Screen Printed Vias for a Flexible Energy Harvesting and Storage Module

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***Abstract*— This case study evaluates a highly flexible screen printed through-hole-via using silver microparticle inks for applications in energy harvesting and storage modules. The printed vias' fabrication and reliability are evaluated by means of a double sided screen-printing method and repetitive (cyclic) bending tests. Vias, in 125 µm thick PET, were laser cut (50, 100, 150, and 200 µm nominal diameter) then filled, and simultaneously connected to adjacent vias, by screen printing. To investigate the use of the printed via in a monolithic energy module, the vias were used for the fabrication of a flexible printed supercapacitor (aqueous electrolyte and carbon electrode).**

The results indicate that the lower viscosity silver ink (DuPont 5064H) does not fill the via as effectively as the higher viscosity ink (Asahi LS411AW), and only the sidewall of the vias are coated as the via size increases (≥ 150 µm diameter). Conversely, the Asahi silver paste fills the via more thoroughly and exhibited a 100 % yield (1010 vias; 100 µm nominal via diameter) with the 2-step direct screen-printing method. The bending test showed no signs of via specific breakdown after 30 000 cycles. The results indicate that this via filling process is likely compatible with roll-to-roll screen printing to enable multi-layered printed electronics devices.

Keywords*— *energy module, printed vias, screen printing, bending reliability, flexible and printed electronics

I. INTRODUCTION

Printed electronics (PE) have provided a means to manufacture electronic devices in a low cost and a high-volume way utilizing printing techniques such as gravure-, flexo- and screen-printing methods. The additive fabrication method can use relatively low temperatures compared to conventional silicon based electronics, and the manufactured devices can be flexible or stretchable. The low cost and high volume manufacturing becomes more significant as we reach the future Internet of Things (IoT) and Internet of Everything (IoE) trillion sensor networks.

An advantage with multi-functional PE components is the ability to print on both sides of the substrate to create multi-layered "monolithic" structures. This would not only help to downsize the required space and to create new device architectures but it will reduce overall material use. Previous studies have shown that a reliable interconnection is achieved by first filling the through-holes with dispensed ink and then printing on both sides of the substrate [1-3]. However, this requires at least three print rounds, increasing production time and cost. The approach in this study is to fill the laser cut via with ink during the double side conductive interconnect print process. With the through-hole via being done on a flexible substrate, the bending reliability was studied in conjunction to printing. Two different commercially available inks (DuPont 5064H and Asahi LS411AW) and four different laser cut via sizes were investigated (50, 100, 150, and 200 µm), which were cut into a flexible polyethylene terephthalate (PET) substrate.

Our vision is to produce an energy harvesting and storage module (i.e. polymer solar cell and supercapacitor) in a high volume roll-roll process with the interconnection between the components done using the screen-printed via (Figure 1). As an initial step, the combined structure of a flexible environmentally friendly supercapacitor [4] with the screen-printed via, using a completely sealed structure, is evaluated.

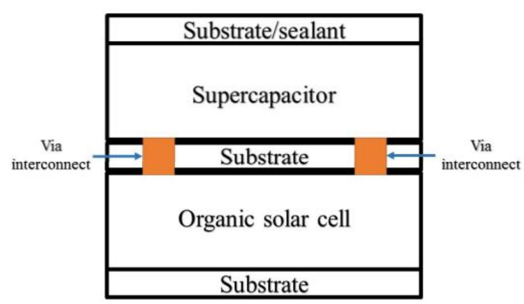

Figure 1: An energy module using screen-printed vias as the interconnection between energy storage and -harvest components.

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II. Materials and Method

A. Experimental setup

A direct screen printing approach was utilized to fill the vias with ink during the print process. Commercially available inks Asahi LS-411AW silver paste and DuPont 5064H silver ink were used as the conductive trace material. The solvent for both inks were butyl cellosolve acetate and isophorone for Asahi and C11-ketone for DuPont. The viscosities were 20 000 – 30 000 cP and 10 000 – 20 000 cP for Asahi and DuPont respectively. Both inks were printed on 125 µm thick Melinex ST506 Polyethylene Terephthalate (PET) substrate. The through-hole vias were cut with a laser beam by Ekspla Atlantic, DPL 015 3 –laser in VTT premises. The laser wavelength was 355 nm, pulse duration 9 ps, repetition range 100kHz-500kHz with 8 W maximum beam power. The laser's scanner head used a galvanometer driven laser scanning mirror, Intelli SCAN III14.

Nominal via sizes cut to the substrate with the laser beam were 50 µm, 100 µm, 150 µm and 200 µm. All substrates were put into an ultrasonic bath for 20-30 min to remove residual PET after laser cutting. After the sonication, all vias were characterized with an optical microscope.

To evaluate the reproducibility of the via filling/printing method, a daisy-chain structure was designed. Each daisy-chain structure had 22 vias in total, with seven daisy-chains placed next to each other on one sample. Along with the daisy chains, each sample had a reference line on the top side. The screens also had a sample structure, with printed current collector, to combine a supercapacitor and screen-printed via. The supercapacitor's current collector (graphite) was printed on top of a silver pad, which was connected to the screen-printed via. The screen design for both studies is shown in Figure 2.

A semi-automatic screen printer (TIC SFC 300 DE screen printer from Eickmeyer) was used for printing on the laser cut PET substrates. The squeegee's pressure, mask height, and alignment can be adjusted on the printer manually. The screen was ordered from FinnSeri Oy. (Finland) and the screen material is a super high modulus po lyester monofilament with 79 mesh count per centimeter and 68 µm mesh thickness with aluminum frames. The screen is

Table 1: A list of different samples, their sampling rate and total number of printed vias.

Via size	Ink	Samples	Number of vias printed
50µm	Asahi LS411AW	5	1010
100µm	Asahi LS411AW	5	1010
150µm	Asahi LS411AW	5	1010
200µm	Asahi LS411AW	5	1010
50µm	DuPont 5064H	8	1616
100µm	DuPont 5064H	8	1616
150µm	DuPont 5064H	7	1414
200µm	DuPont 5064H	5	1010

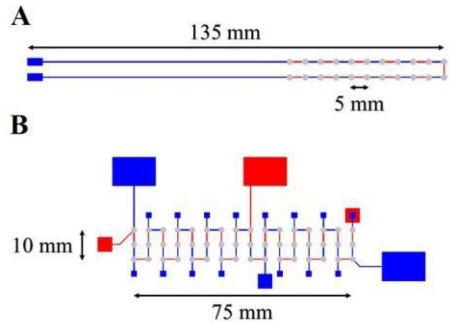


Figure 2: A: Bending structure and B combination with supercapacitor structure.

both water- and solvent resistant. First, the top side of the pattern was printed and then annealed in an oven at 150°C for 20 minutes for Asahi ink and 130°C for 20 minutes for DuPont ink. Subsequently, the bottom side of the pattern was printed and sintered in the same conditions as the top print layer.

B. Characterization

The via was first characterized with an optical microscope from the bottom and top of the substrate.

The resistance of the screen-printed through-hole vias was then electrically characterized with a multimeter to determine their connectivity. The resistance of the via was measured to ensure connection from top to bottom of the substrate. The vias were characterized as pass, poor or failed via. The categorization of via based on its resistance is shown in Table 2.

In the reliability measurements, samples were placed on a custom made pneumatically controlled bending machine. The machine has two parallel plates and the upper moving plate bends the sample that is attached. The distance between plates was 2 cm (i.e. 1 cm bending radius). The bending device and measurement setup are shown in Figure 3. The bending apparatus has been used in other scientific reliability analysis measurements [5].

An iCraft AD-converter and iPlotter software were used to convert bend cycle and resistance reading into a same chart. Resistance was calculated by feeding 4,92 V voltage across a 1 kΩ pull up resistor and the sample line. The sample line's voltage was measured and resistance was calculated with voltage division. The device collected voltage reading into a conventional computer.

All different sample types shown in table 1 were cyclic bent in the pneumatic bending machine. At least 30 000 bending cycles were done in each measurement. 50 µm and 100 µm via sizes were measured twice with a "flipped" sample to have both tensile and compressive stress onto the reference line.

Table 2: Characterization of vias based on their electrical connectivity.

Grade	Criteria
Pass via	$< 1\ \Omega$
Poor via	$1\ \Omega <$ via resistance $< 10\ \Omega$
Failed via	$> 10\ \Omega$ or no connection

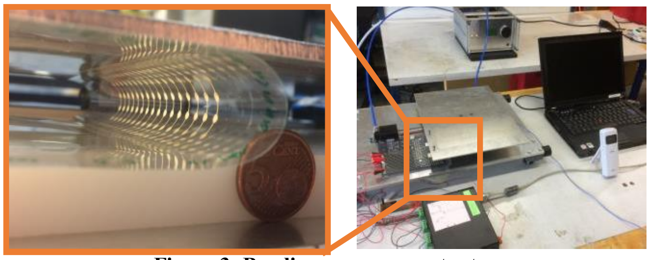


Figure 3: Bending measurement setup

III. Results

A. Optical characterization of laser cut via

Optical characterization results of the vias are shown in Table 3. As seen from the values in Table 3, the laser cut via was then cone shaped instead cylindrical. The measured via size values vary from the nominal values, which results from an offset in the laser focal point and the thickness of PET.

B. Via filling characterization results

Following via printing, cross sections of the vias were taken to evaluate the ink filling properties. The 50 µm via cross section was cut with a broad ion beam and the image was taken with a scanning electron microscope. For the larger via sizes it was possible to use ground resin moulds and an optical microscope. The cross section of after print vias are shown in Table 4.

In all cases, as seen in Table 4, the via is only partially filled during the printing process. However, a reliable electrical connection is still possible. It is also evident that the DuPont ink's filling mechanism is less effective than that of the Asahi ink. In addition, the ink fills larger via sizes poorly and mainly only coats the via sidewalls. When the ink is on the sidewalls, a less reliable connection was noticed. The unreliability is attributed to the top and bottom ink layers not forming a continuous structure inside the via as seen in Figure 5.

C. Electrical characterization of vias

Electrical conductivity of the ink was measured to ensure repeatability and adequate curing conditions. A four point probe (4PP) pattern for sheet resistance (R_S) was measured at a 10 mA current. Four patterns with 400 µm line widths were printed on the top and bottom of the PET (table 5).

As seen from Table 5, the DuPont ink has larger deviation in the sheet resistance values compared to Asahi ink. The high resistance deviation illustrates that the print process was not as optimal for the Dupont ink as for the Asahi ink. Fortunately, printing with DuPont did not affect the filling of the via, since the collar size was large enough and did not get clogged. With the Asahi ink, the R_S values are according to datasheet value, which is lower than 40 mΩ/□ for 10 µm line thickness. The DuPont inks R_S value should be less than 14 mΩ/□ for 25 µm line thickness.

Table 3: Measured via sizes.

Nominal via size (µm)	**via size average,** top (µm)	**StD** (µm)	**via size average,** bottom (µm)	**StD** (µm)	**Number of samples,** (N)
50	125	± 13	67	± 9	29
100	173	± 15	120	± 5	26
150	232	± 18	172	± 10	24
200	281	± 16	224	± 11	26

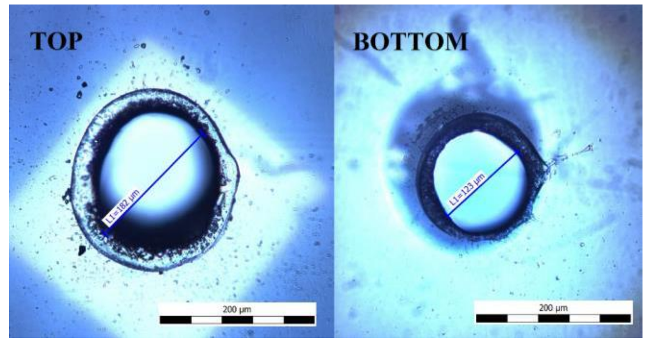
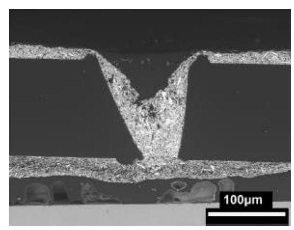
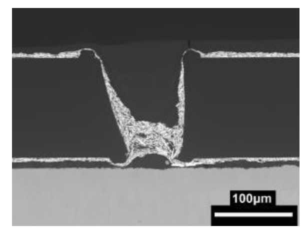
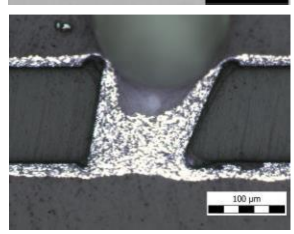
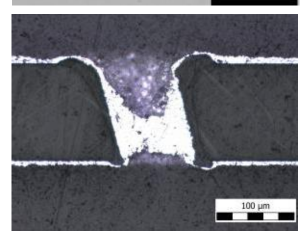
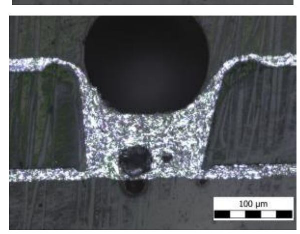
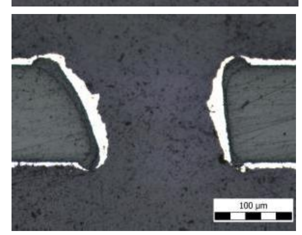
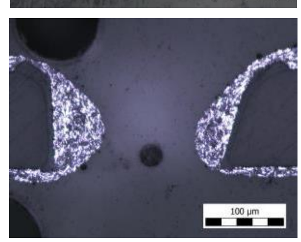
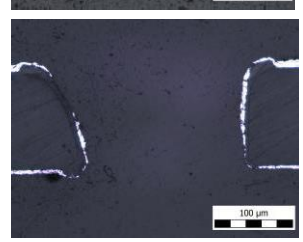


Figure 4: Example of a 100 µm nominal size via.

Table 4: Cross section of printed vias (nominal diameters 50, 100, 150 and 200 µm) using both inks (Asahi and Dupont).

Via size	**Asahi**	**DuPont**
50 µm		
100 µm		
150 µm		
200 µm		

The best ink filling results were achieved with the 100 µm nominal via diameter with both inks. With both inks, the 200 µm vias had the lowest reliability due to the reduced aspect ratio. The 50 µm nominal via size had a high number of poor vias compared to failed vias. It was noticed that with larger via sizes the ink passes the via and lays on the screen stage coating the sidewalls of the via instead of filling and creating a reliable connection. With smaller via sizes the via is filled properly and stays inside the via creating a more reliable connection. On the other hand, the smallest via sizes might limit the amount of ink penetration. The optimal via size in this study was the 100 µm via diameters. When comparing the two inks, it is believed that the Asahi fills the via more effectively, since it has higher viscosity (20 000- 30 000 cP) than DuPont (10 000 – 20 000 cP). More modelling of the ink flow and filling of the via could bring more essential information to search the most efficient via diameter and ink combination.

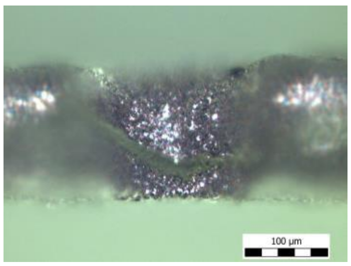


Figure 5: Cross section microscope image of an incomplete screen-printed via with failed connection (150 µm nominal via size).

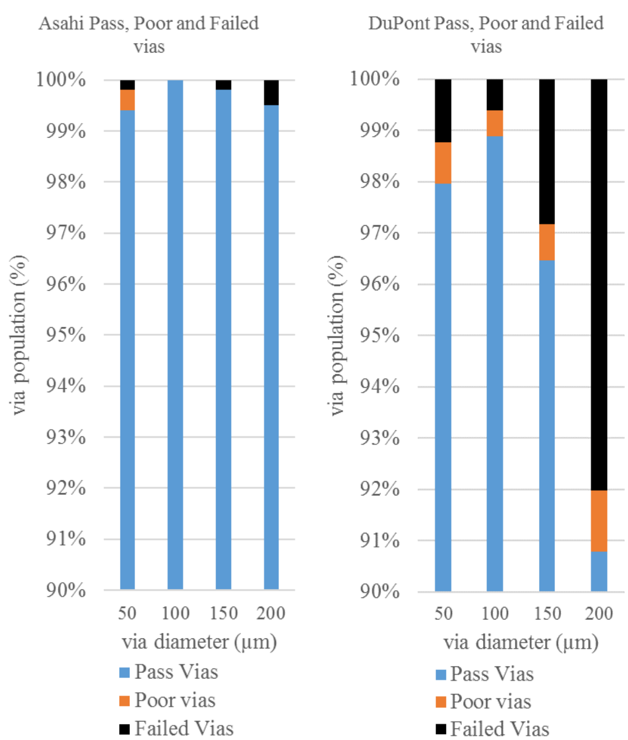


Figure 6: Electrical characterization of via results in eight cases.

Table 5: Sheet resistance (R_S) values of printed samples measured from the 4PP pattern.

Ink	**Mean R_S** Top (mΩ/□)	**Mean R_S** Bottom (mΩ/□)	**Number of 4PP patterns**
Asahi	20,3 ± 4,7	29,5 ± 6	Top: 84 Bottom: 84
DuPont	31,4 ± 22,6	26,67 ± 14,2	Top: 96 Bottom: 132

D. Via bending results

The reliability of the vias was investigated with the cyclic bending study. The results herein combine an average of the results from three bent daisy-chain structures. An average from the reference samples was also taken from tensile and compressive stressed samples and added to the graphs. The average reference values were taken from four compressive stressed samples and two tensile stressed samples for both inks. Both tensile and compressive stressed reference samples are in the chart to help differentiate the via bending effect from the interconnection stress. The daisy-chain samples' resistance change (ΔR) caused from cyclic bending is shown in Figure 7 and Figure 8 for Asahi and DuPont inks respectively.

The daisy-chain structure faces both compressive and tensile stress and thus is between the reference lines' ΔR. The sample lines do still have different ΔR when comparing different via sizes, which is caused by the via bending. An estimation from individual via bending can be given by comparing the daisy-chain structure bending to the fitted compressive and tensile stressed reference average and dividing the results by the number of vias in the daisy-chain structure. The cyclic bending effect estimation to each via size and ink is shown in Figure 9.

Figure 9 illustrates that the largest via sizes are the most resilient to bending with Asahi samples. With DuPont samples on the other hand, 100 µm samples are more resilient to bending compared to 150 µm samples. This is because of high starting resistance value of 150 µm sample seen in Figure 8 caption. Smaller via sizes were thought to have more bending strain to the collar of the via, since the collar is smaller and larger strain is focused on it. Larger via sizes have larger collars respectively, which is beneficial in terms of the bending strains this spreads the bending strain and causes smaller resistance change. In other scientific papers the stress hotspots gather around to the top of the via, since the ink or conductive adhesive, which is used to fill the via, is thicker than the conductive trace [3, 6]. Thicker traces face stronger bending strain [7].

An acceptable resistance change depends on multiple factors such as the application of use. For example, other scientific publications have set the failure criterion to 20 % resistance change, which was achieved after 1555 bending cycles [8]. The failure criterion corresponds to the results that were gathered in this study. The measurement was done for at least 30 000 bending cycles to clearly separate different sample variables from each other.

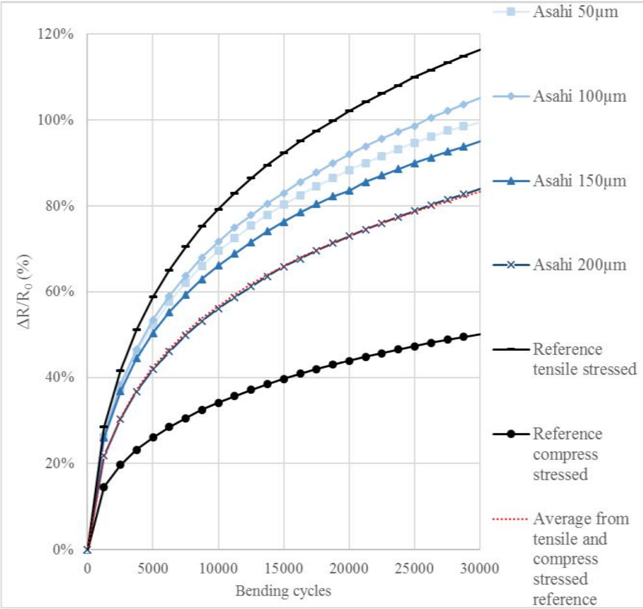


Figure 7: Asahi samples bending results. Initial resistance (R_0) for each case: 50 µm, 13.4 Ω; 100 µm, 12.8 Ω; 150 µm, 11.9 Ω; 200 µm, 11.5 Ω; Reference tensile, 10.7 Ω; Reference compress 10.5 Ω.

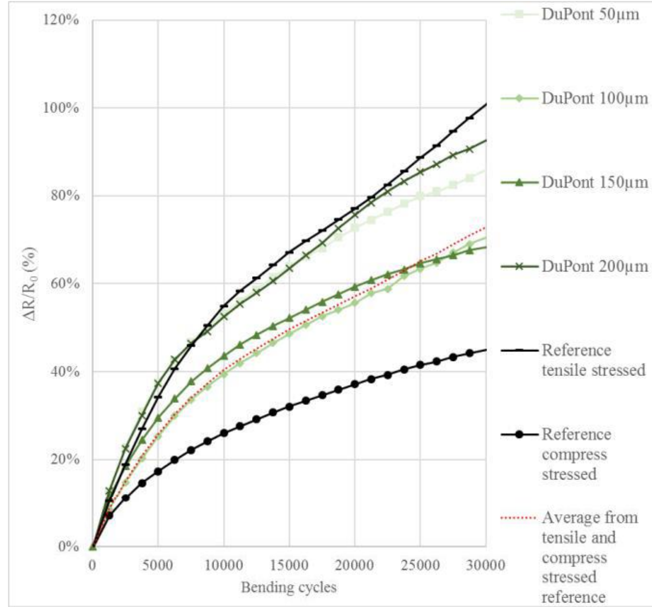


Figure 8: DuPont samples bending results. Initial resistance (R_0) for each case: 50 µm, 16.9 Ω; 100 µm, 17.6 Ω; 150 µm, 20.0 Ω; 200 µm, 12.3 Ω; Reference tensile, 9.9 Ω; Reference compress 15.5 Ω.

E. Optical characterization of bent vias

Scanning electron microscope (SEM) images were taken from the bent vias and compared to a non-bent reference sample. The images were taken on top of the via from the tensile stressed side. The results are shown in Figure 10.

Increased dissociation of silver microparticles (flakes) around the via collar is caused during bending by the deformation of the PET substrate. The local stretching strain of the collar ink relative to the perpendicularly oriented via induces microcraks around the via cavity. This in turn leads to increased resistance of the via seen in Figure 9. This observed effect is corroborated by the theoretical modelling performed by Petér *et. al.* [3].

F. Combination of screen printed via and supercapacitor

A supercapacitor was combined with the printed screen-printed through-hole via as a proof of principle. First, a silver pad was printed on the PET substrate, subsequently, to the other side of the substrate. The supercapacitor's electrode was printed then on the silver pad. The supercapacitor electrical properties were measured with a Maccor (USA) battery testing device, which charged and discharged the supercapacitor. The supercapacitor's capacitance, equivalent series resistance (ESR) and current leakage was calculated from the measurement data. Supercapacitor with vias and a reference supercapacitor structure are shown in Figure 12.

It was seen from the measurement results that the reference supercapacitor and supercapacitor combined with the screen-printed via does not vary greatly. ESR was slightly higher in average, since the printed line increases resistance to the supercapacitor. A completely sealed supercapacitor structure can be built with the screen printed via using R2R printing methods, which is illustrated in Figure 11.

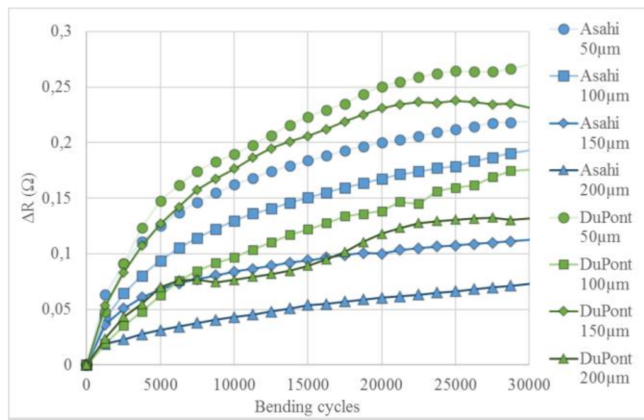


Figure 9: Estimation of the via bending effect in all eight cases.

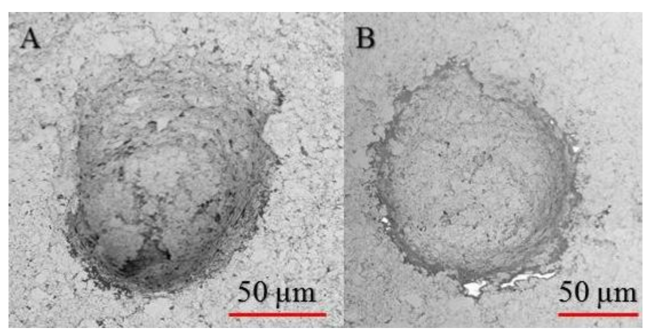


Figure 10: SEM images of (A) before bent via and (B) after bent via. (100 µm nominal via size)

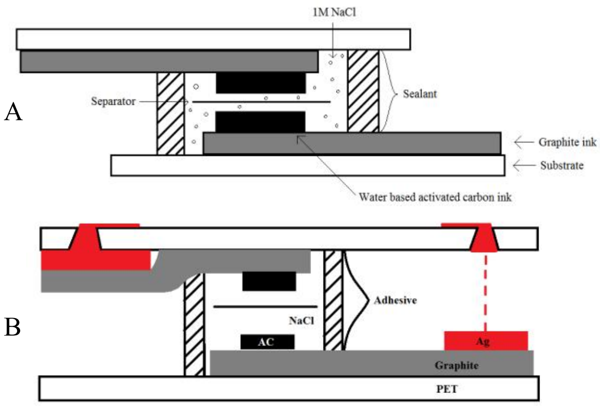


Figure 11: (A) Supercapacitor reference structure and (B) supercapacitor all sealed structure using vias illustrations.

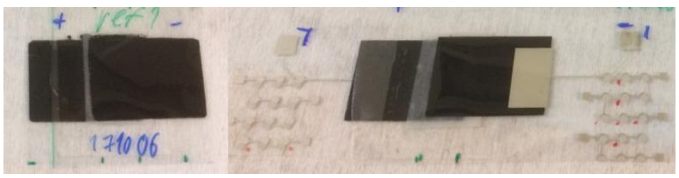


Figure 12: Reference supercapacitor on the left. Supercapacitor with vias combined structure on the right.

IV. Conclusion

The screen-printed through-hole via filling was evaluated with two different silver inks and four different via diameters. The reliability of interconnection between top and bottom print was evaluated with a cyclic bending test. The screen-printed through-hole-via was filled during the screen-printing process. The more viscose Asahi LS411AW paste performed better in the printing process compared to the DuPont 5064H ink. The Asahi ink's yield of fully functional vias was 99.67 % and Dupont's was 95.57 %. Four different laser cut via sizes: 50 µm, 100 µm, 150 µm and 200 µm were compared to each other, where 100 µm had the best yield with both used inks.

In the bending analysis of the screen-printed via, the printed sample structure with vias was attached into a pneumatic bending machine and the resistance of the daisy-chain sample structure line was measured while being cyclic bent 30 000 times. The resistance of a printed line rose in every cyclic bending measurement. However, a via breakdown between the top and bottom side prints was not found in the bending tests. Therefore, bending the vias has little effect on the overall resistance change in the sample line. However, there seems to be an optimal aspect ratio of vias for filling and bending reliability depending on ink properties.

Furthermore, a supercapacitor and a screen-printed through-hole via combined structure was fabricated as an initial step to enable energy module fabrication. The results showed that the screen-printed vias and printed interconnects can be used together with the supercapacitor. In the future the screen-printed vias, with the direct printing process, can be optimized for a R2R process to print PE applications, such as fully printed energy modules, in a low cost and high volume print fabrication line.

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