends to absorb only in UV range. This is one of the major reasons for IPL photonic sintering to be able to cure the inks selectively and efficiently without damaging the underlying transparent substrate (like PET used in our case) which has tendency to absorb only in UV region [5]. Another reason is that the PET film can withstand a maximum temperature of 150°C, which might not be enough to achieve coalescence of the nanoparticle. But IPL allows thin ink layer to go beyond 1000°C for milliseconds which is enough to sinter the ink but short pulse for ms of time isn't enough to reach this heat to bottom of the substrate thus preventing from substrate damaging [7]. R2R printed patterns are in the range of few hundred nanometer to few micrometers [3] whereas the substrate used in R2R printing are generally 100-200um (Figure 1).

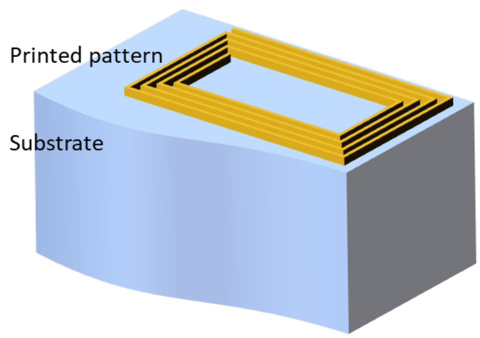

Figure 1 – Printed pattern thickness and substrate thickness comparison.

INK RHEOLOGICAL PARAMETERS

The study on the feasibility of photonic sintering in R2R printing technology was envisioned to carry on different sized near field communication (NFC) antenna patterns. In order to attain the minimum possible resistance by increasing the thickness of the antenna pattern in the R2R printing condition, a high viscosity ink should be prepared. Thus, hydrophilic silver nanoparticle-based ink with two different nanoparticle size as seen in SEM images (Figure 2) were prepared, each having a viscosity of 1000cP and surface tension of 43mN/m. Printing quality was maintained with the roll pressure of 6kgf, printing speed of 6m/min and blade angle of 9°. Shear thinning nature of the silver ink as shown in Figure 2 is also responsible for better ink transfer as the reduced viscosity of ink while printing results in improved printing.

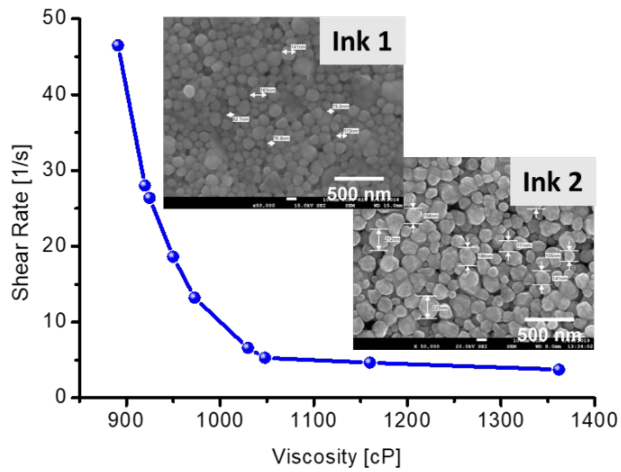

Figure 2 – Viscosity vs shear rate of silver ink to print antenna and SEM image of the silver inks.

ANTENNA DESIGN

Four antenna designs were considered, and their resistance and characterization were done before and after photonic sintering. The dimension of antenna designs is shown in Figure 3 and tabulated in the Table 1.

Antenna	Lx (mm)	Ly (mm)	W(mm)	G(mm)	N
1	83.5	47	1	0.5	7
2	89.25	46.5	1.2	0.55	4
3	60	41.4	1	0.4	5
4	49.3	41.3	1.5	0.2	5

Table 1 – Antenna dimensions.

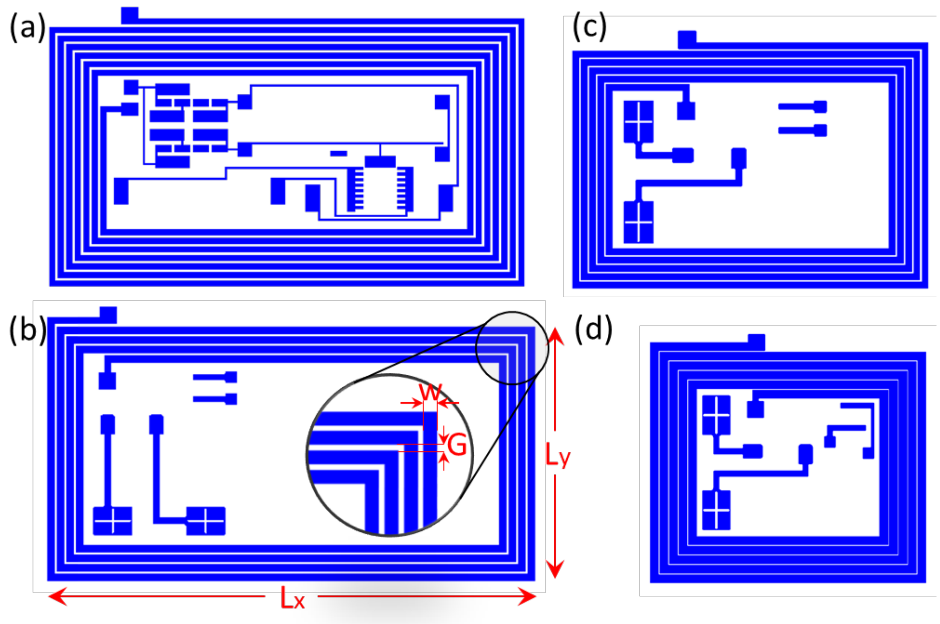

Figure 3 – Antenna designs (a) antenna 1, (b) antenna 2, (c) antenna 3, and (d) antenna 4.

THERMAL CURING

Two silver inks outlined in Figure 2, one with smaller nanoparticle size below 100nm (INK 1) and the other with slightly larger nanoparticle size above 100nm (INK 2) were used for printing. The printing was carried out on a PET web with a width of 250mm and a thickness of 100µm as shown in Figure 4a. For comparison with the photonic sintering, we first conducted the study on the effect of thermal curing under different time instants, during printing and post-printing. Since the 5s curing of the printed pattern passing through R2R heating chamber set at 150°C isn't enough to obtain antenna resistance at a desired range necessitating for post-printing thermal curing at the same temperature but for a long time. The corresponding variation in resistance of the antenna (Antenna 1 in Figure 3) utilizing both types of INKs were measured as a function of time (Figure 4: b-c). As expected, INK 1 showed relatively high resistance than INK 2 owing to its small nanoparticle size. However, as the curing time progresses, there wasn't much difference in resistance of an antenna. The reason for this is believed due to the thermal coagulation during material transfer process sintered for a long time. Furthermore, the results indicate that as long as the printing is fine, slight differences in the size of synthesized nanoparticles does not play significant role following thermal sintering for a long time. Thus, sintering technique are essential to mitigate the limitations of ink synthesis where the associated constraints such as temperature, humidity are difficult to control and thus manipulates the overall synthesis process.

However, long hours of sintering outside the R2R isn't practical from the scalability and industrial manufacturing point of view. Since the low-cost PET film cannot be cured at temperature above 150°C, further reduction of resistance of below this range is ceased as well.

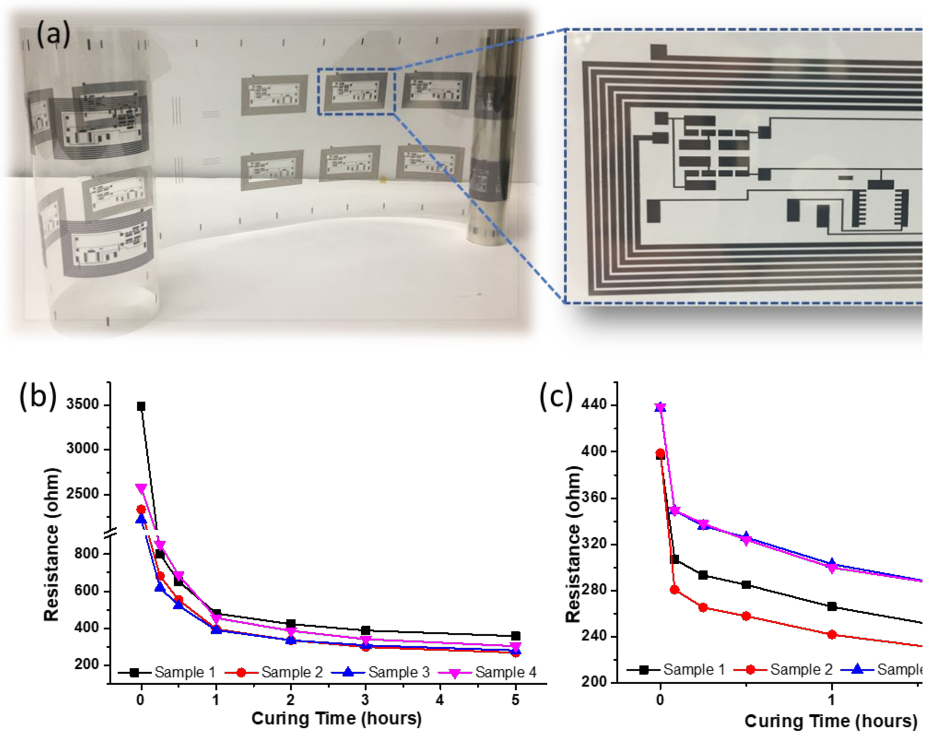

Figure 4 – (a) Image of roll to roll (R2R) gravure printed antenna. Resistance variation with extra curing time at 150°C using (b) Ink 1 and (c) Ink 2.

PHOTONIC SINTERING METHODS

Method 1

In this study, Novacentrix pulseforge 1300 (at KIMM, Korea) was used for photonic sintering. R2R printed antenna was placed at a distance of 1” from the lamp and whole antenna was sintered at once with a single flash. While flashing the light, multiple flashes of light didn’t have any improvement in reducing the antenna resistance. Therefore, only single pulse of light with various energy was applied. The lowest resistance of the antenna thus obtained was 194 Ω for Antenna 1 under a sintering condition of 300V, 4ms pulse width single pulse with 3.3J/cm^2 radiation energy. Other sintering conditions and optimizations are summarized in Table 2. The sintering condition result shows that for every pulse width, there exists the maximum voltage at which the lowest resistance of the antenna is received. Furthermore, it shows that when the pulsewidth is increased, the energy is distributed over larger pulse area and can absorb more energy resulting a low resistance.

Single Pulse IPL Photonic sintering								
V/pulse	0.1ms	0.5ms	1ms	1.5ms	2ms	3ms	4ms	5ms
250					1020 1.110 J		363 2 J	
260					512 1.240 J			
270					395 1.240 J			
280					348 1.530 J	271 2.170 J	299 2.73 J	
300		2540 0.483 J	466 0.977 J	342 1.430 J		260 2.620 J	194 3.3 J	198 3.89 J
310			403 1.080 J					
320		1340 0.568 J	354 1.180 J	277 1.735 J				
330			335 1.290 J	X 1.880 J				
340		840 0.706 J	251 1.410 J					
350	3600 0.113 J	635 0.771 J	248 1.530 J	X 2.235 J				
360		X 1.340 J	X 1.66 J					
500	3460 0.356 J							
600	550 0.632 J							
620	447 0.698 J							
650	X 0.803 J							
700	X 0.999 J							
900	ablation 1.460 J							

	Experiment not done
	Broken/ no resistance
	Shows Resistance Resistance value / Energy (J/cm2)

Table 2 – IPL sintering optimization for Novacentrix pulseforge 1300 for curing antenna 1.

Method 2

In this study, R2R compatible IPL photonic sintering equipment from semisysco (Korea) was used. Unlike Method 1, multiple pulse light is flashed over the moving web on the conveyer. The flashed pulse is stitched with overlapping area as demonstrated in the schematic (Figure 5). This IPL system is compatible for R2R system as it can synchronize with the moving web. For carrying out the IPL experiment, the conveyer was moved at 40 mm/s. The linear lamp used was 200 mm x 10 mm in size. Conveyer is moved in such a way that the 10 mm width of the sintering area is overlapped over 2mm in each pulse. The lowest resistance obtained in this method was 173 Ω for antenna1 with sintering condition of 500 V, Pulse width of 10 ms, single pulse and 3 J/cm^2 radiation energy.

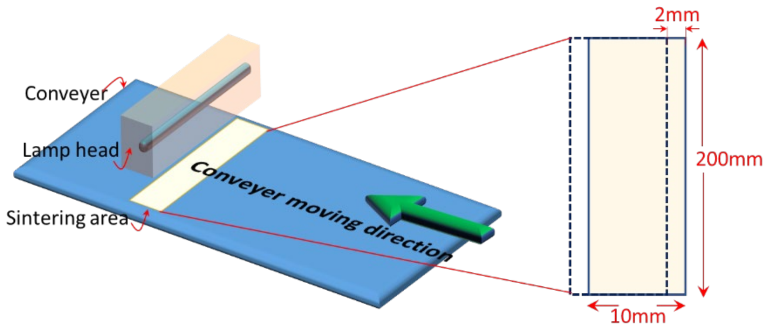

Figure 5 – Schematic representation of the R2R compatible IPL sintering system at semisysco.

Voltage	Pulse width(ms)	Pulse no	Energy(J)	Sintering resistance	
				Before	After
500	10	1	3	412	173
500	8	1	2.5	393	211
500	2	5	3	408	269
530	2	5	3.5	412	216.5
500	1	10	2	410	377
525	1	10	2.5	402	331
545	1	10	3	412	338
500	10	1	3	408	184
500	10	1	3	411	178
510	10	1	3.4	418	178
500	10	1	3.3	415	179
500	4	2.5	3	406	180
510	4	2.5	3.2	431	203
520	4	2.5	3.3	420	191

Table 3 – IPL sintering optimization for semisysco IPL sintering system for curing Antenna 1.

COMPARISON BETWEEN SINTERING METHODS

While method 1 IPL utilizes INK 1 and method 2 IPL utilizes INK 2, we did not find subtle differences in the resistance profile of the printed antenna patterns following sintering (Figure 6). However, the overall resistance in both methods were found to be less than the values obtained by thermal curing. It also proves how IPL photonic sintering system can be used for curing the printed devices like antenna in short time period being compatible for printing through R2R gravure system to realize large scale printed devices. In addition, method 2 IPL photonic sintering system being tested in a moving web in conveyer shows the future possibility of using the IPL system in conjunction to R2R printing.

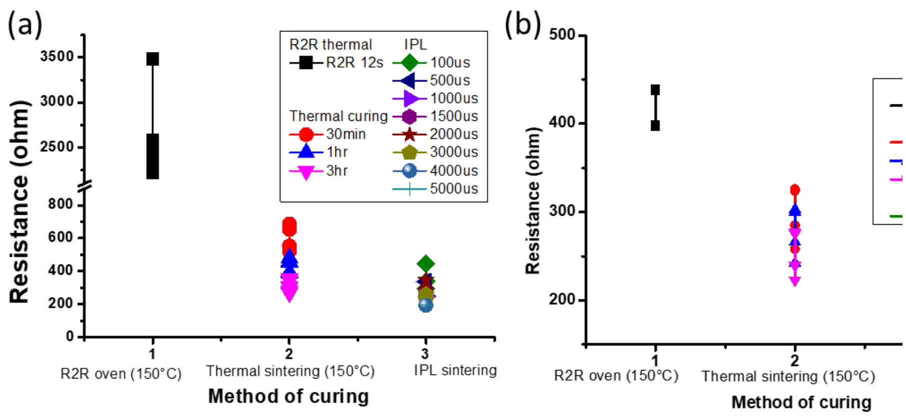

Figure 6 – Comparison of resistance of R2R printed antenna 1 after 12s curing in R2R drying chamber at 150°C with external oven curing under 150°C and IPL photonic sintering for (a) Ink 1 used in method 1 IPL and (b) Ink 2 used in method 2.

For characterizing the antenna, each of the antennas printed (antenna1) with ink1 and ink2, thermal cured and photonic sintered using two different methods were compared by measuring their Q-factor. A network analyzer (E5070B, Agilent Technologies) with test fixture was used in measuring the Q factor. Figure 7a and 9b shows how the Q factor of antenna is improved after 1 hr thermal curing at 150°C compared to only 5s of curing in R2R drying chamber while printing. However, using external oven for longer curing to get antenna performance isn't applicable in manufacturing process. Photonic sintered antennas show even higher Q factor promising to be usable in smartphone reading and proving to be viable technology to be implemented in conjunction to thermal chamber in current R2R system. In addition, Q-factor of different sized antenna was found to be more consistent following photonic sintering than in thermal curing.

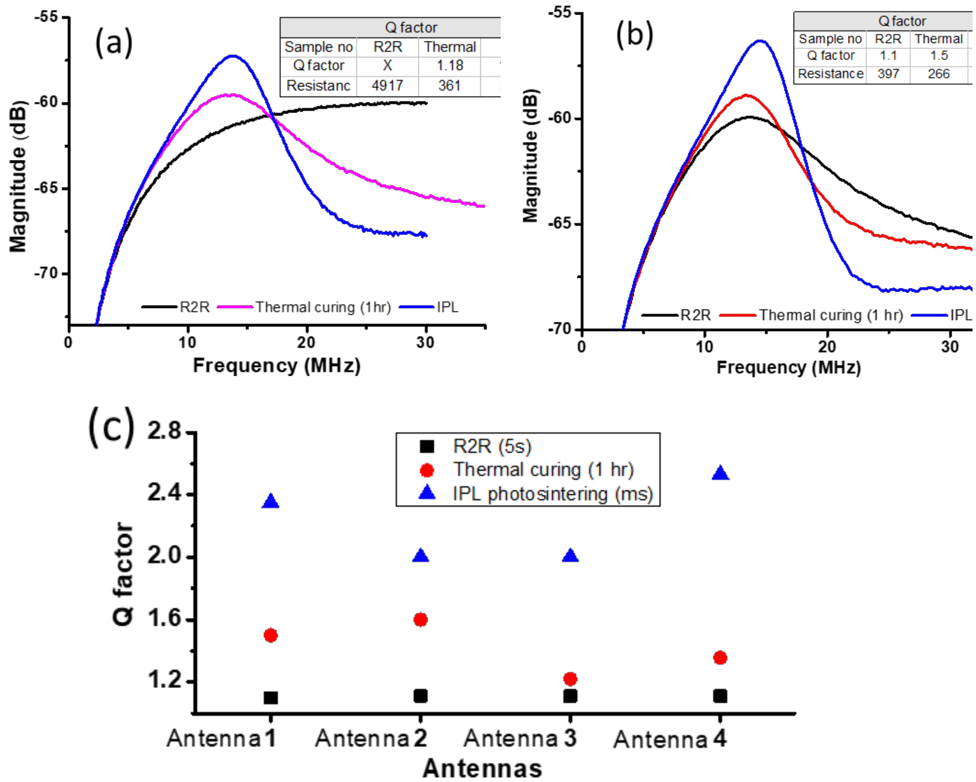

Figure 7 – Q factor characterization of (a) Method 1 IPL photonic sintering (b) Method 2 IPL photonic sintering (c) Q factor comparison of 4 antennas for all R2R 5s cured, external oven 1hr cured and IPL photonic sintering cured.

Photonic sintering challenges

While IPL photonic sintering is quite applicable from manufacturing point of view to implement in R2R system, optimizing the sintering process corresponding to the typical ink condition could be quite challenging. Figure 8 shows the optical images under various conditions showing a crack in the patterns due to higher radiation energy and peeling off from the substrate. A complete ablation of printed layer on higher radiation energy and short pulse high radiation power pulse was clearly noticed.

Figure 9 shows the completely ablated IPL photonic sintered sample and peeling off due to cohesive failure of the ink. This shows the higher radiation energy and radiation power causes such ablation and peeling off issues. This clean film ablation and peeling off of the ink layer are mainly due to high power issues which has been explained by Schroder, K.A. [7] as well.

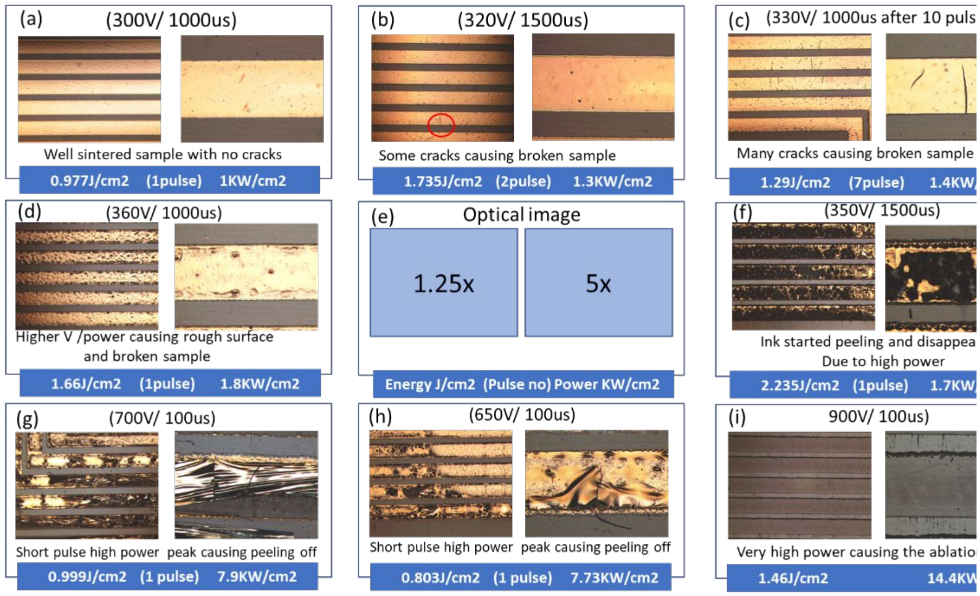

Figure 8 – Optical images of well IPL photonic sintered patterns of antenna from the well-matched photonic sintering condition (a) to the worst one (i) to show completely ablated sample with high radiant energy or high radiant power.

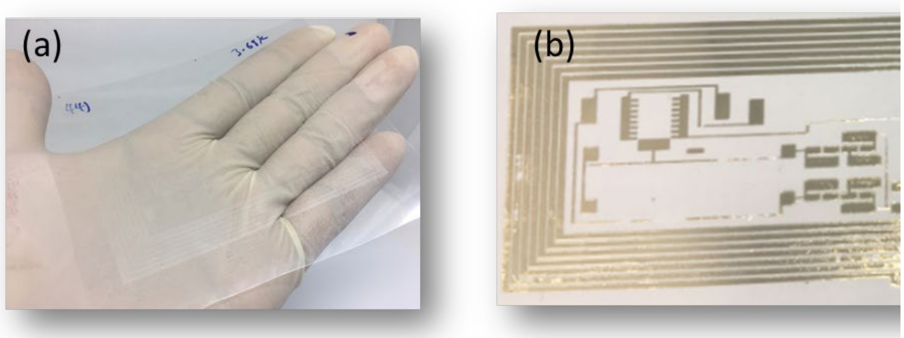

Figure 9 – (a) Completely ablated antenna pattern due to high radiant power (b) Cohesive failure of the antenna pattern causing peeling off of the antenna.

CONCLUSION AND FUTURE WORK

IPL photonic sintering has already been demonstrated in curing silver nanoparticles [8,14,16], copper nanoparticles [9,11,12,13], nickel nanoparticles [15] as well as their hybrid form [10]. However due to high initial cost for lab scale there has been limited research in this field. Moreover, most of the researches are limited to characterizing by measuring the conductivity of the photonic sintered printed patterns. But there hasn't been proper study regarding the device performance after photonic sintering. In this study, we have tried to demonstrate how the photonic curing is viable to R2R system in curing the antenna to get high conductivity and high Q factor compared to thermal curing in millisecond light pulse.

Although the antenna's performance is improved by IPL photonic sintering, its resistance can be further reduced which will result in increased reading range by smartphone as well as higher power harvesting depending on the application of designed antenna. Also, the current research in the R2R printing system where the antenna was first R2R printed with thermal curing for 5s in the R2R gravure printer and then photonic curing in separate IPL system will be the appropriate R2R system to integrate IPL system current R2R printing system.

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